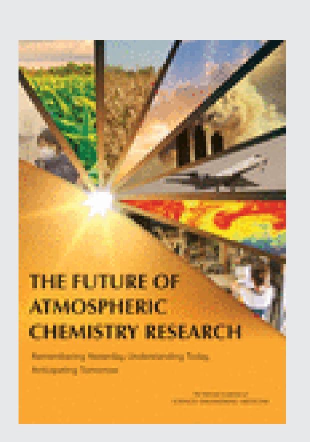

The Future of Atmospheric Chemistry Research: Remembering Yesterday, Understanding Today, Anticipating Tomorrow

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Committee on the Future of Atmospheric Chemistry Research; Board on Atmospheric Sciences and Climate; Division on Earth and Life Studies; National Academies of Sciences, Engineering, and Medicine

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This report has been reviewed in draft form by individuals chosen for their diverse perspectives and technical expertise. The purpose of this independent review is to provide candid and critical comments that will assist the institution in making its published report as sound as possible and to ensure that the report meets institutional standards for objectivity, evidence, and responsiveness to the study charge. The review comments and draft manuscript remain confidential to protect the integrity of the process. We wish to thank the following individuals for their review of this report:

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Although the reviewers listed above have provided many constructive comments and suggestions, they were not asked to endorse the report's conclusions or recommendations, nor did they see the final draft of the report before the release. The review of this report was overseen by Robert F. Sawyer, University of California, Berkeley, and Gregory R. Carmichael, University of Iowa, Iowa City. They were responsible for making certain that an independent examination of this report was carried out in accordance with institutional procedures and that all review comments were carefully considered. Responsibility for the final content of this report rests entirely with the authoring committee and the institution.

Preface

The chemistry of the atmosphere envelops us, affecting the health of humans and our environment, including croplands, forests, grasslands, and oceans. It converts emissions by humans and nature into gases and particles that obscure visibility, acidify rain, harm human health, and influence weather and climate. As human society continues to develop and populations grow, requirements for energy, food, and water also expand. This growth in turn drives changing distributions of chemical emissions from a variety of sources, with increases in anthropogenic air pollution in developing countries and decreases in developed countries, as well as alterations in natural sources caused by changing land use. Emissions are not just a local issue, as hemispheric and global transport of these chemicals and their reaction products have led to substantial changes in the global Earth system. Clearly, fundamental understanding of the chemistry of the atmosphere today and the ability to predict how it will change in response to human activities are integral components of developing policy to ensure societal well being.

It has been more than thirty years since the field of atmospheric chemistry has evaluated its research accomplishments and future directions. In 1984, at the request of the National Science Foundation (NSF), the National Academies of Sciences, Engineering, and Medicine (the Academies[1]) developed the report *Global Tropospheric Chemistry: A Plan for Action*. That report recognized the impact that humans were having on the global atmosphere and called for a comprehensive research program to investigate the chemistry of the lower atmosphere on broad scales. Prior to that report, the emphasis in atmospheric chemistry had been on understanding the polluted troposphere and perturbed stratosphere. The 1984 report represented a paradigm shift—it had become clear that in order to understand human influences on the atmosphere, the scientific community also needed to understand the unperturbed system upon which these influences were imposed. This conclusion led directly to the formation of the NSF Global Tropospheric Chemistry Program, which not only resulted in enhanced support for atmospheric chemistry research, but also has been a major factor in the development of the discipline of atmospheric chemistry over the intervening decades. That report also helped provide a foundation for the formation of the International Global Atmospheric Chemistry (IGAC) program, a highly successful and far-reaching international research effort that is still active today.

Since that time, there have been a handful of specialized reports on atmospheric chemistry published by the Academies—*Rethinking the Ozone Problem in Urban and Regional Air Pollution* (1991), *A Plan for a Research Program on Aerosol Radiative Forcing and Climate Change* (1996), *Global Air Quality* (2001), *Radiative Forcing of Climate Change* (2005), and *Global Sources of Local Pollution* (2010). Atmospheric chemistry was also included in two reports focusing on the future of the atmospheric sciences, including *The Atmospheric Sciences: Entering the Twenty First Century* (1998) and *Strategic Guidance for the National Science Foundation's Support of the Atmospheric Sciences* (2007). None of these reports, however, examined the field of atmospheric chemistry as a whole. Recognizing that there have been tremendous changes in our understanding of the chemistry of the atmosphere and our ability to investigate it through field, theory, laboratory, and modeling efforts over the past three decades, NSF once again asked the Academies to develop a report that would address the rationale and

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[1] In July 2015, the Academies formally changed its name and is no longer known as the National Research Council.

need for a comprehensive and broadly based research program in atmospheric chemistry over the next decade and identify priority areas of research and associated infrastructure that would be needed to successfully accomplish this research. The Committee on the Future of Atmospheric Chemistry Research was formed in early 2015 to address these issues.

A central part of the Committee's activities was seeking the thoughts and advice of the U.S. atmospheric chemistry community on future priority areas in atmospheric chemistry research. This input was solicited by the Committee during a series of "town hall" meetings during the spring and summer of 2015 in Boulder, CO; Cambridge, MA; Washington, DC; Irvine, CA; and Atlanta, GA; as well as informally at the Gordon Research Conference on Atmospheric Chemistry in Waterville, NH. More than 250 individuals participated in these town hall meetings and provided valuable insights and suggestions to the Committee. In addition, almost 50 people expressed their thoughts and concerns about future atmospheric chemistry research to the Committee through an online portal. The Committee then considered the hundreds of collected comments that formed the basis for our analysis. During the preparation of this report the Committee held six meetings: in Washington, DC (three times), Irvine, CA (twice), and Atlanta, GA. A number of conference calls and WebEx meetings were also held during this time.

The Committee was also fortunate to have a number of distinguished atmospheric chemists speak formally to us during several of the meetings. These included atmospheric chemists from the international community, including Len Barrie (Sweden), Guy Brasseur (Germany), Peter Liss (United Kingdom), and Megan Melamed (US, IGAC). We also had the benefit of presentations by Michael Kleinman (UC Irvine) and John Seinfeld (Cal Tech). Mel Briscoe presented invaluable information and guidance to the Committee on prioritization processes. Presentations were made by program managers from several federal agencies, and these agencies also provided us with funding data for atmospheric chemistry over roughly the past decade. We greatly appreciate the efforts of Sylvia Edgerton and Peter Milne (NSF); Jack Kaye and Hal Maring (NASA); Sherri Hunt (EPA); Ashley Williamson, Shaima Nasiri, Sally McFarlane, and Dorothy Koch (DOE); and David Fahey, Steve Fine, Elliot Forest, Jason Donaldson, Laura Letson, Kenneth Mooney, and Monica Kopacz (NOAA) in providing us with this information.

It is clear that the field has advanced a great deal since the 1984 report. First, leaps in fundamental understanding of atmospheric processes have not only affected the field of atmospheric chemistry, but have also led to identification of new processes in the foundational fields of chemistry, physics, and meteorology, advancing those fields as well. Second, technological developments for measurements of chemicals in the atmosphere have led to substantially more complete characterization of the full range of atmospheric chemical composition and chemistry. Third, atmospheric chemistry understanding has advanced sufficiently that it is now clear that the field is central to addressing vitally important societal issues such as the health of humans and ecosystems, climate, and weather. Addressing these issues depends on a deep understanding of atmospheric chemistry as the basis for the development of reliable predictive capabilities across local, regional, and global scales. We believe this perspective represents a significant evolution of the field similar to that captured by the 1984 report.

In words attributed to Isaac Newton, "If I have seen further, it is by standing on the shoulders of Giants." In this spirit, the report traces some of the history and tremendous successes of the field of atmospheric chemistry and highlights a few of the scientists who contributed with major breakthroughs, hence “Remembering Yesterday.” The report addresses the current state of understanding in various areas of atmospheric chemistry (“Understanding Today”), and finally, important challenges for the future that our community faces (“Anticipating Tomorrow”).

The Committee could not have completed its task without the tremendous support of many people at the Academies. This report began with expert assistance and guidance from Katie Thomas at the beginning stages. Edward Dunlea, senior program officer at the Academies, was a tower of strength, knowledge, patience, and wisdom during the entire process of the development of this report. His ability to nudge us all forward constructively but consistently and his contributions and interactions on an almost 24/7 basis were major factors in the report’s successful completion; we could not have asked for a better leader from the Academies. Kristina Pistone and Heather Coleman, both Christine Mirzayan Science and Technology Policy Graduate Fellows at the Academies, provided essential behind-the-scenes work tracking down people, references, and myriad other data for our report, in addition to helping significantly during its writing. Deb Glickson and Laurie Geller provided wonderful perspectives, experience and advice as we began our prioritization process. The Academies’ staff Shelly Freeland and Michael Hudson provided invaluable support that greatly facilitated the logistics of our meetings so that the Committee could focus on the tasks at hand. Their cheerful, efficient, and professional assistance set a very high standard indeed.

Finally, we cannot thank enough the members of this Committee, who have made our jobs as co-chairs a pleasure. They have worked diligently since before our first meeting and throughout the process of report development. Every member has made seminal contributions to the report, and they have done it with a deep understanding of atmospheric chemistry, past and present, and what is needed in the future. They have been very patient with us as we tried to find the best paths forward that considered all points of view. Their sense of responsibility as well as humor has made deadlines and difficult choices achievable.

If the membership of this Committee and those in the community who participated in the process are representative of the current field of atmospheric chemistry, and we think they are, our discipline is in very good hands for the future.

Robert A. Duce, Committee Co-Chair
Barbara J. Finlayson-Pitts, Committee Co-Chair

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Summary

CHANGING WORLD, CHANGING ATMOSPHERE

Our world is changing at an accelerating rate. The global human population has grown from 6.1 billion to 7.1 billion in the last 15 years and is projected to reach 11.2 billion by the end of the century. The distribution of humans across the globe has also shifted, with more than 50 percent of the global population now living in urban areas, compared to 29 percent in 1950. Along with these trends, increasing energy demands, expanding industrial activities, and intensification of agricultural activities worldwide have in turn led to changes in emissions that have altered the composition of the atmosphere.

These changes have led to major challenges for society, including deleterious impacts on climate, human and ecosystem health. Climate change is one of the greatest environmental challenges facing society today. Earth’s average surface temperature has already increased by more than 1.4°F (0.8°C) over the past 100 years and is expected to continue to increase, leading to major impacts on sea level, Arctic sea ice, and precipitation patterns. Air pollution is a major threat to human health, as one out of eight deaths globally is caused by air pollution. Future food production and global food security are vulnerable to both global change and air pollution. Atmospheric chemistry research (see Box S.1) is a key part of understanding and responding to these challenges.

ATTAINING A PREDICTIVE CAPABILITY

Atmospheric chemistry research has supported policy decisions that have greatly improved human health and welfare. For example, research findings guided policies to reduce poor air quality in urban areas (“smog”), acid deposition (“acid rain”), and stratospheric ozone depletion (including the Antarctic “ozone hole”). These historical examples share some common elements. Knowledge drawn from laboratory experiments, theory, field measurements, and models provided a fundamental understanding of atmospheric behavior. When rapid environmental change was observed, additional and targeted research helped elucidate the physical processes and human activities driving these changes. A predictive capability was then developed that enabled evaluation of future scenarios of environmental and societal impacts that could inform policy choices (see Figure S.1). Continuing research refines the predictive capability and provides a context for interpreting observations from ongoing atmospheric monitoring and assessment efforts. The goal of atmospheric chemistry research is to anticipate and prepare for future environmental challenges with an enhanced predictive capability that foresees environmental changes and societal impacts, rather than just react to them after they occur.

THIS STUDY

The last comprehensive report to examine the field of atmospheric chemistry as a whole (*Global Tropospheric Chemistry: A Plan for Action* [1984]) was published more than 30 years ago.

BOX S.1
The Field of Atmospheric Chemistry

In the words of Walt Whitman, the atmosphere is the “common air that bathes the globe.” Indeed, the atmosphere serves as the planet’s great integrator—mixing, transforming, and transporting both natural and human-caused emissions around the globe. The atmosphere delivers elements necessary to sustain life, but also pollutants that harm humans and ecosystems. Atmospheric constituents alter the structure of clouds and heat balance, changing the Earth’s climate, weather patterns, and hydrological cycle.

The field of atmospheric chemistry is focused on describing the chemical composition of the atmosphere, understanding sources of gases and particles, quantifying the fundamental chemical transformations that happen within the atmosphere, and applying that knowledge to explore how the composition of air responds to changing human and natural inputs. Atmospheric chemistry research leads to new discoveries about the nature of the air we breathe, elucidates changes occurring to the planet, and provides a crucial link to understanding the Earth system.

In 2015, the National Science Foundation (NSF) requested that the National Academies of Sciences, Engineering, and Medicine undertake a study to identify priorities and strategic steps forward for atmospheric chemistry research for the next decade. The Academies formed the Committee on the Future of Atmospheric Chemistry Research (referred to as the Committee) to summarize the rationale and need for supporting a comprehensive U.S. research program in atmospheric chemistry; comment on the broad trends in laboratory, field, satellite, and modeling studies of atmospheric chemistry; determine the priority areas of research for advancing the basic science of atmospheric chemistry; and identify the highest priority needs for improvements in the research infrastructure to address those priority research topics (see Box 1.2 for the full statement of task for this report). The report also describes the scientific advances over the past decade in six core areas of atmospheric chemistry: emissions, chemical transformation, oxidants, atmospheric dynamics and circulation, aerosol particles and clouds, and biogeochemical cycles and deposition. As shown in Figure S.1, these topics define core components of the scientific discipline of atmospheric chemistry. The report was developed for the NSF’s Atmospheric Chemistry Program; however, the results presented will be of interest to other agencies and programs that support atmospheric chemistry research.

RECOMMENDED SCIENTIFIC PRIORITIES FOR ATMOSPHERIC CHEMISTRY RESEARCH

A central part of the Committee’s activities was to seek advice from the U.S. atmospheric chemistry community. Community input was solicited during a series of “town hall” meetings and through an online “virtual town hall” website (see Appendix B). Using this community input and a survey of today’s research as the underlying basis, the Committee ultimately chose five Priority Science Areas that it believes will drive atmospheric chemistry research over the next decade. The first two Priority Science Areas are necessary for building the foundation of atmospheric chemistry, aimed at providing further growth in understanding of how the atmosphere works. The next three Priority Science Areas directly address major challenges facing society, for which advances in atmospheric chemistry are required to make progress. In total, these Priority Science Areas cover a broad range of research questions. Specific areas of focus are provided to address key science gaps within each area, along with examples of actions needed to address those gaps (in Chapter 5).

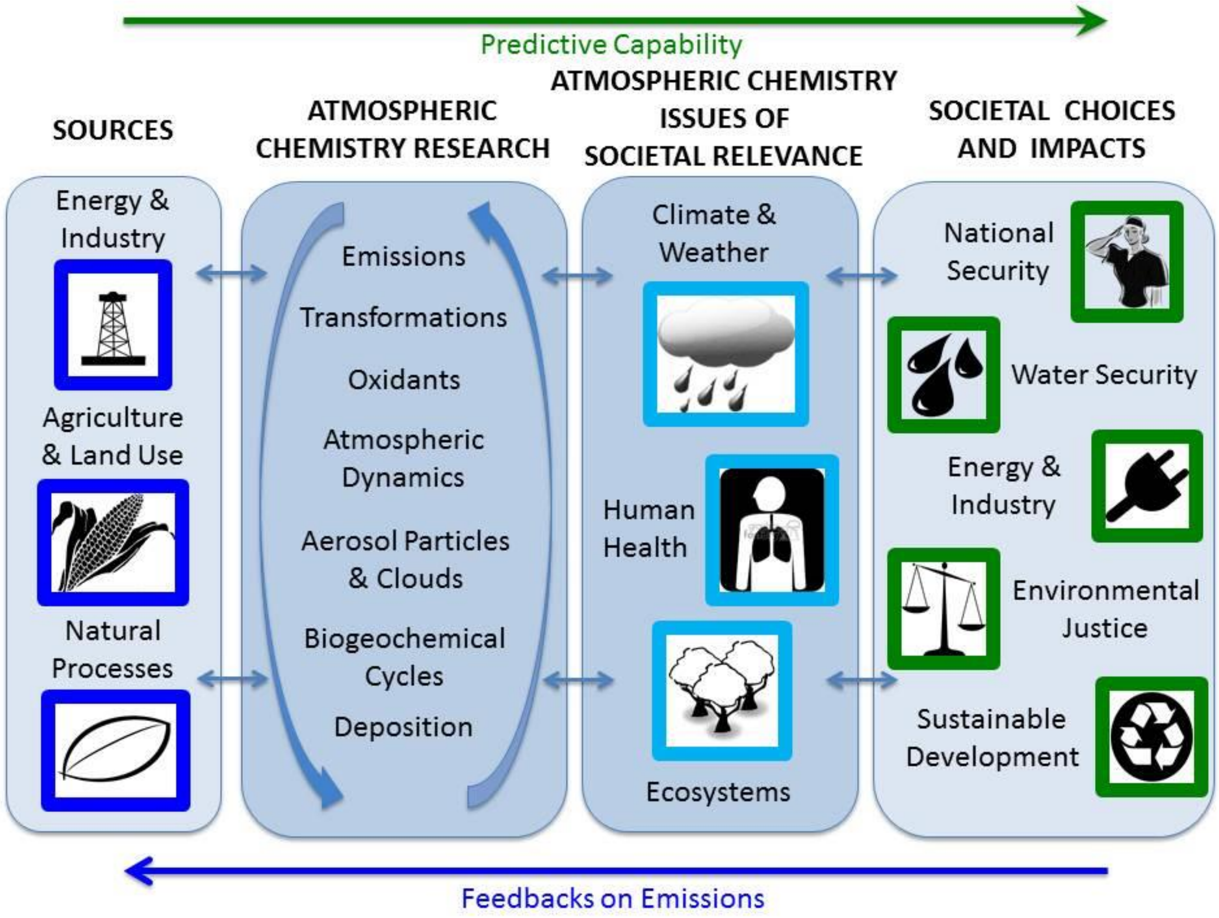

FIGURE S.1 Schematic showing the connections among major emission sources to the atmosphere, atmospheric chemistry research, scientific issues of societal relevance, and subsequent societal choices and impact areas. Research in atmospheric chemistry helps to build a predictive capability for future environmental conditions that can indicate likely impacts to society of various policy choices.

__Priority Science Area 1__: Advance the fundamental atmospheric chemistry knowledge that enables predictive capability for the distribution, reactions, and lifetimes of gases and particles.

Predictive capability starts with a fundamental understanding of the atmospheric chemistry occurring now. While some predictions can be made with confidence using the current understanding, important gaps and inconsistencies remain. Advances in atmospheric chemistry experiments, theory, modeling, and observational capabilities enable atmospheric chemists to identify and begin to narrow those gaps and to resolve discrepancies for today’s atmospheric composition and chemical reactions.

Actions needed to address key scientific gaps include:

A. Quantify reaction rates and understand detailed chemical mechanisms in multi-pollutant and multi-phase environments that cover the chemical and dynamical regimes from polluted urban to natural remote regions.
B. Identify and quantify important atmospheric oxidants or other reactants that lead to transformation and removal of chemical species from the atmosphere across broad spatial and temporal scales.
C. Develop a stronger understanding of the influences that heterogeneous chemistry exerts on tropospheric composition.
D. Understand and quantify the influence of the coupling between chemical and meteorological processes on the distribution of trace constituents in the troposphere.
E. Understand and quantify the influence of the coupling between chemical, dynamical, and radiative processes involving stratospheric chemistry.

***Priority Science Area 2*: Quantify emissions and deposition of gases and particles in a changing Earth system.**

Emission and deposition processes govern concentrations and spatial distributions of gases and particles in the atmosphere. A predictive capability of these distributions is key for assessing the impacts of atmospheric processes on human and ecosystem health, weather, and climate. Research is needed to reduce uncertainties in emissions for known sources and constrain emissions of poorly understood constituents (e.g., bioparticles), as well as to understand deposition processes that remove reactive species. Sources of atmospheric constituents change as humans make new decisions about technology, energy systems, pollution control, agriculture, and transportation. Natural sources respond to meteorological conditions, changes in land use, longer-term changes in climate, and biogechemical and ecosystem feedbacks.

Actions needed to address these key scientific gaps include:

A. Better determine emissions from both anthropogenic and natural sources and their spatial and temporal variations and trends.
B. Identify mechanisms and measure rates by which wet and dry deposition removes aerosol particles and trace gases from the atmosphere.
C. Determine the role of meteorology, including temperature, precipitation, and extreme events, on emissions and removal of atmospheric species.
D. Determine the role of global change and societal choices (including changes in climate, energy choices, and land use) on the emissions and removal of atmospheric species.

***Priority Science Area 3:* Advance the integration of atmospheric chemistry within weather and climate models to improve forecasting in a changing Earth system.**

Greenhouse gases and atmospheric particles impact the Earth's radiation budget and dynamics of the atmosphere and thereby alter weather, for example, via changing precipitation patterns and monsoon circulations. Aerosol particles play a critical role through their influence on the growth, formation, and development of clouds and precipitation. In global climate models, the effects of atmospheric aerosol particle concentrations on radiation and the distribution and radiative properties of the Earth's clouds is the most uncertain component of overall global radiative forcing. Changes in atmospheric dynamics and circulation that are linked to changes in atmospheric composition (e.g., precipitation patterns, monsoon circulations) are even more uncertain than radiative forcing. During the past decade of intensive aerosol-cloud-climate research, some scientific gaps have been closed, and additional processes have been identified that still elude quantification. As with many complex systems in intermediate stages of understanding, this progress has not yet reduced the overall magnitude of uncertainty, leaving major deficiencies in the ability to project future climate.

The chemical reactions involving aerosol particles and gases determine not only particle formation but also the processes by which climate-relevant trace species are removed from the atmosphere. Atmospheric chemistry, therefore, remains the crucial component that allows estimates of the atmospheric lifetimes of many species, and consequently the ability of pollution to accumulate in the atmosphere and thus influence climate and weather. Some, but not all, studies of regional climate change (seasonal-to-interannual or longer-term projections) or weather forecasting have included heterogeneously distributed aerosol particles or ozone in their models. The atmospheric chemistry community needs to continue to work with the climate and weather research communities in several major areas so that knowledge of the many roles that atmospheric composition plays in climate and weather can be built into dynamical models.

The atmospheric chemistry community should expand interactions with the climate and weather research community to address these key scientific gaps:

A. Determine the global distributions and variability of atmospheric trace gases and aerosol particles, and better understand their climate-relevant properties.
B. Understand the role of aerosol particles as a modulator of cloud microphysics and precipitation efficiency in natural and anthropogenically perturbed environments.
C. Develop accurate descriptions of the complex chemical and physical evolution of atmospheric constituents that can be implemented in models for robust prediction of the impact of the chemical state of the atmosphere on climate and weather.

***Priority Science Area 4*: Understand the sources and atmospheric processes controlling the species most deleterious to human health.**

Atmospheric gases and particles have documented effects on multiple adverse health outcomes, including chronic and acute effects that can lead to mortality and different types of morbidity. It is estimated that air pollution is responsible for 1 out of 8 premature deaths (more than 7 million annually) worldwide. However, the specific chemical species that cause these various effects and potential synergisms among them are not well understood. Advanced atmospheric chemistry research techniques (e.g., models, analytical methods, and instrumentation) are necessary to understand the identities, sources, and fates of the air pollutants that negatively affect human health.

The atmospheric chemistry community should expand interactions with the exposure, epidemiology, and toxicology research communities to address these key scientific gaps:

A. Develop mechanistic understanding to predict the composition and transformations of atmospheric trace species that contribute to impacts on human health.
B. Quantify the distribution of atmospheric constituents that impact human health.

C. Determine what unique sources and chemical reactions occur in indoor environments that have implications for atmospheric chemistry and human health.

***Priority Science Area 5*: Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.**

Biogeochemical cycles control the elements that are necessary for life and connect chemistry in the atmosphere with oceans, the solid earth, and the terrestrial and marine biospheres. This exchange of compounds is tightly coupled to global food security (e.g., agriculture, fisheries) and certain energy sources (e.g., biofuels, wood). These exchange processes are influenced by human activity and global climate and are directly tied to natural and managed ecosystem health. In addition, biogeochemical cycles and ecosystem health play a central role in climate by regulating carbon uptake by the biosphere and the exchange of greenhouse gases and aerosol particle precursors. Finally, the biogeochemical cycling of toxic constituents (e.g., mercury) directly affects ecosystems as well as human health.

Major scientific goals include understanding the cycling of elements through the various components of the Earth system, the impacts of deposition of atmospheric nutrients and contaminants to natural and managed ecosystems, and the feedbacks of ecosystems onto the atmosphere. New laboratory and field studies are needed to characterize these atmospheric chemistry processes for future use in predictive models.

The atmospheric chemistry community should expand interactions with the ocean, land surface, and other Earth science research communities to address these key scientific gaps:

A. Quantify the full suite of trace gases and particles deposited from the atmosphere and connect these to ecosystem responses.
B. Identify and quantify the chemical composition, transformations, bioavailability, and transport of nutrients and contaminants in the global atmosphere and their interactions with the biosphere.
C. Identify important feedbacks between atmospheric chemistry and the biosphere under global change.

SUPPORTING PROGRAMMATIC AND INFRASTRUCTURE PRIORITIES FOR ADVANCING ATMOSPHERIC CHEMISTRY RESEARCH

To understand the trends that are currently occurring in the field of atmospheric chemistry and the research emphases of different federal groups supporting work in this area, the Committee requested information from five government agencies. In summary, the judgement of the Committee is that the field of atmospheric chemistry has been expanding for the past several decades, but the amount of funding for research in the field has not increased substantially.

In this context, the Committee made recommendations to help support research for the next decade to enable the Priority Science Areas identified in Chapter 5 (see Figure S.2). These programmatic and logistical actions are primarily directed to the NSF Atmospheric Chemistry program and include the development of "tools;" the collection, analysis, and exchange of data; and increased collaborations within atmospheric chemistry and with other communities. The

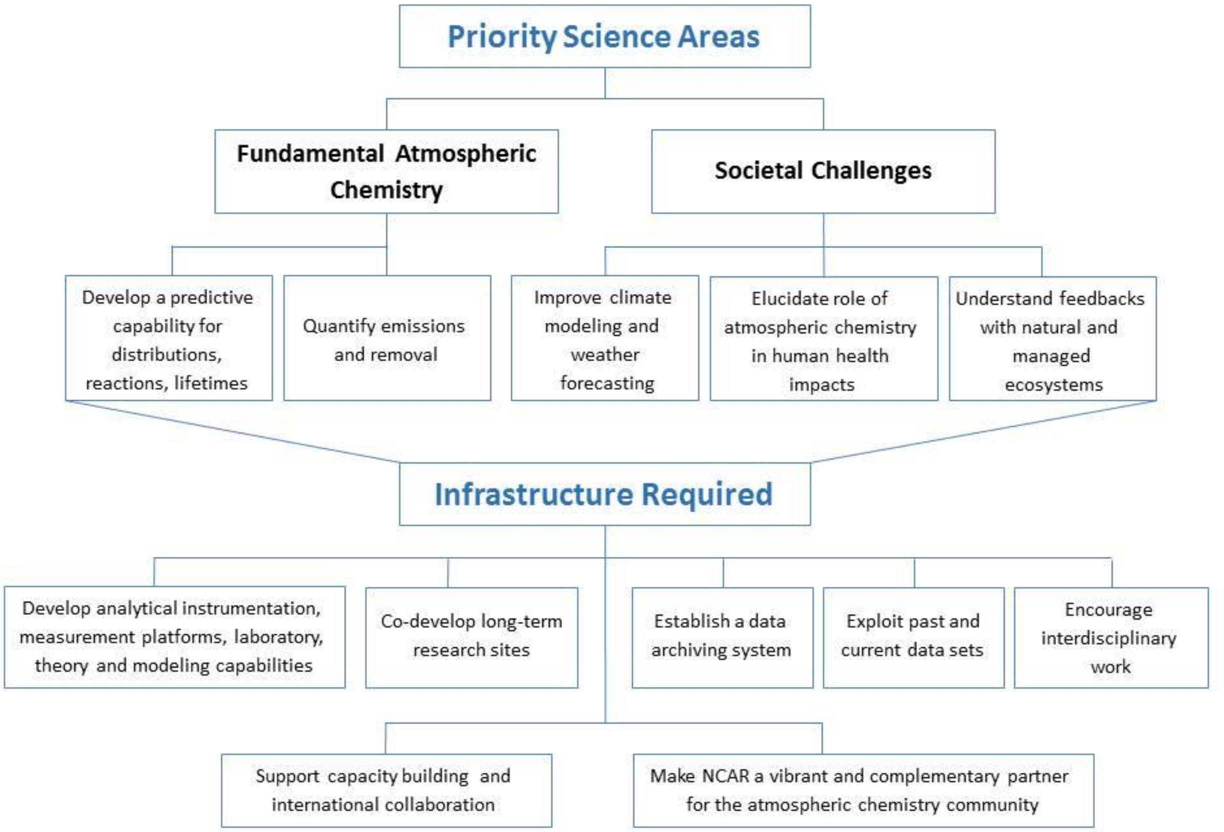

FIGURE S.2 The Committee's Priority Science Areas (Chapter 5) and recommended infrastructure and programmatic actions (Chapter 6). The National Center for Atmospheric Research (NCAR) is part of the infrastructure of atmospheric chemistry in the United States, and the peer-reviewed Atmospheric Chemistry program at NSF relies on partnering with NCAR; thus, the Committee also comments on NCAR's role.

Development of Tools for Atmospheric Chemistry Research

Instruments used in laboratory experiments and deployed for remote and in situ measurements are key tools used by atmospheric chemists. New analytical techniques, instruments, and instrument platforms are needed to support the Priority Science Areas above. Similarly, a range of modeling tools is central to the development of a predictive understanding of atmospheric chemistry. NSF will play an essential role in fostering the development of the next generation of many of these tools, as could industry and other government funding agencies.

NSF's Major Research Instrumentation (MRI) and Small Business Innovation Research/Small Business Technology Transfer Research (SBIR/STTR) programs have produced valuable breakthrough technologies that enabled improvements in atmospheric chemistry research. However, there are generally few of these opportunities for high-risk, high-reward proposals for instrument development in atmospheric chemistry at NSF. The Committee encourages the Atmospheric Chemistry Program to consider mechanisms for providing more support for development of instruments and measurement platforms by working more closely with other NSF programs and directorates. Viable mechanisms need to also be available within the Atmospheric and Geospace Sciences Division to submit proposals for new instruments and techniques that take extended periods of time to develop and test before they can be used to generate accurate and reliable data.

There are a wide variety of available modeling tools that vary in both spatial and temporal scale and technical approach. This diversity of approaches is needed to develop a broad toolbox to understand the complex problems in atmospheric chemistry. The disparate spatial and temporal scales of chemistry and transport in the atmosphere present a major challenge in building modeling tools and methodological approaches that effectively integrate across the scales. NSF should continue its investments in atmospheric chemistry and tracer-transport model development and applications, from developing and incorporating theoretical chemistry to predicting global composition. NSF could also emphasize endeavors that focus on modeling across spatial scales (urban to global) and temporal scales (weather to climate) to promote collaboration and coordination across agencies and different NSF divisions.

Recommendation 1: NSF should ensure adequate support for the development of the tools necessary to accomplish the scientific goals for the atmospheric chemistry community, including the development of new laboratory and analytical instrumentation, measurement platforms, and modeling capabilities.

Information Collection, Analysis, and Archiving in the Era of "Big Data"

The collection of measurement data over long periods of time allows the discernment of trends that are not apparent in one-time field projects. Research at long-term field sites representing different environments is not adequately supported in the United States. Changing this would require a large scale coordinated effort and commitment by the research community and multiple funding agencies.

Established research sites with core measurement capabilities and long-term knowledge about regional photochemistry, meteorology, ecosystem properties, and biosphere-atmosphere exchange processes are a critical resource for making and interpreting new measurements. A distributed set of research sites that take advantage of existing infrastructure in other programs as much as feasible would be most cost effective. An interagency panel could prioritize the long-term sites and determine:

- the required infrastructure;
- whether the sites would be centrally managed or managed by individual principal investigators;
- core measurements to be included with each site;
- procedures for the archiving of the samples collected at these sites; and
- criteria and a review process to support funding decisions.

Recommendation 2: NSF should take the lead in coordinating with other agencies to identify the scientific need for long-term measurements and to establish synergies with existing sites that could provide core support for long-term atmospheric chemistry measurements, including biosphere-atmosphere exchange of trace gases and aerosol particles.

Answers to research questions are often apparent only after intensive data analysis; for example, the synthesis and analysis of existing datasets can be applied to test models across various regimes and guide future research directions. However, funding is often insufficient to mine field data deeply for thorough analysis or to re-analyze existing datasets. Longer grant periods or supplemental installments to afford principal investigators the time and effort to continue analyses may be needed to accomplish these efforts. NSF should also encourage and support new projects that use data mining to advance the science—a cost-effective way to advance the atmospheric chemistry research agenda. For a fraction of the cost of a field study, NSF could dedicate funds to encourage analysis of existing high-quality data sets using powerful data mining techniques developed by the computer science community and perform detailed intercomparisons using satellites, field measurements, and models.

Recommendation 3: NSF should encourage mining and integration of measurements and model results that can merge and exploit past datasets to provide insight into atmospheric processes, as well as guide planning for future studies.

Management of large volumes of “big data” is becoming ubiquitous in atmospheric chemistry research as vast and multidimensional datasets are continuously generated. These datasets require increasingly large resources to manage. Mechanisms are needed for effectively and efficiently archiving, sharing, and mining data, including making them easily available to the broad scientific community and to the public.

The current availability of datasets varies substantially; some are archived at data centers while others are available only upon request from individual scientists. Agencies (in particular the National Aeronautics and Space Administration [NASA] and the National Oceanic and Atmospheric Administration [NOAA]) maintain data separately, and there is not a common data format that allows integration between models and measurements. NSF-funded research does require data management plans, but no central coordinated data archive and sharing system exists for atmospheric chemistry research.

NSF should require that future NSF-funded datasets be handled in a manner that allows ready access and comparison with previous datasets. The Committee envisions a centralized system for providing and supporting data management for atmospheric chemistry. Apart from providing facilities for data archiving, accessibility, and transparency, a centralized responsibility could assure that datasets are managed with expert preparation and fostering, along with sufficient and standard documentation and metadata. NSF could take the lead in coordinating with other federal and state agencies to establish such a system.

Recommendation 4: NSF should establish a data archiving system for NSF-supported atmospheric chemistry research and take the lead in coordinating with other federal and possibly state agencies to create a comprehensive, compatible, and accessible data archive system.

Imperative for Collaborations

Understanding and addressing challenges faced by society will rely on close integration of knowledge from multiple disciplines, including the physical, biological, and social sciences and engineering. While there are examples within NSF of programs that encourage interdisciplinary work, the Committee is concerned that mechanisms to support interdisciplinary work may encounter barriers due to NSF institutional and review structures. Interdisciplinary research often requires sustained long-term funding, which can be difficult to achieve using the typical three-year NSF grants.

Given the important cross-disciplinary aspects of the science priorities for atmospheric chemistry, the NSF Atmospheric Chemistry Program should explore multiple options to address these well recognized challenges that the Foundation faces. In some NSF directorates, an effective approach has been to fund either virtual or on-the-ground centers that draw together scientists who have different expertise and are often geographically dispersed. Other options include defining funding mechanisms whereby small, focused teams can integrate the necessary expertise from multiple disciplines; identifying and altering structures that discourage integration across disciplines; and placing value on a “cross-disciplinary integration” component of proposal evaluation. Both single agency and cross-agency efforts are an essential component of the Priority Science Areas identified above.

Recommendation 5: NSF should improve opportunities that encourage interdisciplinary work in atmospheric chemistry and facilitate integration of expertise across disciplines and across academia, institutes, government, and industry. This improvement may include support of focused teams and virtual or physical centers of sizes appropriate to the problem at hand.

The past success in understanding and applying atmospheric chemistry should be leveraged to improve air quality in many parts of the world. Although emissions have been reduced and air quality improved in the United States, many individuals are still living with dangerously high pollutant levels that impact their communities’ health and society. Working with underrepresented groups within the United States and with the international community is important for developing a global understanding of atmospheric chemistry and its impacts on human activities, especially as the importance of long-range transport of air pollution has become evident.

Understanding the interface between the processes controlling regional air quality and global atmospheric chemistry often requires establishing experimental programs with communities not traditionally included in the atmospheric chemistry community, especially those outside of domestic borders. This work may require nontraditional or international partnerships, and in some cases can encourage local scientific capacity building. Building the expert human capacity and observational and modeling capability at the regional level within the developing world through collaborations between U.S. atmospheric chemists and scientists in these regions has the potential to be a sustainable approach to addressing global air quality challenges. One possibility is establishing sites in developing countries that are part of larger networks for global scientific measurements. Another is the transfer of measurement and modeling capabilities to scientists in developing nations. Yet another is engaging underserved communities within the United States in addressing air quality issues through citizen science.

NSF already does a substantial amount to promote international collaborations. Activities within NSF (e.g., Partnerships for International Research and Education) and across multiple U.S. agencies (e.g., Partnerships for Enhanced Engagement in Research) are important for fostering global research programs and building global capacity in atmospheric chemistry.

Recommendation 6: NSF, in coordination with other agencies, should continue to encourage and support U.S. scientists involved in atmospheric chemistry research to engage with underserved groups, in capacity building activities, and in international collaborations.

Role of a National Center

Answering today's science questions requires significant resources and expertise to develop, maintain, and operate an array of instruments, platforms and laboratory tools, as well as significant expertise and computational resources to develop and run complex weather, chemistry, and climate models. A national center can be an optimal approach for providing these observational and computational capabilities because (1) dedicated center staff with expertise are most efficient at maintaining these complex capabilities; (2) NSF competitive processes are suitable for making support of these abilities available to individual or groups of principal investigators (PIs); and (3) the center can help foster the collaborative research needed to identify and solve critical science and societal problems. A national center can provide these resources and expertise to investigators in the broader community, primarily those served directly by NSF, while at the same time contributing directly to scientific advancements. In addition, a national center can also bring the community together by hosting a large steady flow of visitors of all stages in their careers, facilitating collaborations, holding conferences and workshops, and playing a leading role in the production of scientific assessments.

NCAR was established as a federally funded national center dedicated to achieving excellence in atmospheric science research, of which atmospheric chemistry is an essential part. Atmospheric chemistry research occurs within many divisions of NCAR, especially the Earth Observing Laboratory and the Atmospheric Chemistry Observations and Modeling (ACOM) Laboratory. ACOM serves as one of NCAR's primary connections to the atmospheric chemistry community; yet that community regularly questions how well ACOM has been fulfilling this complete vision of its role.

Many on the Committee have observed that ACOM's capabilities have diminished in the past decade or so—including departures of some prominent atmospheric chemists. This situation occurred at the same time that NCAR ACOM scientists have been pushed to provide instruments, measurements, and models for numerous projects, reducing the time for them to pursue their own research and/or development interests.

The Committee believes that it is essential for NCAR to find its unique role in atmospheric chemistry research, complementing and enhancing research by the broader community, and engaging individual PIs from universities, federal labs, and the private sector. NCAR can improve its standing within the atmospheric chemistry community by aligning its strategic vision with the role of a national center laid out above and with the original founding charter of NCAR. To achieve this vision, NCAR, in conjunction with NSF, will need to provide strong leadership, strategic allocation of resources, and guidance on balancing scientific excellence and community service.

In summary, the partnership between the competitively funded NSF atmospheric chemistry program and the facility-funded NSF programs at NCAR needs to be strengthened; if this vision can be realized, NCAR can play a pivotal role in facilitating greater scientific advances across the atmospheric chemistry community.

Recommendation 7: NCAR, in conjunction with NSF, should develop and implement a strategy to make NCAR a vibrant and complementary partner within the atmospheric chemistry community. This strategy should ensure that scientific leadership at NCAR has the latitude to set an energizing vision with appropriate personnel, infrastructure, and allocation of resources; and that the research capabilities and facilities at NCAR serve a unique and essential role to the NSF atmospheric chemistry community.

FINAL THOUGHTS

Similar to 30 years ago, the field of atmospheric chemistry research is in the midst of redefining its role in science and society. While atmospheric chemistry research has its foundations in the traditional disciplines of chemistry, physics, biology, geology, oceanography, engineering, and meteorology, as the field has grown it has become a robust area of basic science in and of itself.

In addition, atmospheric chemistry research is now clearly a vital part of building predictive capability for the Earth system and human impacts. Today, the field covers a wide swath of topics that integrate how an understanding of the fundamental chemistry of the atmosphere relates to the climate system, weather, ecosystems, and human society. These challenges involve nothing less than the health of our planet and its inhabitants. The Committee sees a deliberate shift in the field of atmospheric chemistry in the future to fully embrace its dual role—observing, learning, and discovering for the sake of fundamentally understanding the Earth system and the underlying chemical, physical and biological processes, while also making major contributions to addressing those challenges that directly affect society. Atmospheric chemistry research alone will not solve the challenges of global climate change or the impacts of air pollution on human and ecosystem health, but these challenges will not be solved without the knowledge that comes from atmospheric chemistry research.

Moving in both of these directions will take effort, investment, and a willingness to adapt by NSF, other agencies, and the atmospheric chemistry community as a whole. But after conversations with many members of our community during the course of this study, the Committee is convinced that we are ready for these challenges. The future of atmospheric chemistry research relies on the community to continue advancing our scientific knowledge and applying these findings to improve the world around us.

Chapter 1—Why This Report?

1.1 A CHANGING WORLD, A CHANGING ATMOSPHERE

Our world is changing at an accelerating rate. The global human population has grown from 6.1 billion to 7.1 billion in the last 15 years, and is projected to reach 11.2 billion by the end of the century (United Nations, 2015b). The distribution of humans across the globe has also shifted, with more than 50 percent of the global population now living in urban areas, compared to 29 percent in 1950.[2]

The world's demands have evolved with this expanding population and with advancing technologies. Energy demands are altering energy sources and extraction locations, atmospheric emissions, land use, and transportation. Industrial activity supporting increasing standards of living has altered the quantity and quality of goods transported worldwide. Changes in agricultural practices, urbanization, and other anthropogenic (human-caused) activities in response to increasing population have led to land cover and land use change around the globe, expanding not only the landscapes used for crops and livestock cultivation, but also the use of fertilizers and other techniques that improve food production. This growth in human activities and changes in their spatial distribution, along with natural disturbances such as volcanic eruptions and lightning-induced wildfires, have a substantial impact on the Earth system. As a world, we have entered a period when human activities are overriding some natural processes and leading to a discernable impact on the Earth system. In recent decades, this period has been labeled the "Anthropocene," an era in which human activities are having irreversible impacts on the Earth system (Crutzen and Stoermer, 2000; Steffen et al., 2007).

All of these processes directly affect the atmosphere—the only environmental medium in which humans are unavoidably and continuously immersed and linked to other humans around the globe. Atmospheric motion transports anthropogenic and natural emissions around latitude bands within weeks, between the tropics and the poles within months, and between northern and southern hemispheres within a year (Jacob, 2000). Because the atmosphere mixes far more quickly than other media such as water or soil and flows unimpeded across political boundaries, emissions to the atmosphere are rapidly distributed from one region to another, requiring global cooperation to achieve positive change. The atmosphere is a global commons that connects all life.

The composition of the atmosphere impacts society in a number of ways—from changes in weather and climate to food production and human health. Climate change is one of the greatest environmental challenges facing society today (NRC, 2011). Earth's average surface temperature has increased by more than 1.4°F (0.8°C) over the past 100 years, and the 21st century has already experienced 15 of the 16 warmest years since 1880.[3] Sea level, Arctic sea ice, and other parts of the planet are changing, including alteration of the water cycle, changes in biogeochemical cycles, and biodiversity loss (Sala et al., 2000). Recent studies establish a link between climate change and certain types of extreme weather events (NASEM, 2016), which come with high economic costs. For the period of 1980–2011, it was estimated that droughts and

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[2] World Bank Open Data: http://data.worldbank.org/.

[3] Global Analysis from the NOAA National Centers for Environmental Information: http://www.ncdc.noaa.gov/sotc/global/201513.

heat waves cost the U.S. economy a total of $210 billion (Smith and Katz, 2013). The Pentagon's strategic plan indicates that climate change, which is largely caused by changes in atmospheric composition, is an immediate leading security risk to the United States (DOD, 2014). Atmospheric chemistry plays an important role in controlling climate change; for example, aerosol particles impact climate through direct radiative forcing, by indirectly modifying clouds, and by modifying carbon uptake. Atmospheric chemistry also leads to the formation of air pollutants that are greenhouse gases, such as ozone. Society needs forecasts of those aspects of the changing climate structure that most urgently threaten human health, security, economic opportunity, social stability, and confidence in the future. Such forecasts require a far more fundamental understanding of the physical, chemical and biological relationships intrinsic to the Earth's climate system than is currently within the grasp of the scientific community.

Air pollution represents a serious threat to human health both in the United States and worldwide. The annual human health costs due to air pollution in the United States alone are estimated to be in the range of $71–$277 billion per year (Muller and Mendelsohn, 2007). One out of eight deaths globally is caused by air pollution (WHO, 2014), including those due to high levels of indoor air pollutants in both homes and places of work.

Lastly, future food production is vulnerable to both climate change and air pollution, with serious implications for global food security (Tai et al., 2014). Reduced production of soybeans and maize from ozone exposure during the period from 1980 to 2011 is estimated to have caused losses of about $9 billion per year in the United States (McGrath et al., 2015). Projections to the year 2030 for those crops along with wheat predict losses of $12–$35 billion per year globally (Avnery et al., 2011).

Climate change and other human-induced global changes contribute to a growing sense of urgency to improve overall understanding of the physical, chemical, and biological processes controlling the composition of the atmosphere and how it impacts the health of humans and ecosystems.

1.2 THE SCIENCE OF ATMOSPHERIC CHEMISTRY

Atmospheric chemistry research encompasses *understanding* the processes controlling the chemical composition of the atmosphere through development of fundamental scientific knowledge, and *applying* that knowledge to improve the quality of life for humans and ecosystems. As such, the field bridges "basic" research, which addresses fundamental questions about how the world works, and "applied" research, which is motivated by a desire to answer questions of relevance to society. This marriage has been described as "use-inspired basic research" (Stokes, 1997).

Fundamental atmospheric chemistry research focuses on natural processes and human activities, and how they control the chemical composition of Earth's atmosphere. Ultimately, the research addresses processes that control the local, regional, and global distributions of atmospheric chemical composition—chemical inputs to, transport and transformations within, and removal from the atmosphere. Because the atmosphere includes a wide range of temporal and spatial scales and many different chemical species whose concentrations can be affected by emissions, transport, reactions, removal, and other meteorological variables, a combination of approaches are essential. These include laboratory and theory studies to elucidate processes on a molecular scale, field observations that provide atmospheric composition, and models that integrate the current understanding from laboratory, theory and field studies, and provide a mechanism for assessing different future alternative scenarios.

These approaches are mutually synergistic, with a discovery in one area often leading to new advances in many others, and each has its challenges. Laboratory studies provide controlled conditions to evaluate individual chemical pathways and provide molecular properties required for atmospheric measurements. Theory provides support for interpretation of laboratory results and can provide predictions that can then be tested by experiments. Challenges include the large number of different species and phases involved—gas, liquid, and solid, as well as interfaces and partitioning between the phases. An additional challenge involves developing a wide range of approaches for laboratory studies that encompass single reactions to complex systems that simulate conditions like the real atmosphere, or can be reliably extrapolated to atmospheric conditions, providing a bridge to field measurements and modeling studies.

Field studies of atmospheric composition provide discoveries in previously unexplored environments, test the knowledge of atmospheric chemistry through intensive field campaigns involving simultaneous measurements of many different species, and discern meaningful trends when time scales are sufficiently long. Challenges include the need to study chemical processes as they happen, recognizing that most of the atmosphere's volume is decoupled from the surface of the Earth and human emissions. This requires simultaneous measurements of a multitude of atmospheric constituents at low concentrations, which relies on sensitive and specific instrumentation as well as observing platforms that can carry these instruments to measurement locations that can be difficult to access.

Models integrate the knowledge from laboratory, theory, and field studies to predict atmospheric chemistry and composition across a range of temporal and spatial scales, an integral part of simulating the complex Earth system. These models are also central to testing the relative importance and synergies between mechanisms and processes identified in the laboratory and field for controlling the regional and global distributions of atmospheric species. Challenges include developing models that cover the wide ranges of temporal and spatial scales, while accurately capturing processes from the molecular and sub-second scales on which chemical processes occur, to the global and decadal scales relevant to describe societal impacts.

The pioneers who settled the frontier of atmospheric chemistry were trained in a range of disciplines. Today's atmospheric chemists are from an equally rich range of backgrounds, including chemistry, physics, mathematics, statistics, biology, meteorology, oceanography, engineering, and environmental science. These diverse backgrounds and skills are required to tackle the problem-centered, use-inspired challenge of developing a predictive capability for the changing atmosphere.

1.3 ATTAINING A PREDICTIVE CAPABILITY

Choices made by society change the chemical composition of the atmosphere, which in turn affects societally important issues such as air quality, climate and weather, water resources, human health, agriculture, and ecosystems. These impacts feed back into societal choices, e.g., a choice to move out of highly polluted areas or change behavior that drives emissions. Development of a more comprehensive understanding of societal challenges and their possible solutions is a complex problem spanning responses in human behavior and the natural world.

This endeavor has led to the rise of "big science" that requires coordinated and focused interdisciplinary efforts.

Minimizing negative impacts, maximizing standards of living, and avoiding unintended negative consequences requires a comprehensive understanding of the systems and feedbacks involved in the atmosphere. Well-guided research in atmospheric chemistry can improve this understanding. Finding solutions for problems such as the ozone hole, acid deposition, and polluted urban regions air in the United States all followed a similar pattern (see Chapter 2). Beginning with a sufficient understanding of the atmospheric system, societal impacts of particular human activities were identified, the gaps in scientific understanding were defined, and fundamental research was conducted to fill the gaps. The core contribution of atmospheric chemistry research was the creation of a predictive framework that integrated the physical understanding of atmospheric chemistry with the outcomes of potential policies. This research was then synthesized for policy makers to provide a scientific basis for informed choices (see Box 1.1). As one example of quality of life improvement following policy changes, decreasing concentrations of chemical pollutants in the atmosphere (including nitrogen dioxide, ozone, and particulate matter) in the 1990s and 2000s have been associated with significantly lower rates of childhood bronchitis (Berhane et al., 2016).

Society would benefit from further development of the ability to accurately predict the impact of human choices on the atmosphere. A predictive framework must synthesize atmospheric chemistry research results with information from physical and ecological, technological, and social science fields related to a specific societal problem. Attaining such a predictive capability requires an understanding of the Earth system developed from laboratory and theoretical studies of fundamental atmospheric chemistry and physics, instruments and atmospheric observations to establish atmospheric constituents and processes, and computational models to integrate understanding. This integration allows attribution of causes of an observed societal impact to particular societal choices, such as linking stratospheric ozone depletion with the use of chlorofluorocarbons (CFCs), acid deposition with burning high-sulfur coal, or air quality impairment due to emissions from fossil fuel combustion. Evaluation of these models with observations is critical because uncertainties affect predictions in the causal chain from human activity to impact, and thus the associated costs and benefits of actions. Assessment of complex models is then required to evaluate their fidelity and completeness, as well as to develop simplified response functions that capture key relationships and sensitivities and facilitate integration into analyses that cover many facets of physical, economic, and human systems.

The predictive framework must be able to replicate immediate societal impacts with high fidelity and also predict the responses to alternative scenarios beyond the range of immediate observations. The framework has predictability when it is able to simulate with confidence not only what is, but also what will be. A predictive framework also needs to represent the observed extremes of a societal impact, as well as slowly evolving societal choices. If there is political motivation, predictions enabled by a robust framework can lead to the development of societal and technological solutions, as well as adaptation or mitigation strategies. Examples can be found within each of the success stories described below in Chapter 2. ***Establishing an effective predictive capability is an ultimate goal of atmospheric chemistry research.***

BOX 1.1
Support for Decision Makers

Decision makers require science to provide a predictive capability that links human activity to environmental impacts. It is possible to affect the level and nature of these impacts by choosing policies, for example, that favor certain forms of energy or that set emission standards. Decision makers can optimize these policies with tools that characterize the Earth system response. Some of these tools for decision support include:

Risk management: Air quality risk management has been used for decades by governmental agencies to reduce risk to human health (Davidson et al., 2008). Corporations and governments are also increasingly using predictive modeling of weather and climate to mitigate financial risks and to gain competitive advantage. A more advanced form of risk management tightly integrates environmental statistics and predictive models with a numerical representation of perceived cost and benefit; this approach can be used for applications ranging from atmospheric science research (Small et al., 2011) to decisions affecting air quality (Garner and Thompson, 2012).

What-if scenarios: Environmental changes attributable to a particular policy or action can be estimated by taking the difference between two simulation results. For example, comparative simulations have been used to evaluate the change in regional air quality and health impacts after more stringent particulate matter regulations were in place (EPA, 2012), and to assess benefits to climate, health, and agriculture caused by eliminating short-lived climate forcers (Shindell et al., 2012).

Environmental economics: The study of the way in which decisions about the use and distribution of resources affect human, environmental, and ecosystem health is known as environmental economics (Field and Field, 2009). This analysis requires some valuation of environmental impacts, often in monetary terms. These costs or benefits can then be used to evaluate the monetary impact of decisions, regulations, or development trajectories. Incorporation of environmental externalities to create a fuller cost accounting can be used to seek pathways that have the greatest overall benefit.

Integrated assessment: The response of linked economic and environmental system components is investigated by combining components of many disciplines, from international trade to atmospheric concentrations to agricultural productivity, into integrated assessment models (Cofala et al., 2007; Nordhaus, 1993). This type of model has been used to explore the circumstances required to meet environmental goals such as climate stabilization (Krey et al., 2014). Notable global-scale assessment successes include the recent scientific assessment of ozone depletion (WMO, 2014), and the Intergovernmental Panel on Climate Change (IPCC) process and its series of reports on climate change, including the most recent synthesis report (IPCC, 2014).

1.4 CONTEXT FOR THIS STUDY

It has been more than 30 years since the last comprehensive report to examine the field of atmospheric chemistry (*Global Tropospheric Chemistry: A Plan for Action* [NRC, 1984a]) was published. That report led to increased emphasis on understanding the global character and extent of natural and human perturbations to the global troposphere. Since the 1984 report, the analytical capabilities and tools used to measure the composition of the atmosphere have improved substantially in terms of sensitivity, timescale, and chemical complexity. These advances allow scientists to ask new and detailed questions that probe the chemistry of the atmosphere, and design and perform studies that support the development of predictive models. At the same time, dramatic advances in computational resources have enabled the development

BOX 1.2
Statement of Task for This Report

An ad hoc committee will identify priorities and strategic steps forward for atmospheric chemistry research for the next decade, in the context of the current state of knowledge, ongoing research activities, and resource availability. The Committee will report a compelling research strategy and identify where additional investments in research infrastructure could best advance scientific understanding. The report will include the following elements:

- A brief summary of the rationale and need for supporting a comprehensive U.S. research program in atmospheric chemistry, including how research in this area contributes to advancing our understanding of climate change, air quality, the carbon and nitrogen cycles, the energy and water cycles, and the overall role of the atmosphere in Earth system science.
- A commentary on the broad trends in laboratory, field, satellite, and modeling studies of atmospheric chemistry, as well as application of atmospheric chemistry knowledge that may influence the overall field of earth Sciences in the coming decade.
- A determination of the priority areas of research for advancing the basic science of atmospheric chemistry over the coming decade. In prioritization, the Committee should consider the need for a balance among laboratory studies, field campaigns, modeling efforts, and instrument development. The Committee is requested to provide research areas/ topics sorted by their prioritization, and to explain how the priorities were developed.
- An analysis of the research infrastructure needed to address the priority research topics identified in the preceding point and identification of the highest priority needs for improvements in this infrastructure. This analysis will include an assessment of the need for new measurement technologies, observational platforms, and major infrastructure investments in atmospheric chemistry over the next decade.

The Committee's report should incorporate input from the broader atmospheric chemistry research community, including scientists working in academia, government, and private sector. The Committee should consider how the proposed research agenda relates to the broader federal agency and international context for atmospheric chemistry, but focus on those activities that might best be supported by the National Science Foundation. The Committee should not make specific budget recommendations, but should comment generally on budget implications as part of determining priority areas for research.

of more complex and comprehensive model descriptions of atmospheric chemistry within the Earth system. These models, informed by new laboratory, theory and observational studies, can provide better predictions of the rapidly changing future atmosphere to inform policy choices, potentially protecting human health and well-being in the face of global change. In light of the rapid changes to the Earth system combined with impressive scientific advancements of the past few decades, it is clear that the next decade of research will be pivotal for advancing the understanding of atmospheric chemistry and its role in human welfare.

At the request of the U.S. National Science Foundation (NSF), the National Academies of Sciences, Engineering, and Medicine established the Committee on the Future of Atmosphere Chemistry Research. The Committee was charged with identifying priority areas of research for the next decade that will advance the basic science of atmospheric chemistry, developing a compelling strategy for the future of atmospheric chemistry research, and identifying where additional investments in research infrastructure could best advance scientific understanding. See Box 1.2 for the Committee's Statement of Task and Appendix A for the list of Committee

Members. The report was developed for the NSF's Atmospheric Chemistry Program, which "supports research to measure and model the concentration and distribution of gases and aerosols in the lower and middle atmosphere."[4] However, the results presented will be of interest to other agencies and programs that support atmospheric chemistry research.

In addressing its task, the Committee has considered the past contributions and future priorities for atmospheric chemistry research to examine natural processes and human-derived changes over time—from yesterday, through today, and continuing to tomorrow. The Committee gathered extensive input from the atmospheric chemistry community—including scientists working in academia, government, and the private sector—in a series of open public forums and incorporated that input into the results presented here (see Appendix B for a description of the community input gathering process).

This report focuses on activities that the NSF Atmospheric Chemistry Program might best support, within the broader context of atmospheric chemistry research as supported by other agencies both nationally and internationally. The report was written with a variety of audiences in mind, from funding agencies to researchers to policy makers, and as such, the technical level varies. In Chapter 2, the Committee demonstrates how atmospheric chemistry has historically played a central role in building basic scientific understanding and applying that knowledge for the benefit of society. Chapter 3 reviews the fundamental principles underlying atmospheric chemistry research at a higher technical level using recent advances over the past decade. Chapter 4 identifies key areas where atmospheric chemistry needs to contribute to societally relevant challenges, and Chapter 5 identifies priorities for a comprehensive U.S. research program in atmospheric chemistry to advance the most relevant basic and applied science over the coming decade. The grand challenge of predicting and ameliorating societal impacts requires integration with research domains beyond atmospheric chemistry. While this report addresses the Statement of Task by focusing on potential contributions of atmospheric chemistry research, Chapter 5 also identifies areas in which cross-disciplinary integration is likely to lead to major advances. Chapter 6 reviews broad trends in laboratory, field, satellite, and modeling studies within atmospheric chemistry and provides recommendations for improved research infrastructure and approaches to better address the priorities outlined in Chapter 5.

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[4] http://www.nsf.gov/funding/pgm_summ.jsp?pims_id=11692&org=AGS&from=home (accessed June 2016)

Chapter 2—Remembering Yesterday

Atmospheric chemistry research has provided some spectacular successes, including in improving air quality, reducing acid deposition, and addressing upper atmosphere ozone depletion. These advances have saved lives, protected ecosystems, and improved the social welfare of people in the United States and around the world. There is a similar theme to each of these stories. Each involves atmospheric chemistry research focused on rapid change in the environment, which led to improved understanding of the drivers of a problem with real-world implications. Further research improved predictive capability to inform policy choices and responses to the problem, with research continuing to inform the status of the problem today. A brief history of these examples and the central role played by atmospheric chemistry research and researchers, including those whose contributions were recognized with Nobel Prizes and the U.S. National Medal of Science, follows.

2.1 PAST SUCCESSES OF ATMOSPHERIC CHEMISTRY IN A CHANGING WORLD

Improving Air Quality

Air pollution had been recognized as a problem in cities for hundreds of years (Brimblecombe, 1978; Gaffney and Marley, 2003), but by the 19th century it became much more visible and pervasive. The industrial revolution led to dramatically increased emissions of gases and particles into the atmosphere from fossil fuel combustion. Early episodes of air pollution were typically driven by high emissions of particles and sulfur dioxide, which is converted to sulfuric acid in a process that is accelerated in fog and clouds. This situation led to the description of air pollution as “smog,” a combination of the words “smoke” (from particles) and “fog.” The adverse health effects of smog have been documented for hundreds of years (Brimblecombe, 1978). In the United States, 20 deaths were attributed to extreme smog conditions in Donora, PA, in 1948, and the “poster child” for severe air pollution was an episode in London in December 1952 that led to an estimated 4,500 excess deaths during the episode, and a total of 13,500 through the following March (Bell et al., 2004; Snyder, 1994).

In the late 1940s, a new type of damage to agricultural crops was observed in Southern California and traced to interactions with atmospheric constituents (Middleton, 1956; Middleton et al., 1950). Shortly thereafter, chemist Arie Jan Haagen-Smit (see Box 2.1) demonstrated in a series of laboratory studies that photolysis in air of a mixture of hydrocarbons and nitrogen oxides (NO_x) led to the formation of ozone (O_3) and a number of associated pollutants whose effects were consistent with the observed plant damage. Despite recalcitrance on the part of industry to acknowledge the contributions from vehicle exhaust emissions (Hahn, 1967), this explanation is now well-established: the increasing emissions of volatile organic compounds (VOCs) and NO_x that came in large part from automobile exhaust were major contributors to this new kind of air pollution, known as “photochemical” smog (e.g., Haagen-Smit, 1950). In his classic book “Photochemistry of Air Pollution,” Philip Leighton described the fundamental reaction mechanisms involved (Leighton, 1961). Although it would be almost a decade before the importance of hydroxyl radicals in air was recognized, this book presented the likely

BOX 2.1
National Medal of Science Winners Who Made Major Contributions to Atmospheric Research and Its Impacts on Humanity

Several scientists have not only made seminal contributions to the development of the discipline of atmospheric chemistry, but also provided leadership and innovation that has had a major impact on the protection of the atmospheric environment and its impact on human health and welfare. Each of these individuals has been recognized with the U.S. Presidential National Medal of Science (see Figure A). Arie Haagen-Smit elucidated the basic ingredients and chemistry of photochemical smog formation. An organic chemist, he made some of the first measurements of atmospheric ozone in Los Angeles by following the rate of cracking of rubber strips mounted outside his laboratory. Haagen-Smit was also involved in the policy and regulation of air pollution as the first Chair of the California Air Resources Board in 1966, and his stakeholder engagement skills helped the state lead the way in understanding and addressing photochemical air pollution (medal awarded in 1973).

Arie Haagen-Smit (1973)

Harold Johnston (2002)

Susan Solomon (1999)

Gene Likens (2001)

Charles Keeling (2002)

FIGURE A U.S. Presidential National Medal of Science winners who have made seminal contributions to atmospheric chemistry research and the years the medals were awarded. SOURCES: Haagen-Smit: Photo by Floyd Clark, courtesy of Caltech Archives; Johnston: Lawrence Berkeley National Laboratory; Solomon: American Chemical Society; Likens: Cary Institute for Ecosystem Studies; Keeling: University of California at San Diego.

A number of other scientists whose research was crucial to the progress of atmospheric chemistry in the 20th century have also been awarded the Presidential National Medal of Science. Harold Johnston studied fundamental reaction mechanisms and kinetics of nitrogen oxides (NO_x) and showed that NO_x emissions from stratospheric aircraft would have dramatic consequences in terms of depleting the protective upper atmosphere ozone layer. Susan Solomon's theories helped elucidate the unique chemical processes occurring in the Antarctic ozone hole, confirming the role of chlorofluorocarbons (CFCs) in stratospheric ozone depletion. She led the first expedition to Antarctica to determine if the ozone was being depleted by reactive halogens formed by reactions on polar stratospheric ice crystals. Solomon also helped support the Montreal Protocol in calling for CFC reductions and later co-chaired the Intergovernmental Panel on Climate Change. Gene Likens connected fossil fuel combustion and increased acidity of rain in the eastern United States. His long-term research at the Hubbard Brook Experimental Forest in New Hampshire spurred public awareness and further study into the effects of human activities on ecosystems. Charles Keeling, a geochemist, conducted seminal measurements of carbon dioxide (CO_2) that established the link between fossil fuel burning and climate change. His records at Mauna Loa, Hawaii established the "Keeling Curve" as the longest continuous record of atmospheric CO_2 in the world.

importance of free radical chemistry and recycling in the formation of photochemical air pollution.

Levels of fine particles and O_3 in the atmosphere are key determinants of air quality (e.g., Jerrett et al., 2009a; Rao, 2015; Samet et al., 2000) because they are associated with adverse effects on human health and welfare, agricultural damage, and reduced ecosystem productivity (EPA, 2014a,b; UNEP and WMO, 2011). Understanding the processes driving photochemical smog formation has developed over many decades. Ground-level O_3 is known to be formed from reactions involving NO_x, VOCs, and sunlight (Finlayson-Pitts and Pitts, 2000; Haagen-Smit and Fox, 1954; Leighton, 1961; NRC, 1991; Seinfeld and Pandis, 2006). Particles can be directly emitted or formed by reactions of gaseous precursors in air. Particulate matter ranges in size from particles smaller than 10 nanometers in diameter to particles as large as 10 micrometers. A measure of fine particles widely used by regulatory agencies is $PM_{2.5}$, which refers to particulate matter smaller than 2.5 micrometers in diameter. The majority of urban $PM_{2.5}$ forms in the atmosphere from reactions of NO_x, sulfur dioxide (SO_2), ammonia, amines, and volatile organic compounds (VOCs). There is increasing concern about ultrafine particles (particulate matter smaller than 100 nanometers in diameter) from outdoor and indoor pollution sources, and their toxicity. Understanding the emissions and chemical transformations of precursor gases to PM and O_3 has been instrumental in driving regulations to improve air quality.

A number of regulatory policies to address air pollution have been put into place at both the federal and state levels in the United States. These include the Air Pollution Control Act (P.L. 84-159, 69 Stat. 322, 1955) and the Clean Air Act (P.L. 88-206, 77 Stat. 392, 1963), which with subsequent amendments have significantly improved air quality and reduced adverse effects over the last 50 years. For example, regulatory policies emphasizing NO_x control have led to significant decreases in O_3 in polluted areas such as Southern California, along with declines in CO, $PM_{2.5}$, and related pollutants over recent decades (Hidy and Blanchard, 2015; NRC, 1991). Figure 2.1 shows the trend in air pollutants falling from unsafe levels to values below National Ambient Air Quality Standards over the United States.

Reasons for this decrease in pollutant emissions include progress in energy efficiency, shifts to renewable technologies, and the role of regulations in raising the cost of business for polluting firms that then increases the competitive advantage of companies using clean technologies (EIA, 2015; IEA, 2014). As a result, California air quality improvements, for example, have been associated with increased productivity, life expectancy, and medical cost savings (CAPCOA, 2015). On a national scale, economic growth has occurred in concert with increased energy consumption and reduced pollutant emissions (see Figure 2.2). The atmospheric chemistry research community has made major contributions to the development of such demonstrably effective control strategies.

The lessons learned from reducing air pollutant levels in the United States provide a scientific foundation for other areas in the world where emissions have increased dramatically in recent years. Poor air quality remains an issue in many parts of the developing world. It has been estimated that outdoor air pollution is responsible for 3.3 million premature deaths per year globally, with more than two-thirds of the deaths occurring in Asia (Lelieveld et al., 2015). Because long-range transport can carry pollutants to downwind regions or countries, international strategies may be a part of future air quality management. In addressing these challenges, atmospheric chemistry will continue to play a vital role in improving the understanding of the impact of air pollution on human health (see Chapter 4.2).

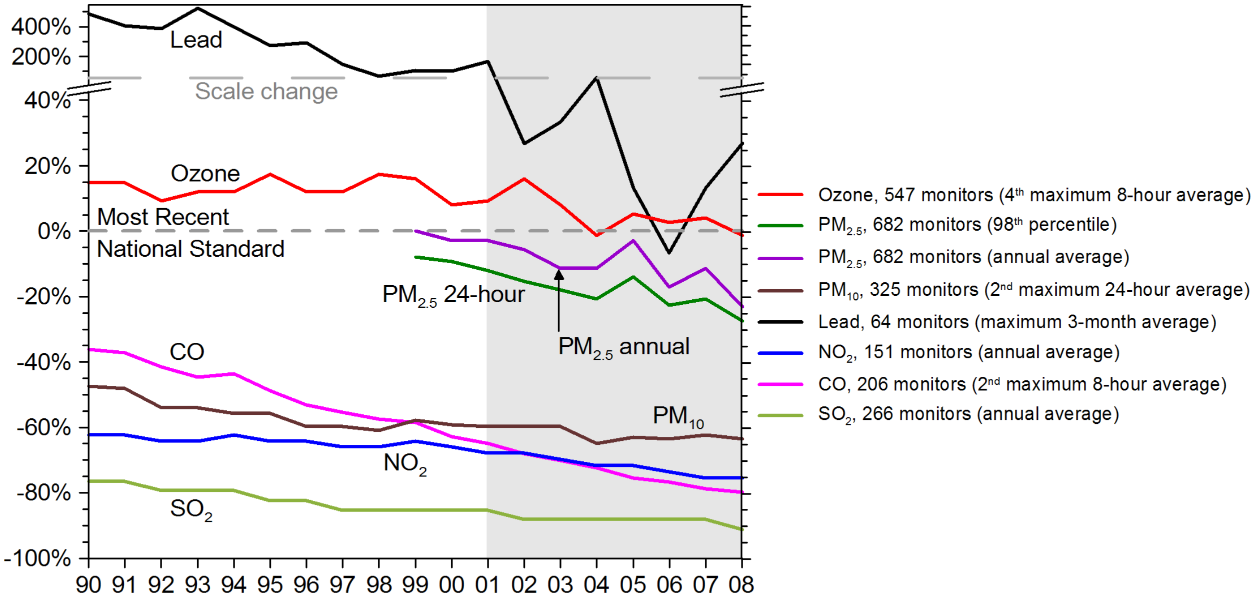


FIGURE 2.1 Trends in percentage deviation from National Ambient Air Quality Standards (NAAQS) for ozone, particulate matter smaller than 2.5 and 10 micrometers in diameter ($PM_{2.5}$ and PM_{10}, respectively), lead, nitrogen dioxide (NO_2), carbon monoxide (CO), and sulfur dioxide (SO_2) concentration over the United States from 1990 through 2008. Declines in contaminant concentration have been made possible by advances in understanding sources of emission and mechanisms of transformation in the atmosphere. Note the scale change for the trend in lead, designated by the gray shaded area. SOURCE: https://www3.epa.gov/airtrends/2010/dl_graph.html.

Reducing Acid Deposition

Deposition of acids from the air onto surfaces is a direct consequence of fossil fuel burning. Combustion emits NO_x and SO_2, which are converted in the atmosphere into nitric and sulfuric acids, respectively (NRC, 1983, 1986; Oppenheimer, 1983). When these compounds are returned to Earth, acidification of ecosystems and surface waterways occurs, inducing a cascade of deleterious effects. The writer John Evelyn first described the negative effects of city air on ancient marble in 17th century England (Evelyn, 1661). However, the topic of acid pollution coming from the atmosphere was not widely reported until the Industrial Revolution dramatically increased emissions of NO_x and SO_2. For example, in 1852, chemist Robert Smith coined the term “acid rain” when he noted higher acidity rainfall in industrial regions relative to coastal areas in England and indicated it was a result of sulfuric acid from coal combustion (Smith, 1852).

More than a century after Smith’s finding, Odén (1968) showed that much of the acidity in Scandinavian rains resulted from long range atmospheric transport following emissions of sulfur from England and central Europe. Also in the 1960s, multi-disciplinary research was initiated to understand the consequences of acid rain falling in the eastern United States and Canada. Biologists and limnologists conducted large-scale manipulative experiments on farms and lakes to probe the links between reduced agricultural crop production, “dead” lakes, damaged forests, whole-ecosystem effects, and acid deposition (Beamish and Harvey, 1972; Lee et al., 1981). Through their studies of the regional distribution of acid rain and its importance for aquatic and terrestrial ecosystems at the Hubbard Brook experimental forest in New Hampshire

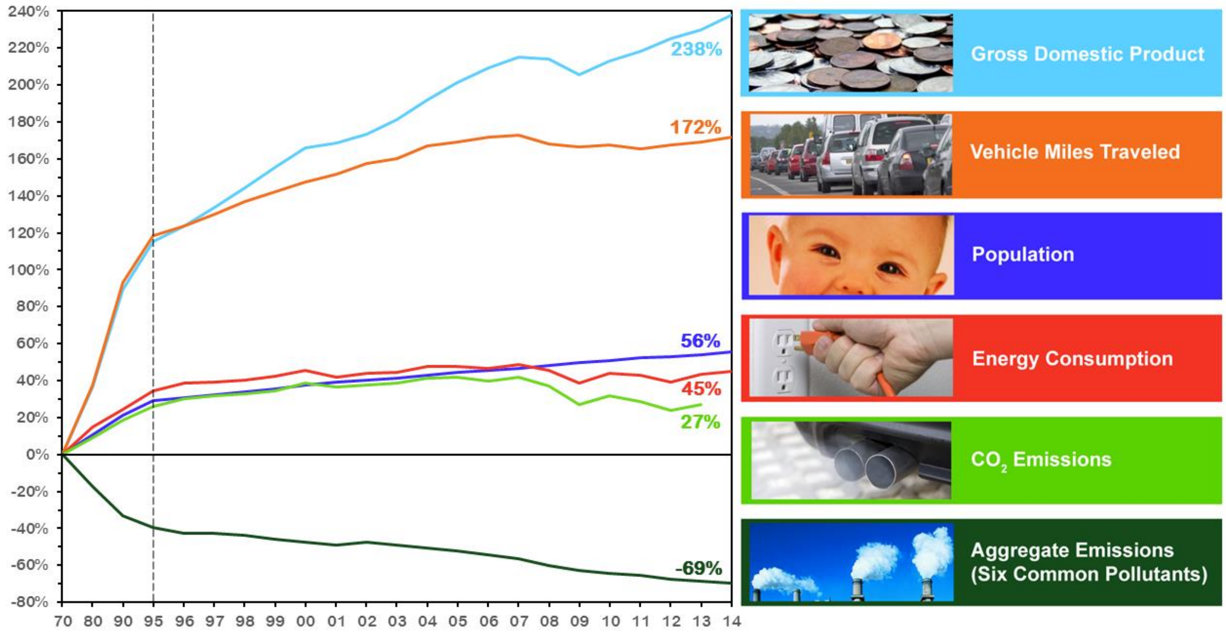
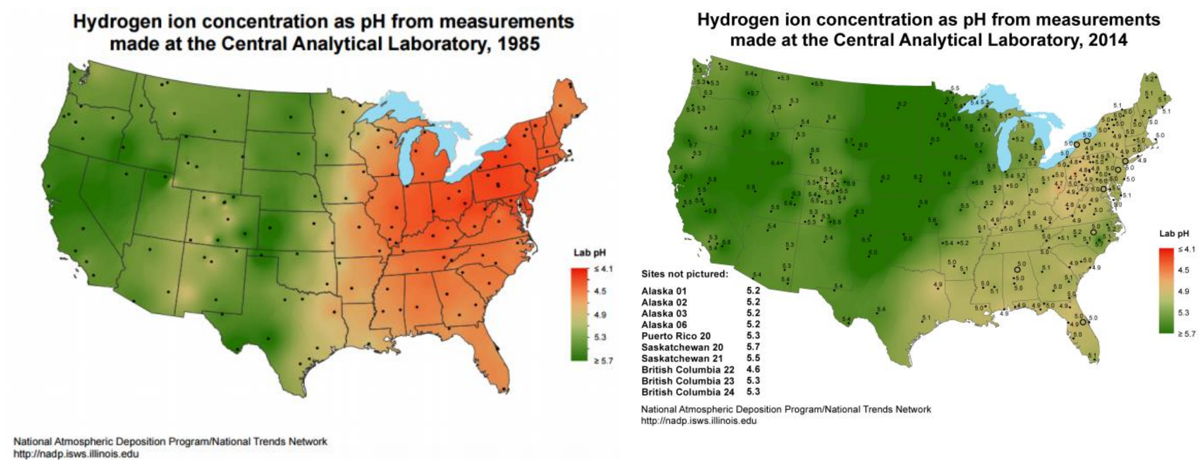


FIGURE 2.2 Percent changes from values in 1970 for gross domestic product, vehicle miles traveled, population, energy consumption, CO_2 emissions, and aggregate emissions from the six most widespread air pollutants (carbon monoxide, ozone, lead, nitrogen dioxide, particulate matter, and sulfur dioxide) through 2014 nationally over the United States. The graph shows decreasing air pollutant emissions over time since passage of the Clean Air Act, despite growth in U.S. population, economy, energy use, and miles driven. SOURCE: https://www3.epa.gov/airtrends/aqtrends.html.

beginning in the early 1960s, Gene Likens (see Box 2.1) and colleagues were instrumental in bringing the problem of acid rain to public attention in the United States (Likens et al., 1972). Coupled with research to better understand long range transport of atmospheric pollutants and consistent with Odén's work in Sweden, chemists proposed that power plant emissions from Midwestern states could arrive in the Northeast region of the United States and damage ecosystems through the deposition of acid rain (Nelson et al., 1984).

Throughout the 1970s and 1980s, researchers discovered that acid deposition reduces biodiversity and can substantially alter ecosystems with smaller changes in acidity than previously believed (Schindler, 1988). Furthermore, the negative impacts of direct acid deposition are compounded because acidity causes toxic species, such as trace metals, to remobilize in the environment (Exley et al., 1991). In addition to the ecosystem effects of acid deposition, the formation of highly concentrated inhalable droplets of acidic fog was proposed to likely have substantial effects on human and ecosystem health (Hoffman, 1984; Jacob et al., 1985).

Elucidating the causes and effects of acid deposition has been a broad effort involving researchers from a variety of fields. Fundamental atmospheric chemistry research played a vital role in understanding the mechanism behind the conversion of SO_2 and NO_x into acids and with this knowledge, researchers were able to predict that decreases in emissions of SO_2 and NO_x would lead to lower acid rain levels. The recognition that pollution can cause effects well beyond local environments began reaching the public in the 1970s, leading to widespread pressure for political and legal action. This realization resulted in the 1980 Acid Deposition Act, which was



Hydrogen ion concentration as pH from measurements made at the Central Analytical Laboratory, 1985

National Atmospheric Deposition Program/National Trends Network
http://nadp.isws.illinois.edu

Hydrogen ion concentration as pH from measurements made at the Central Analytical Laboratory, 2014

National Atmospheric Deposition Program/National Trends Network
http://nadp.isws.illinois.edu

FIGURE 2.3 Change in acidity in the atmosphere over the continental United States between 1985 and 2013. Comparison of the two maps shows the positive effects of legislation based on research in chemical transformation and transportation from sources to affected areas. SOURCE: NADP, 2014.

passed to comprehensively support a program addressing the sources of acid rain and dry deposition and their long-term effects over a broad geographic area. Research was also funded to examine potential pollution control measures, which resulted in Clean Air Act amendments and supporting legislation such as the Cross-State Air Pollution Rule[5] to ameliorate effects of pollutant transport between states. Thus, informed public support for policy change ultimately led to the development of strategies to dramatically reduce emissions, and hence acid deposition in the Northeastern United States (Driscoll et al., 2001). Figure 2.3 illustrates the dramatic success of this legislation in leading to significant decreases in the acidity of precipitation over the eastern United States.

The success in reducing these emissions was largely due to improvements in environmental technology that emerged in response to the Clean Air Act and its amendments. As one example, between the late 1880s and the late 1960s the number of patents filed for flue gas desulfurization systems to limit emissions of SO_2 from coal-fired power plants, did not exceed four per year; after passage of the Clean Air Act amendment of 1970 the number of patents filed annually was substantially greater (Taylor et al., 2003). Furthermore, since 1970 capital costs for new systems and operating costs decreased significantly (Taylor et al., 2003), highlighting the potential for environmental regulation to drive technological innovation.

Addressing Upper Atmosphere Ozone Depletion

Absorption of solar radiation by O_3 in the stratosphere (the region of the atmosphere 10–-50 km above the Earth's surface) makes life on Earth possible because it reduces the flux of harmful ultraviolet radiation to the surface (NRC, 1979, 1982, 1984b). Decreases in this O_3 ultraviolet filter lead to increases in skin cancer, as well as ecosystem and climate effects such as reduced productivity, altered reproduction and development, and increased mutations (Hader et al., 2007; van Dijk et al., 2013; WMO, 2014). Sydney Chapman described the natural cycle in

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[5] Cross-State Air Pollution Rule: https://www.epa.gov/airmarkets/interstate-air-pollution-transport.

the upper atmosphere that involves oxygen molecules, oxygen atoms and ozone. Known as the "Chapman Cycle," this reaction sequence generates a steady-state concentration of O_3 that provides the filter for ultraviolet radiation from the sun. The recognition that anthropogenic emissions could alter the balance of this cycle came in the early 1970s when Paul Crutzen (see Box 2.2) predicted that oxides of nitrogen could alter the Chapman Cycle (Crutzen, 1970, 1971) via photolysis of nitrous oxide that had been proposed to be transported from the troposphere (Bates and Hays, 1967). In 1971, Harold Johnston (see Box 2.1) predicted that direct emissions of NO into the stratosphere by a proposed fleet of supersonic transports would also lead to significant ozone loss (Johnston, 1971).

Use of chlorofluorocarbons (CFCs) expanded greatly after World War II in a number of applications such as propellants in spray cans and as refrigerants and blowing agents. Because they do not have carbon-hydrogen bonds with which the hydroxyl radical (OH) can react, they are inert in the lower atmosphere and do not contribute to photochemical air pollution. However, their long lifetimes in the lower atmosphere mean that they can reach the stratosphere where they absorb short-wavelength ultraviolet radiation, generating highly reactive chlorine atoms. Stolarski and Cicerone (1974) proposed that if there were a source of chlorine atoms in the stratosphere, chain destruction of ozone could occur in a manner qualitatively similar to that by oxides of nitrogen, but at the time there was no obvious source of chlorine to the upper atmosphere. Inspired by the detection and measurement of a global distribution of CFCs in the troposphere by James Lovelock and coworkers (Lovelock et al., 1973), Mario Molina and Sherwood Rowland probed the potential atmospheric fates of CFCs and showed that they could indeed be a major source of stratospheric chlorine, perturbing the Chapman Cycle and leading to stratospheric O_3 loss (see Box 2.2).

Measurement of CFCs also enabled new developments in predictive modeling tools, as Lovelock recognized with his seminal 1971 paper (Lovelock, 1971). Tropospheric measurements of semi-inert tracers like $CFCl_3$ and radiogenic gases such as ^{222}Rn and ^{85}Kr, provided fundamental tests for the development of 3-D chemistry transport models (Jacob et al., 1987; Prather et al., 1987) and became a community standard for testing chemistry-transport modeling (Jacob et al., 1997; Pyle and Prather, 1996).

While initial doubt existed regarding the predictions of the atmospheric effects of CFCs, the loss of ozone in the Antarctic "ozone hole" discovered in the mid-1980s was clear and much more dramatic than expected based on known chemistry at the time. Confirmation of stratospheric ozone depletion by as much as 10-35 percent was first noted by Shigeru Chubachi, Joseph Farman, Brian Gardiner, and Jonathan Shanklin (Chubachi, 1985; Farman et al., 1985), and corroborated by satellite measurements (Krueger et al., 1987; Stolarski et al., 1986). These findings motivated Antarctic expeditions by a burgeoning community of scientists charged with discovering the causes and mechanisms of ozone hole formation. The fact that the ozone loss was severe over Antarctica and localized in the lower stratosphere near the ozone concentration maximum was an unforeseen phenomenon. Given what was known at the time, it was expected that CFC-induced ozone loss would be greatest at high altitudes in the tropical and mid-latitude stratosphere. Researchers used a combination of laboratory and theory studies, field observations, and predictive modeling to show that reactions of gases on polar stratospheric cloud particles activated temporary reservoirs for chlorine, which had originated in the stratospheric breakdown of CFCs (Anderson et al., 1991; Crutzen and Arnold, 1986; McElroy et al., 1986; Solomon et al., 1986; see Box 2.2). The integration of laboratory studies, theory, models, and field observations built a detailed understanding of the phenomena responsible for

BOX 2.2

Nobel Prize winners: Sherwood Rowland, Mario Molina, and Paul Crutzen

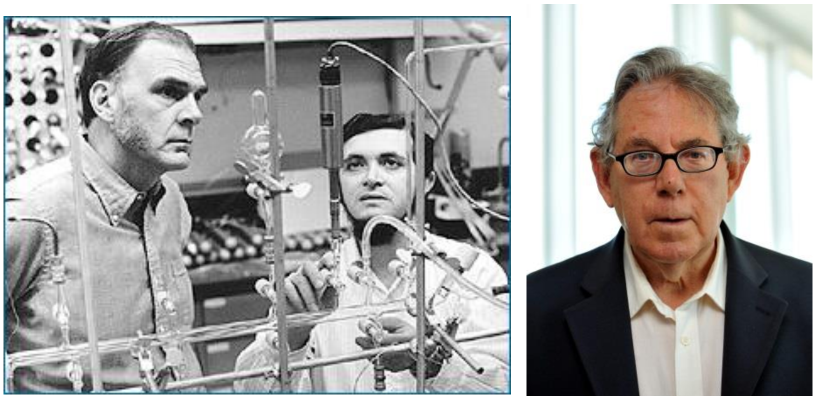

FIGURE B Sherwood Rowland (left), Mario Molina (center), and Paul Crutzen (right) laid the groundwork towards understanding ozone depletion in the stratosphere. SOURCES: (left) Sciencephoto (http://www.sciencephoto.com/media/445822/view), (right) Wikimedia Commons (http://www.mpic.de/en/service/picture-gallery.html).

Early laboratory studies by Paul Crutzen, as well as Sherwood Rowland and Mario Molina (see Figure), were central in identifying causes of, and ultimately curbing, depletion of stratospheric O_3. These scientists identified the primary mechanisms of damage to the upper atmosphere caused by chlorofluorocarbons (CFCs) and oxides of nitrogen, and predicted the environmental consequences of continuing “business as usual.” This basic research on a rapidly changing issue (increased emissions of CFCs) led to a breakthrough in understanding of a major environmental issue. Their translation of the implications of the science for public policy formed the basis for the ban on the use of CFCs in spray cans in the United States in 1978 and later, in 1987, the first international agreement (known as the Montreal Protocol) to phase out substances that deplete the ozone layer. The Montreal Protocol has since been amended a number of times as new scientific information became available. The three scientists were awarded the Nobel Prize in chemistry in 1995 in recognition of their contributions to atmospheric chemistry and the resulting understanding of the consequences for human health and welfare.

stratospheric ozone depletion, including the unique chemical processes that led to formation of the Antarctic ozone hole (Cicerone et al., 1983; Molina and Molina, 1987; Prather et al., 1984). This defining decade of atmospheric chemistry research demonstrated unequivocally that CFCs were the cause of ozone depletion.

The overwhelming evidence for the role of CFCs as the source of chlorine that caused stratospheric ozone depletion led to enhanced public awareness and demand for action. In 1987, an international agreement resulted in the Montreal Protocol[6] treaty and its amendments, which phased out the production of CFCs and other ozone depleting substances. Atmospheric chemists played a key role in providing information to help develop the initial Montreal Protocol, and the information leading to its amendments that broadened the suite of ozone depleting substances included and accelerated the timeline for their phase-out. Figure 2.4 shows that the previously increasing trends in atmospheric concentrations of CFCs reversed in the period 1994–2002 in response to these international controls. By decreasing atmospheric CFC concentrations, this

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[6] The Montreal Protocol on Substances that Deplete the Ozone Layer: http://ozone.unep.org/en/treaties-and-decisions/montreal-protocol-substances-deplete-ozone-layer.

science-based treaty is projected to have averted a number of adverse effects, including millions of skin cancer deaths (van Dijk et al., 2013) and the Antarctic ozone hole has been shown to be healing (Solomon et al., 2016). Emission controls have also curbed the contribution of CFCs to global climate change, since they are also greenhouse gases (Hansen et al., 1989; IPCC, 2014).

However, even though it is on a positive path towards healing, the Antarctic ozone hole is expected to continue to have an impact on regional climate (e.g., on temperature, winds, and precipitation) in the Southern Hemisphere for some time (WMO, 2014). Atmospheric chemists will continue to play an important role in understanding stratospheric chemistry and how stratospheric ozone is changing in response to changes in emissions as well as changes in atmospheric temperature and structure (see Figure 2.5), in particular as the climate continues to change. This ongoing work is needed to monitor these changes and predict future changes with implications for human health and welfare.

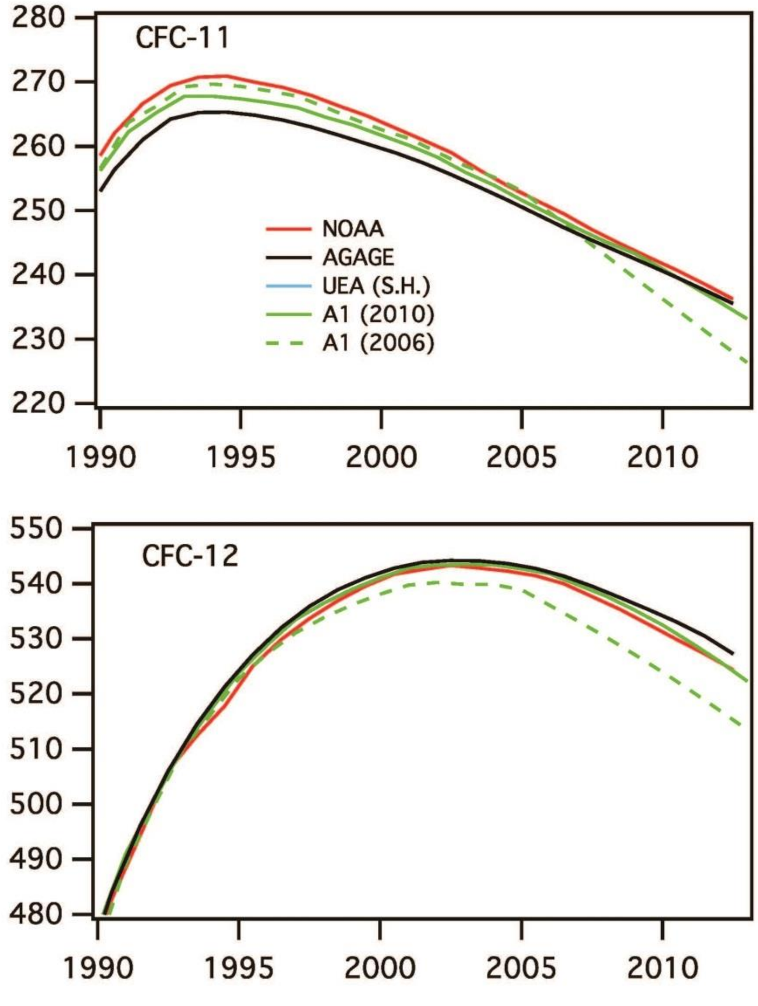

FIGURE 2.4 Trends in mean global concentration (in parts per trillion) of two forms of chlorofluorocarbons containing different numbers of carbon atoms (CFC-11 and CFC-12). These graphs show past increase in ozone depleting substances as well as the dramatic impact of the Montreal Protocol to curb production and release of CFCs, which are now declining in concentration. SOURCE: WMO, 2014.

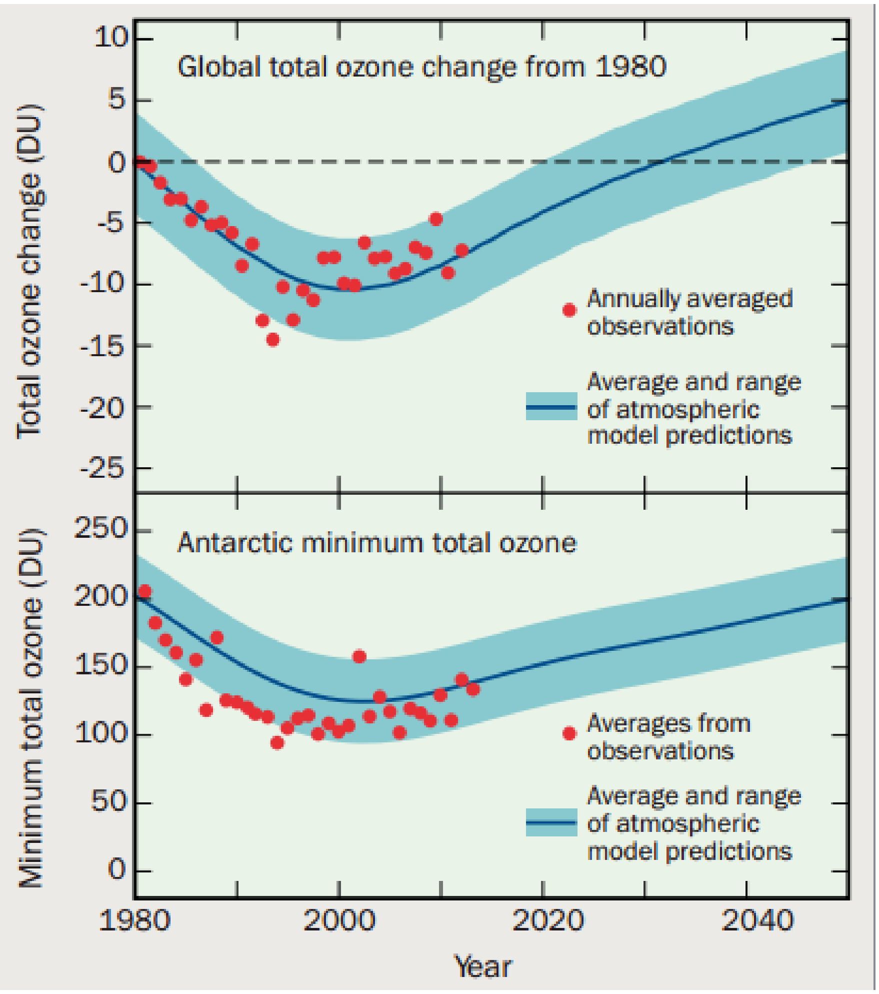

FIGURE 2.5 Simulations of stratospheric ozone depletion resulting from chemistry-climate models (blue shading indicates their range of projections) compared with observations of global total ozone (red dots in the top panel) and minimum total ozone values over Antarctica (red dots in the bottom panel) in Dobson units (1 DU = 0.01 mm thickness at standard temperature and pressure). This figure shows the effect of changes in atmospheric CFC concentrations (as depicted in Figure 2.4) on stratospheric ozone. SOURCE: Hegglin et al., 2015.

2.2 CONCLUSION

As discussed in the three case studies presented in this chapter, the results of atmospheric chemistry research have historically provided a strong foundation of scientific understanding of issues that affect human and global welfare. This understanding has enabled the identification of the root causes to major problems and the development of predictive capabilities that informed scientifically based control strategies. Research continues to inform ongoing assessment of environmental issues as they affect the United States, as well as in developing countries where emission rates of many pollutants are increasing rapidly.

As will be discussed in the chapters that follow, as the atmosphere undergoes changes at an unprecedented rate due to human activities, emerging problems have the potential to significantly affect the health of humans and ecosystems, as well as climate and weather on many scales. These changes will undoubtedly include those that are unforeseen and/or happen abruptly (NRC, 2013), similar to the emergence of the Antarctic ozone hole. The ability to stay ahead of rapidly evolving environmental issues will require having a range of the most advanced tools possible in order to be ready to respond to the next event, be it a volcanic eruption, an oil spill, or emissions of methane from destabilizing permafrost.

Some of the problems that have been successfully addressed in the United States are being repeated in other countries around the world as economic growth outstrips the implementation of regulations. Given the importance of long range transport and of global consequences of emissions, what happens in those countries can have substantial impacts in other regions around the world. It is therefore in our national interest to advance the understanding of atmospheric chemistry to provide a sound basis for the global development and implementation of rational, scientifically based strategies to minimize the impacts of these challenges.

Chapter 3—Understanding Today

Research in atmospheric chemistry is a basic, discovery-driven field of science that is needed to understand the world in which we live. This fundamental research is also at the core of a number of societally relevant issues such as climate and weather, and the health of humans and ecosystems. The Committee identified six core areas that encapsulate the chemical nature of the atmosphere and provide the building blocks for breakthroughs in the future.

- *Human activities and natural processes govern emissions that determine the chemical composition of the atmosphere*. Because atmospheric composition responds to changes in emissions as a result of societal choices and a changing Earth system, the development of accurate emission inventories is a necessary foundation for understanding and predicting changes in the atmosphere.
- *Chemical transformations affect the spatial and temporal variability of gases and particles*. The fundamental study of reactions that occur in the atmosphere is clearly central for predicting the distribution and nature of constituents in the air.
- *Atmospheric oxidants control the lifetimes, distribution, and products of emitted species*. Highly reactive chemicals like ozone and the hydroxyl radical participate in many chemical reactions, so their sources, sinks, and concentrations, as well as their spatial and temporal variability, are required to understand reactions that drive atmospheric chemistry.
- *Chemical processes interact with atmospheric dynamics to control the distribution of trace gases and particles*. Motions of the atmosphere, not just emissions and reactions, affect atmospheric composition, and the understanding of both must be coupled to evaluate distributions.
- *Particle chemical and physical properties affect cloud and aerosol particle radiative properties, cloud microphysics, and precipitation processes*. The distribution and nature of particles in the atmosphere alters the climate as well as behavior of the water (including precipitation) in the Earth system.
- *Atmospheric trace gases and aerosol particles impact and alter global biogeochemical cycles*. An understanding of atmospheric chemical composition is required to evaluate the present and future health of natural and managed ecosystems, marine and other aquatic environments.

The first topic, emissions, highlights the substantial influence of humans on the atmosphere. Both human emissions and changes in the natural system are accelerating; a predictive capability that provides foresight into the consequences of those rapid changes will aid in preparation and decision making. The second through fourth topics—chemical transformations, oxidant distributions, and relationship with atmospheric dynamics—are fundamental elements of the predictive capability required to determine the distribution, transport, and fate of chemicals throughout the atmosphere. These processes determine how emissions and other human activity lead to societal impacts. The fifth topic connects atmospheric chemistry to other physical systems that affect humans, while the sixth connects the atmosphere to biogeochemical cycles affecting ecosystems and food production.

Support of research in these core topics remains vital for both atmospheric chemistry itself and for understanding its connections to human health and welfare as discussed in the next chapter. The remainder of this chapter provides an overview of each of the six topics described above, followed by examples of recent and ongoing advances within the past decade. These recent advances are described using language common in the atmospheric chemistry field to capture the nature and complexity of topics, rather than directing the presentation to the non-expert in the field, as we have attempted to do throughout most of the report.

3.1 HUMAN ACTIVITIES AND NATURAL PROCESSES GOVERN EMISSIONS THAT DETERMINE THE CHEMICAL COMPOSITION OF THE ATMOSPHERE

Overview

An overarching goal of atmospheric chemistry research is to understand human-induced changes in the atmosphere so that accurate predictive capabilities can be developed to assess future scenarios and aid in the development of effective policies to minimize risk. This goal requires accurate data on the emissions to the atmosphere, including gases and particles from all sources, anthropogenic as well as natural. Although substantial improvements have been made in recent years, the current knowledge of emissions into the atmosphere in both the pre- and post-industrial eras is inadequate.

The amount, location, and timing of emissions to the atmosphere are controlled by both environmental conditions and human activities. Global change and economic development alter these emissions, and the economic growth of human society has changed the distribution of both natural and anthropogenic sources. The development of inventories that track emissions from their sources is a scientific endeavor that contributes to the entire discipline of atmospheric chemistry and should be managed in parallel with other core research areas. Emissions link human and natural activity to changes in Earth system behavior, and are required for traceable, attributable, and quantified assessments of how global change and social choices will affect the atmosphere. Improved understanding of the magnitude and the location of emission sources, as well as connections with the technological advances, social decisions, and environmental conditions that determine emissions, is crucial for predicting climate and weather and mitigating health impacts.

Recent Advances

The composition of the atmosphere and the controlling chemistry derive from emissions of gases and particles. Emissions include a very large suite of organic and inorganic compounds, including trace metals such as mercury. Quantification of these emissions in an inventory provides data needed for a multitude of atmospheric chemistry applications, such as inputs to chemistry and climate models, evaluation of trends, and explanation of in situ observations. Substantial advances have been made in the development of emission inventories via techniques that combine a variety of scientific approaches that may include laboratory, in situ and remotely sensed observations, assessment of technology and energy use, and evaluation of human behavior.

Integrated Approaches

There are several approaches to build, interrogate, and evaluate emission estimates. "Bottom-up" approaches typically apply combined knowledge of emission rates and the activities that contribute to them. "Top-down" approaches use models of the atmosphere to infer emissions from measured concentrations of atmospheric constituents. Improvements to quantifying emission data require the integration of these different approaches. Analysis of remote sensing data of the atmosphere near urban centers has produced a compelling constraint on emissions (e.g., Berezin et al., 2013; Hilboll et al., 2013; Konovalov et al., 2008; Pfister et al., 2005). For example, remote sensing of tropospheric NO_2 from space-based satellite instruments has revealed a dramatic change in the global distribution of anthropogenic emissions over the past several decades. Emissions strongly increased over China, the Middle East, and India, while simultaneously declining throughout the more developed world (including the United States, Western Europe, and Japan) (Hilboll et al., 2013). Using a variety of observations and chemical modeling tools, "bottom-up" emission inventories and emission trends can be more thoroughly constrained (e.g., Brioude et al., 2013; McDonald et al., 2012; Palmer, 2008; Polson et al., 2011). Novel detection methods and tracer techniques have revealed gaps in inventories for emissions and their variability from specific sectors (e.g., Mellqvist et al., 2010; Pétron et al., 2012; Roscioli et al., 2015). Addressing these gaps results in better predictions, for example, linking precursor emissions with impacts on ozone (Ahmadov et al., 2015). Other observational advances, such as improved direct flux estimates from high time resolution measurements from the surface (e.g., Karl et al., 2001) and aircraft (Karl et al., 2009; Warneke et al., 2010), have provided the means to quantify emissions over local and regional scales. These various techniques not only enable the quantification of species emitted to the atmosphere, but also evaluate the conditions that control the emissions. It is important to continue application and development of new approaches to verify and predict emissions and to evaluate their impact on atmospheric chemistry and climate (Bond et al., 2013; Tong et al., 2012). Some world regions are undergoing rapid growth or transition, requiring dynamic methods of emission estimation that can rapidly respond to changes in circumstances.

As emission inventories are developed, key scientific questions are generated through their subsequent use. An unanticipated disagreement between atmospheric measurements and an emission inventory is usually an indication of a lack of sufficient understanding of sources and emissions (see Box 3.1). With additional observations and study, causes of gaps and errors in emission estimates can be identified and improved in subsequent inventories. A recent example involves long-term measurements of ethane at sites in the northern hemisphere (see Figure 3.1), which show increases in ambient concentrations. This increase was not predicted by models until the emission inventories for ethane were revised to include those from oil and gas extraction and production activities in North America.[7] Continued analysis of these datasets up to 2015 suggests increased and previously unrecognized emissions of ethane and propane from natural gas development in North America are important (Helmig et al., 2016).

Recent discoveries of high wintertime ozone concentrations in rural areas of the United States in Wyoming (Rappengluck et al., 2014; Schnell et al., 2009) and Utah (Warneke et al., 2014) are other examples of unanticipated disagreements between measurements and emission inventories. Traditionally, high O_3 concentrations are observed during summertime in urban and suburban areas with high motor vehicle, residential, and industrial emissions; this paradigm has

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[7] Ethane emission inventory: https://nar.ucar.edu/2015/acom/d2-ethane-emissions-inventory.

BOX 3.1
Atmospheric Chemistry in the News: Volkswagen Emissions Scandal

In 2015 the public learned of research suggesting that automaker Volkswagen' installed devices that could override emissions controls technology in more than 11 million diesel vehicles sold worldwide. Research revealed that programming in certain models had been set to activate nitrogen oxide (NO_x) emission controls only during laboratory emission testing (Franco et al., 2014). As a result, certain models of on-road vehicles may be emitting between 10 and 40 times more NO_x than anticipated. Estimates are that exposure to excess pollution from diesel models sold in Europe could be responsible for thousands of premature deaths per year (Knapton, 2015; Mathiesen and Neslen, 2015), particularly in Europe. Though relatively few of these diesel vehicles were sold in the United States, researchers estimated excess NO_x emissions in the United States and calculated that such excess emissions over the sales period 2008–2015 could cause 60 premature deaths have been attributed to pollution exposure from these excess vehicle emissions in the United States (Barrett et al., 2015). The revelation of "defeat devices" in the Volkswagen vehicles confirmed what a number of atmospheric chemists in Europe had been reporting—that NO_x concentrations appeared far higher industry-wide than expected given estimated emissions (Schiermeier, 2015[8]). Volkswagen is the subject of regulatory investigations in multiple countries and has announced plans to spend billions of dollars to rectify this issue. This incident has sparked awareness in the general public about the public health risks associated with vehicular air pollution.

subsequently guided ozone measurement and management strategies. Atmospheric models do not predict winter peak ozone episodes in Western rural areas, which is now attributed in part due to the rapid development of oil and gas activities and the inability of bottom-up emission inventories to sufficiently characterize the rapidly changing emissions from this sector (Ahmadov et al., 2015).

Emissions from Specific Sources

The relative magnitude of emissions from biogenic or anthropogenic sources can play a key role in atmospheric chemistry and the chemical pathway of emissions. Biogenic and anthropogenic emissions typically have fundamentally different governing processes, temporal scales, and spatial scales that require their own inventory methodologies. Additionally, particle phase emissions represent a unique challenge as they can be emitted directly or formed from secondary processes in the atmosphere.

Vegetation is the dominant source of reactive hydrocarbons to the atmosphere (Guenther et al., 2006) in the form of volatile organic compounds (VOCs) such as isoprene and monoterpenes (Guenther et al. 1996). Model descriptions of the emissions of biogenic VOCs have improved considerably over the past decade (e.g., Arneth et al., 2007; Guenther et al., 2012) and are included in most regional and global models. At the same time new measurements, including for example flux measurements from aircraft (Karl et al., 2013) and over the open ocean (e.g., Kim et al., 2014; Marandino et al., 2007), are providing critically needed observational constraints on these emissions. These constraints are vital to quantifying the gas-phase emissions that drive atmospheric chemistry processes in many regions of the globe.

In urban areas, anthropogenic emissions drive the chemistry and resulting impacts, such as ozone and particulate pollution. In former decades, ozone mitigation strategies have been

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[8] Also see http://www.bbc.com/news/business-34333216.

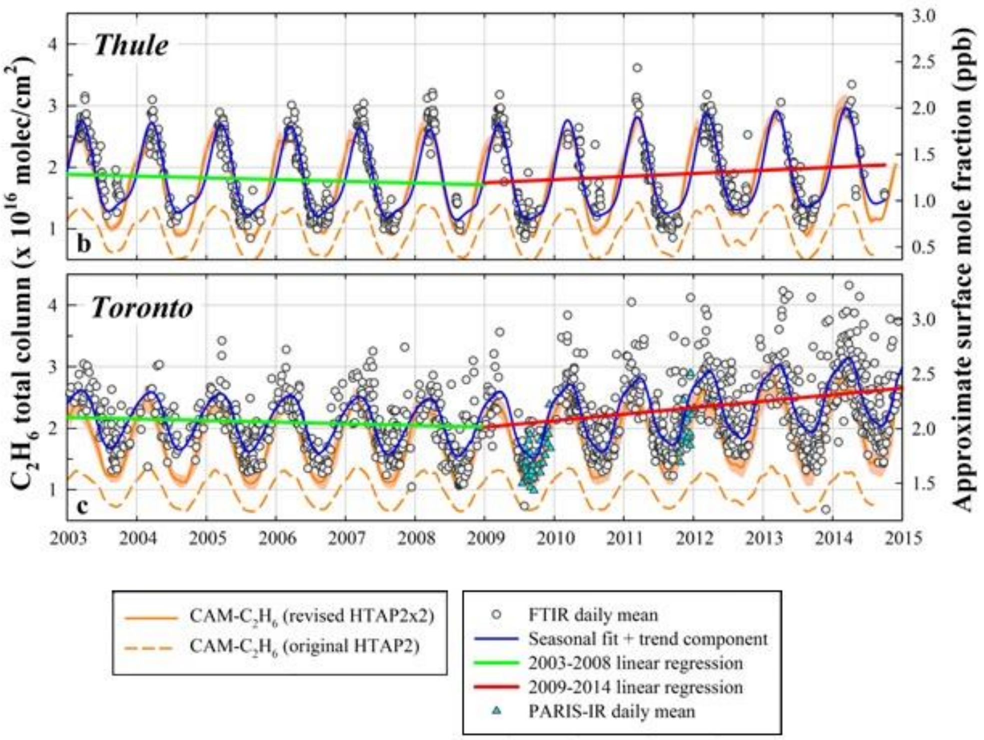

FIGURE 3.1 Measurements of ethane from multiple sites in the northern hemisphere have suggested an increase in concentrations of this long-lived compound in the last 6 years (Thule and Toronto shown here). Models can replicate the measurements only after the emission inventories are revised and ethane emissions, predominantly from oil and gas extraction and production activities in North America, are increased. Graph displays daily mean ethane total columns derived from the FTIR (gray circles) and PARIS-IR (light blue triangles) observations performed between January 2003 and December 2014. The right *y*-axis scale converts the total columns into approximate surface mole fraction. The blue curve visualizes the function (including seasonal modulation and trend component) fitted to all daily FTIR means over the periods 2003–2008 and 2009–2014, using a bootstrap tool. The green and red lines are the associated linear regressions (as solid line for FTIR and dashed line for PARIS-IR). The dashed and solid orange curves are the monthly mean ethane total columns simulated by a model (CAM), implementing the original HTAP2 and revised HTAP2x2 (with increasing North American emissions) inventories, respectively. The shaded area corresponds to the 1σ standard deviation. SOURCE: Franco et al., 2016.

informed and validated by combined model/measurement programs. In more recent decades, the field of atmospheric chemistry has been tasked with gaining insight into the difficult problem of urban and anthropogenic aerosol particles (Heald et al., 2005; Odum et al., 1997; Volkamer et al., 2006). Because particulate matter is frequently formed by processes occurring in the atmosphere, it is a dynamic component that does not lend itself to conventional inventory development approaches (Donahue et al., 2006; Zhang et al., 2015a). Current and future research examining urban outflow will expand the understanding of the mechanisms and impacts of anthropogenic and biogenic emissions as they are processed in the atmosphere and contribute to particle mass (Camredon et al., 2007; Gentner et al., 2012; Huang et al., 2014). Constraints on the magnitude of each source are vital to this research.

The atmosphere has been a known habitat for microbes for centuries (Womack et al., 2010). Aerobiology studies focusing on determining the concentrations of microbes in the atmosphere remain disproportionally low relative to water and soil (Barberán et al., 2015;

Behzad et al., 2015; Kellogg and Griffin, 2006; Womack et al., 2010). The understanding of the sources and impacts of bioparticles has increased in recent years but the overall understanding of their impacts on atmospheric composition, climate, and human health remains weak. The first attempts to model emissions and transport of fungal spores, bacteria, and pollen (Burrows et al., 2009b; Heald and Spracklen, 2009; Jacobson and Streets, 2009; Mahowald et al., 2005) are now starting to be tested against new observational constraints from fluorescence measurements (Gabey et al., 2010; Huffman et al., 2010; Perring et al., 2015). The oceans have become recognized as a significant source of not only sea salt but also surface active biological species including bacteria, lipids, proteins, sugars, and viruses. When waves break, bubbles burst at the surface ocean releasing sea spray aerosols which can be heavily enriched in biogenic material (Blanchard, 1964; O'Dowd et al., 2004). Because the biota concentrates important nutrients, phosphorus can be a good indicator of primary biogenic particles (Mahowald et al., 2008b). Studies have also explored the ability of bioparticles from different sources to act as cloud condensation nuclei or ice nuclei (DeMott et al., 2016; Steiner et al., 2015; Tobo et al., 2013), the importance of which is discussed in Section 3.4.

Biomass burning is another activity that produces substantial amounts of trace gases and particulate matter to the atmosphere. Estimation methods for biomass burning emissions (e.g., Urbanski et al., 2011; van der Werf et al., 2006; Wiedinmyer et al., 2011) have taken advantage of controlled laboratory experiments (e.g., Stockwell et al., 2015) and ambient observations (e.g., Burling et al., 2012; Christian et al., 2007) to constrain the particle and gas emissions from this important source on time and spatial scales relevant for local, regional and global applications, although uncertainties in these estimates remain high (e.g., Al-Saadi et al., 2008; Urbanski et al., 2011). Remote sensing observations of land surface characteristics (e.g., Friedl et al., 2002) and burning activities (e.g., Giglio et al., 2006; Wooster et al., 2003) have enabled further constraints on biomass burning emission estimates. Large-scale open biomass burning, including wildfires (see Figure 3.2), emit close to half of the global particulate emissions and are an important contributor to global trace gas emissions (c.f., EDGAR database[9]).

In short, quantitatively defining the composition and sources of all emissions to the atmosphere requires taking both natural and anthropogenic sources into account, while using many different observational and modeling tools to constrain the estimates and to provide realistic uncertainties on those estimates. Such data need to be provided on spatial and temporal scales appropriate for each type of source and need to interface with scales used in models and measurements.

3.2. CHEMICAL TRANSFORMATIONS AFFECT THE SPATIAL AND TEMPORAL VARIABILITY OF GASES AND PARTICLES

Overview

Most emitted gases and particles can further react to form new chemical species. Many compounds that are unreactive in the lower atmosphere, such as chlorofluorocarbons (CFCs), are transported to the stratosphere where they break down, leading to dramatic impacts on natural

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[9] http://edgar.jrc.ec.europa.eu/overview.php?v=42.

FIGURE 3.2 Wildfires are an important source of emissions to the atmosphere, for example, this photo of smoke from wildfires burning in the Angeles National Forest filling the sky behind the Los Angeles skyline on Monday, June 20, 2016. SOURCE: Associated Press.

ozone cycles (Burkholder et al., 2015; Crutzen and Lelieveld, 2001; Finlayson-Pitts and Pitts, 2000; Rowland, 2001, 2006). Transformations are at the core of atmospheric chemistry.

Figure 3.3 provides an overview of the gas-phase chemistry of organic compounds, oxides of nitrogen and sulfur that alter the chemical composition of directly emitted species to form a variety of products in both the gas and condensed phases (Atkinson and Arey, 2003; Finlayson-Pitts and Pitts, 2000; Seinfeld and Pandis, 2006). The gas-phase inorganic chemistry of oxides of nitrogen and sulfur are relatively well known, while their interactions with organic compounds—for example to form organosulfates—are less well understood. Oxidation of organic compounds generally produces compounds that are more polar and less volatile (Goldstein and Galbally, 2007). Products with different vapor pressures and solubility behaviors, combined with direct emissions of VOCs of varying molecular mass, lead to a continuum of organic species that have been designated intermediate, semi-, low, and extremely low volatility organic compounds (Donahue et al., 2012). Such products are major components of secondary organic aerosol particles (SOAs) and are likely involved in the earliest stages of particle nucleation and growth (Bianchi et al., 2016; Ehn et al., 2014; Kulmala et al., 2013; Riipinen et al., 2012; Tröstl et al., 2016; Zhang et al., 2011). The condensed phase of particles, fogs, and clouds provides the opportunity for species to partition between phases, to deposit into rivers, lakes, and oceans. Atmospheric transformations of particles containing micronutrients, such as iron, are likely to be of great importance to both cycling of sulfur and organic acids as well as iron bioavailability (Meskhidze et al., 2005).

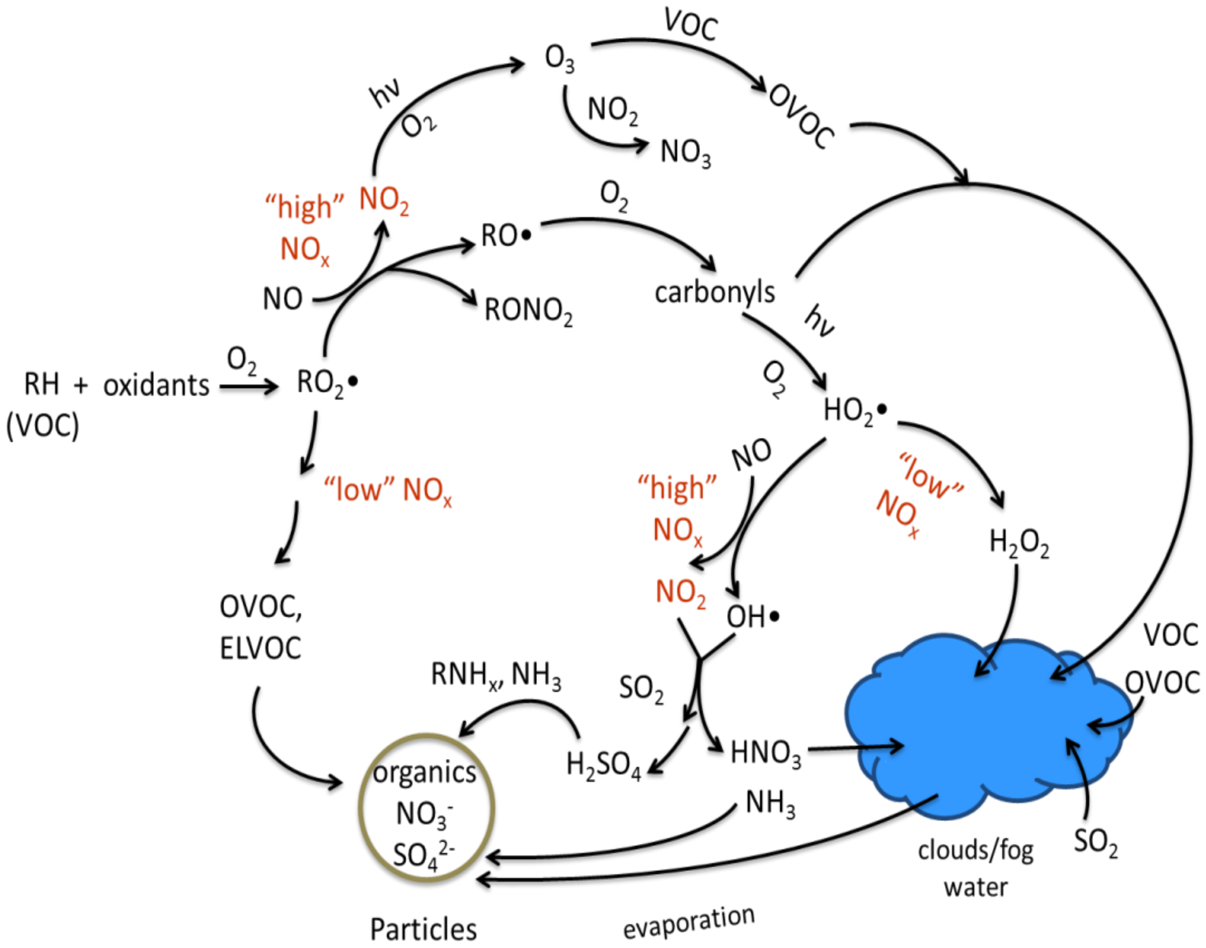

FIGURE 3.3 Overview of some important cycles in the troposphere. Nitric oxide (NO), the major nitrogen oxide species emitted during combustion, is converted to nitrogen dioxide (NO_2) in air and ultimately forms ozone, nitric acid, and organic nitrates (Finlayson-Pitts and Pitts, 2000; Pusede et al., 2015; Seinfeld and Pandis, 2006). Sulfur dioxide emitted during combustion of sulfur-containing fossil fuels is converted by oxidation in both the gas and condensed phases to form sulfuric acid as well as organosulfates. Nitric and sulfuric acids react in air with ammonia and amines to form particles. Oxidants include OH, O_3, NO_3, and halogen atoms.

Recent Advances

Many important advances over the last few decades have contributed to the understanding of the atmospheric transformations of chemical constituents. Following are a few examples whose importance for understanding atmospheric processes and impacts has recently been recognized, but currently lack solid understanding.

Kinetics, Mechanisms, and Products of Gas-Phase Reactions

Rate constants for the gas-phase reactions of organic compounds with OH, O_3, NO_3, and Cl, and their dependence on temperature and pressure have been measured for many reactions (IUPAC, 2015; Sander et al., 2011), and structure-reactivity relationships have been developed for the initial oxidation steps of relatively small VOCs (Calvert et al., 2015). Mechanistic understanding has also substantially improved (Calvert et al., 2015), an example being the isomerization of some organic peroxyl radicals to form products with high oxygen to carbon (O:C) ratios and low volatility.

Advances in theoretical chemistry have provided an important tool to understand and predict both kinetics and reaction pathways in increasingly complex systems that may not be possible to study experimentally at present (Vereecken et al., 2015). For example, autooxidation reactions are well known in the condensed phase (Sevanian et al., 1979), but were highlighted in theoretical studies as also being possible in the gas phase (Sevanian et al., 1979; Vereecken et al., 2007), which was subsequently confirmed in environmental chamber studies of isoprene oxidation products (Crounse et al., 2011; Krechmer et al., 2015).

Many gas-phase smog chamber reaction studies have been carried out in the absence of water vapor or under low humidity conditions. There is increasing recognition of the role of water vapor in altering reaction pathways of some organic compounds, particularly those that are highly soluble in water such as glyoxal, methylglyoxal, and organic acids. For example, one study reported the observation of glyoxylic acid and its germinal diol counterpart in the gas-phase (Plath et al., 2009), illustrating a gas-phase pathway to convert aldehyde into acid and aerosol particles.

The Importance of Phase and Partitioning

The importance of exchange between the gas and particle phases has become abundantly clear for understanding the composition of the atmosphere. While the phase behavior of inorganic salts in pure form as a function of relative humidity (RH) is well known, that of SOA is not. Models have historically treated SOA as an oily material, where internal diffusion is sufficiently fast that quasi-equilibrium between the gas-phase and particles holds. However, recent laboratory and field studies have shown that SOA can be a highly viscous semi-solid material (Bateman et al., 2015; Kidd et al., 2014; Perraud et al., 2011; Pöschl and Shiraiwa, 2015; Renbaum-Wolff et al., 2013; Shiraiwa et al., 2011; Virtanen et al., 2010, 2011) in which diffusion in the particles is much slower, and the time scales to reach equilibrium can be longer than represented by quasi-equilibrium in atmospheric models (You et al., 2014). A further recent discovery is that liquid-liquid separation can occur in particles (O’Brien et al., 2015; You et al., 2014), and that this separation and the particle morphology depends on the composition and the relative amounts of the different components. These properties can affect gas uptake and partitioning, gas concentrations, optical properties of the particles (You et al., 2012) and their ability to act as cloud condensation nuclei and ice nuclei (Schill and Tolbert, 2013).

Chemistry and Photochemistry in the Condensed Phase and at Interfaces

The presence of all three phases—gas, liquid, and solid—in the atmosphere dictates that partitioning between the phases and chemical/photochemical processes occurs in all three phases. In addition to multiphase chemistry involving gases and liquids, it is known that unique chemistry can occur at interfaces. Reactions on solids and in surface films are generally described in the community as "heterogeneous chemistry."

Fogs and clouds present a common aqueous medium for reactions in the troposphere, and water and organic species can also be an important component of particles. While oxidation of dissolved SO_2 (in all of its forms collectively referred to as S(IV)) in the aqueous phase to sulfuric acid and sulfates is well known, recent studies show that multiphase chemistry involving water may be important in atmospheric transformation for a variety of species (Biswas et al., 2008; Davidovits et al., 2011; Ervens, 2015; Herrmann et al., 2015; McNeill, 2015; Zhang et al., 2015b; Zhu et al., 2005). This mechanism has been shown to influence the chemical signature of trace constituents, such as formation of SOA from species vented from the boundary layer to the free troposphere (e.g., Froyd et al., 2007; 2010; Sorooshian et al., 2006). These precursors generate particles aloft, which have large radiative impacts because they scatter and absorb incoming solar energy as well as diffuse backscattered radiation from clouds below (Seinfeld, 2008). Ubiquitous and abundant small, water-soluble compounds from gas-phase oxidation of biogenic and anthropogenic emissions can undergo oxidation in the aqueous phase to form SOA (Herrmann et al., 2015; Zhang et al., 2015a; Zhu et al., 2005). Reactions with inorganic constituents in wet aerosol particles (Noziere et al., 2009; Perri et al., 2010; Yasmeen et al., 2010), and with oxidants (e.g., ozone [Grgic, 2010]) also contribute to this aqueous phase oxidation. Evidence for SOA formation is provided by laboratory experiments demonstrating that the aqueous chemistry of small water-soluble compounds forms organic acids/salts (e.g., oxalate) as well as high molecular weight compounds that are also found in atmospheric aerosol particles.

Multiphase chemistry also plays a role in determining the optical properties of particles, for example the formation of visible light-absorbing products known as "brown carbon" (Laskin et al., 2015; Moise et al., 2015), which adds to particulate mass emitted directly from some combustion processes such as biomass burning (Dutkiewicz et al., 2011; Laskin et al., 2015). However, the nature and number of individual chromophores, their formation mechanisms, and atmospheric fates remain open questions (Phillips and Smith, 2014, 2015).

There has been increasing recognition of the importance of heterogeneous chemical processes, which occur not only on the surfaces of particles (Al-Abadleh and Grassian, 2003), but also on surfaces in the boundary layer, including the built environment, ocean, vegetation, and ice (George et al., 2015). The air/sea interface covers nearly three-quarters of the Earth's surface and can profoundly impact atmospheric composition (Ryder et al., 2015). Reactions at interfaces can modify surface properties, changing their ability to act as cloud condensation and ice nuclei, and altering the bioavailability of the particle constituents to both ecosystems and humans. In addition, reactions on biological surfaces can cause damage to humans and ecosystems (Pöschl and Shiraiwa, 2015). In an effort to study heterogeneous chemistry on more realistic atmospheric surfaces, new flow tube approaches have been developed (Bertram et al., 2009). These studies offer the potential for bridging the existing gap between laboratory studies and field observations.

On a molecular level, the mechanisms and kinetics of chemical and photochemical reactions on surfaces differ from those in the gas-phase or in aqueous solutions. For example, enhanced thermal and photochemical reactions have been observed at the interface compared to the bulk for organic systems (e.g., Donaldson and Valsaraj, 2010; Enami et al., 2015; Griffith and Vaida, 2012; Heath and Valsaraj, 2015; Kameel et al., 2014), for inorganic ions (Knipping et al., 2000; Tobias and Hemminger, 2008) and for nitric acid/nitrate photochemistry on "urban grime," and on other environmental surfaces (Baergen and Donaldson, 2013; Du and Zhu, 2011; George et al., 2015; Zhou et al., 2003; Zhu et al., 2008). Similarly, there is increasing evidence that some organic species in particles and/or on surfaces in the atmosphere act as photosensitizers at interfaces (George et al., 2015), initiating a cascade of unique interfacial chemistry.

Water plays an important role in heterogeneous chemistry by reacting with particles and surfaces or changing their reactivity, affecting particle growth, or competing for surface sites for heterogeneous reactions (Du et al., 2014; Kolb et al., 2010; Rubasinghege and Grassian, 2013). Reactions on ice or snow surfaces play an important role, particularly in the chemistry of the polar regions (Anastasio and Jordan, 2004; Grannas et al., 2002; Hamer et al., 2014; McNeill et al., 2006; O'Driscoll et al., 2008; Thomas et al., 2012; Toom-Sauntry and Barrie, 2002). Gas uptake in/on snow and subsequent chemistry and photochemistry in the snow contribute to changing chemical composition above or in the snowpack, oxidant formation, and ozone depletion, showing the important role of surface atmospheric chemistry within the cryosphere component of the Earth system (Abbatt et al., 2012).

Understanding the Molecular Basis for Particle Nucleation and Growth in the Atmosphere

Sulfuric acid is known to be a significant source of atmospheric particles on a global basis (Kuang et al., 2009; Kulmala et al., 2013; Zhang et al., 2011, 2015a). However, it is common to have orders-of-magnitude differences between measured rates of new particle formation and those predicted using classical nucleation theory. The discoveries that have dramatically altered the understanding of atmospheric nucleation include the following: (1) amines are far more efficient than ammonia in forming particles with sulfuric acid and are ubiquitous in air (Almeida et al., 2013; Ge et al., 2011; Glasoe et al., 2015; Pratt et al., 2009a; Smith et al., 2010); (2) assumptions inherent in classical nucleation theory may not be directly applicable to at least some of the important nucleation processes occurring in the atmosphere (Kupiainen-Maatta et al., 2014); and (3) organic species likely play a key role in nucleation and growth (Bianchi et al., 2016; Bzdek et al., 2013; Donahue et al., 2011; Kulmala et al., 2013; Metzger et al., 2010; Riccobono et al., 2014; Riipinen et al., 2012; Schobesberger et al., 2013; Tröstl et al., 2016; Zhang et al., 2011).

Theoretical approaches, including quantum chemistry, have played a central role in elucidating mechanisms on a molecular level and providing a predictive capability that was previously lacking. The combination of theoretical and experimental approaches have provided new understanding of how small clusters form and grow via stepwise additions of acid, base, and water, and the decomposition mechanisms for the clusters that occur in competition with their growth (Bzdek et al., 2013; DePalma et al., 2014; Olenius et al., 2014; Ortega et al., 2012; Vehkamaki and Riipinen, 2012).

An area in which less progress has been made is in reconciling measurements and models for the formation and growth of the mass of SOA associated with particles. It was almost a decade ago that it became clear that models could not reproduce the SOA particle mass measured in many locations (de Gouw et al., 2005; Heald et al., 2005; Johnson et al., 2006; Volkamer et al., 2006). Better agreement has been attained with improved emission inventories and identification of additional precursors and semi- and intermediate volatility species, as well as updated chemical mechanisms and chemistry in the condensed phase (Hayes et al., 2015; Heald et al., 2011; Shrivastava et al., 2011). However, even when models reproduce measured mass concentrations, more specific properties such as volatility and O:C ratio may not be consistent (Chen et al., 2011). The new ability to measure extremely low volatile organic compounds (Ehn et al., 2014) has highlighted how ubiquitous these compounds are and their likely importance in SOA formation. In addition, as discussed above, multi-phase chemistry and photochemistry is likely to play an important role in SOA formation, particularly in high relative humidity environments.

Ability to Measure Composition of Gases and Particles

A number of techniques, particularly those involving mass spectrometry, have been developed to measure gases at sub-part per trillion levels, particles as small as approximately 8 nm in size, as well as clusters. For example, chemical ionization combined with high resolution time-of-flight mass spectrometry has been applied to measure a variety of both inorganic gases such as nitryl chloride (Simpson et al., 2015), organic species with extremely low vapor pressures (Ehn et al., 2014; Nozière et al., 2015), and molecular clusters (Schobesberger et al., 2013). On-line real-time analysis of particles using laser desorption/ionization or thermal desorption techniques (Bzdek et al., 2012; Canagaratna et al., 2007; Middlebrook et al., 2003; Murphy, 2007; Murphy et al., 2006; Nozière et al., 2015; Prather et al., 2008; Tobias and Ziemann, 2000; Zelenyuk et al., 2009; Zhang et al., 2014) has provided insight into particle composition and how it changes with the nature of precursors and aging in air. The development and widespread use of commercial aerosol mass spectrometry has enabled characterization of particle composition in many locations throughout the world, providing insights into spatial and temporal variations under a number of conditions (Jimenez et al., 2009) and enabled the observation of the oxidation of low-volatility organic compounds to form organic aerosol particles in the lab (Robinson et al., 2007; Sage et al., 2008). Obtaining molecular structures of complex organic species in particles remains challenging using online particle mass spectrometry techniques. However, advances in off-line techniques applied to collected particle samples—such as multi-dimensional and combined chromatography-mass spectrometry (Nozière et al., 2015; Zhang et al., 2014) as well as ambient ionization methods (Ifa et al., 2010) and in situ chromatography with online derivatization (Isaacman et al., 2014) for gas and particle phase species—have improved understanding substantially. Advances in instrumentation have been major contributors to propelling the field forward over the last decade and is likely to continue in the coming years as well.

3.3 ATMOSPHERIC OXIDANTS CONTROL THE LIFETIMES, DISTRIBUTION, AND PRODUCTS OF EMITTED SPECIES

Overview

Trace oxidants are present in extremely low concentrations in the atmosphere, yet they determine the chemical fates of many atmospheric species. These oxidants are highly reactive, with the most reactive oxidants having lifetimes on the order of seconds or less, while tropospheric ozone (O_3) can last for weeks. The hydroxyl radical (OH) is the dominant oxidant in the atmosphere. However, a suite of other gas-phase and aqueous-phase oxidants in the oxygen, hydrogen, organic carbon, nitrogen, and halogen chemical families are important drivers of atmospheric chemical reactions. The hydroperoxyl radical (HO_2) and organic peroxyl radicals are also key players. Another important atmospheric oxidant is the Criegee intermediate, which is formed in the reaction between ozone and organic compounds that have double bonds. Lastly, a suite of oxidants grouped under “reactive oxygen species” (ROS) in the condensed phase are also of importance, especially for their role affecting human health (see Chapter 4.2).

Recent Advances

Over the past few decades, atmospheric chemistry research has transitioned from a dearth to a wealth of oxidant observations of gas-phase OH, HO_2, unspeciated peroxy radicals (Heard and Pilling, 2003; Monks, 2005; Stone et al., 2012), NO_3 (Brown and Stutz, 2012), and halogens (Simpson et al., 2015). Advances in understanding oxidant chemistry in certain environments have generated a good first-order understanding of this chemistry in the stratosphere and free troposphere. In more complex environments—particularly those with more VOCs or heterogeneous reaction pathways that compete with gas-phase reactions, and in the condensed phase—the understanding is less complete.

Stratospheric Ozone Depletion

As described in Chapter 2, one success story in atmospheric chemistry is discovery of the processes that cause stratospheric ozone depletion (Newman et al., 2009). An integral component of this understanding is in stratospheric oxidant chemistry, where observations and models generally agree within uncertainties for odd-hydrogen chemistry (Wennberg et al., 1994) and halogen chemistry (von Hobe et al., 2013; WMO, 2014). However, despite this success, there are still outstanding questions about oxidant sources in the lower stratosphere. For example, overshooting mid-latitude cumulus convection may inject water vapor into the lower stratosphere, setting the stage for enhanced heterogeneous conversion of inorganic chlorine to forms that can catalytically destroy ozone (Anderson et al., 2012). Continued monitoring of stratospheric composition is essential to ensure that the stratospheric halogen oxidant concentrations continue to decrease and that stratospheric ozone recovers (seeFigures 2.5 and 2.6).

Global Atmospheric Gas-Phase Oxidation Potential

A key issue for atmospheric chemistry research is the global atmospheric oxidation capacity, especially associated with OH since it controls the lifetime of methane and other important greenhouse gases. Studies have shown that the interannual variability in global atmospheric OH concentrations is small (<5 percent; Ciais et al., 2013; Montzka et al., 2011). However, the distribution of OH regionally and between hemispheres (Patra et al., 2014) and the abundance and impact of the full range of atmospheric oxidants on atmospheric composition, are still uncertain.

Oxidants seem to be best understood in cleaner environments, such as much of the tropical free troposphere where oxidation chemistry involves primarily OH in the gas-phase and hydrogen peroxide in the aqueous phase. In these regions, measured and modeled OH generally agree (Cantrell et al., 2003; Ren et al., 2008, 2012). However, measured HO_2 is less than modeled in the mid-troposphere (Ren et al., 2008; Tan et al., 2001), perhaps because of heterogeneous reactions of HO_2 on aerosol particles (Mao et al., 2010).

The nitrate radical (NO_3) is an important nighttime oxidant for anthropogenic and biogenic VOCs (Brown and Stutz, 2012). NO_3 is coupled to dinitrogen pentoxide, which is lost to the surface where it can react with sea spray to produce volatile nitryl chloride, a daytime Cl oxidant source (Thornton et al., 2010). Several studies in urban areas and power plant plumes indicate that the chemical processes producing NO_3 are generally understood, but questions remain about the distribution of NO_3 in the free troposphere and its vertical nocturnal distribution in urban areas.

Halogens can also influence oxidant budgets. In the Arctic, bromine drives a catalytic cycle that can lower boundary-layer ozone to near-zero levels (Barrie et al., 1988; Pratt et al., 2013; Simpson et al., 2015). On a global scale, satellite measurements suggest that bromine monoxide (BrO) might be distributed in the troposphere throughout the mid-to-high latitudes, giving it a role in determining global oxidant levels (Wagner et al., 2001). In remote coastal regions, iodine and other emitted biogenic species can play an important role in the local oxidation chemistry and in the formation of particles (Carpenter and Nightingale, 2015; Carpenter et al., 2001). Chlorine has also been postulated to be an important oxidant in the marine boundary layer, making a small contribution to atmospheric oxidation in some urban port cities (Riemer et al., 2008; Young et al., 2014), and according to some recent studies, may be an important oxidant over continents (Mielke et al., 2011; Thornton et al., 2010; Wild et al., 2016).

There is growing evidence that Criegee intermediates (highly reactive and unstable biradicals) may be important atmospheric oxidants. Criegee intermediates were postulated almost 70 years ago (Criegee, 1948) and laboratory studies using indirect methods have inferred their existence and reaction kinetics (Calvert et al., 2015). The Criegee intermediate has recently been directly generated and measured in laboratory systems (Taatjes et al., 2013; Welz et al., 2012), and its existence in the atmosphere has been inferred from measurements of sulfuric acid, an oxidation product of the reaction between SO_2 and Criegee intermediates (Mauldin et al., 2003, 2012). However, the role of the Criegee intermediate as an atmospheric oxidant has not been firmly established (Berndt et al., 2014; Huang et al., 2015; Nguyen et al., 2015b; Osborn and Taatjes, 2015; Taatjes et al., 2014).

Gas-Phase Oxidants in Globally Dispersed Forested Environments

Forests cover about one third of the Earth's land mass and their emissions of reduced gases strongly influence the global atmospheric oxidation potential. The chemistry in these environments is controlled by abundant biogenic VOCs and sparse nitrogen oxides so that the biogenic VOCs determine the destruction and the production of OH and O_3. Measured OH has greatly exceeded modeled OH in studies of several forests (Carslaw et al., 2001; Hofzumahaus et al., 2009; Lelieveld et al., 2008; Ren et al., 2008; Tan et al., 2001; Taraborrelli et al., 2012; Whalley et al., 2011) but not all (McKeen et al., 1997). These higher-than-expected OH abundances in forests have led to the hypothesis that an unknown mechanism recycles HO_2 to OH at a maximum rate without generating O_3 (Rohrer et al., 2014). To explain these OH abundances, the photochemistry of isoprene has been explored in greater detail, and the mechanisms are now much better understood (Crounse et al., 2011, 2012, 2013; Paulot et al., 2009; Peeters and Muller, 2010; Peeters et al., 2009). Confounding this disagreement between measured and modeled OH are the recent discoveries of interferences in the most common detection method for HO_2 (Fuchs et al., 2011; Whalley et al., 2013) and in some (Hens et al., 2016; Mao et al., 2012) but not all (Fuchs et al., 2016) laser-based instruments that measure OH. After these interferences were discovered, interference-free detection methods were devised and are being applied. For some recent field studies, when improved mechanisms are included in the model and interference-free OH measurement techniques are used, the observed OH levels agreed with the modeled OH levels to within the combined uncertainties (Feiner et al., 2016; Hens et al., 2014; Mao et al., 2012). While recent results are encouraging, more work is needed to develop a predictive understanding of the oxidation chemistry in forest atmospheres.

An important constraint on understanding atmospheric oxidation is OH reactivity (defined as the inverse of the OH lifetime [Kovacs and Brune, 2001]). In forests, unidentified OH reactivity is between 30 and 90 percent of the measured total (Di Carlo et al., 2004; Nolscher et al., 2014; Sinha et al., 2008). However, in some cases, the estimated OH reactivity (constrained to measured primary chemical emissions) agrees within 20 percent of the measured values (Mao et al., 2012; Zannoni et al., 2015). Further work is needed to address the lack of consistent agreement between estimated and measured OH reactivity in order to develop a predictive understanding.

Gas-phase Oxidants in Urban Regions

The atmospheric oxidation chemistry of urban areas and regions characterized by copious emissions of VOCs, NO_x, and particles has been studied for decades and is thought to be understood. However, measured ozone generally exceeds modeled concentrations at high ozone abundances (Appel et al., 2007; Im et al., 2015). The agreement between measured and modeled OH and HO_2 in cities is somewhat mixed, with good agreement in some studies and differences of a factor of two in others (Griffith et al., 2016; Stone et al., 2012). Some of this disagreement may be explained by the recently discovered interferences in OH and HO_2 measurements, but persists even when interference-free methods are used (Ren et al., 2013). The measured and calculated OH reactivity only agree sometimes (Chatani et al., 2009; Griffith et al., 2016; Kovacs and Brune, 2001; Ren et al., 2003; Yoshino et al., 2006), suggesting the presence of unknown chemical species in some urban areas.

For ozone production, some studies indicate that the concentration of ozone-producing HO_2 behaves as expected as NO increases (Mihelcic et al., 2003; Thornton et al., 2002); while for others, the measured-to-modeled HO_2 ratio increases as a function of NO, resulting in more ozone production than is predicted by current models (Brune et al., 2016; Emmerson et al., 2005, 2007; Griffith et al., 2016; Kanaya et al., 2007, 2008; Martinez et al., 2003; Ren et al., 2003, 2006). Consistent with this result are direct measurements of ozone production that suggest the calculated rates are too low in the morning when NO_x concentration is the greatest (Cazorla et al., 2012). These findings of greater-than-expected measured HO_2 and measured ozone production versus NO appear to be inconsistent with the weekend/weekday effect, in which more ozone is produced on weekends when NO_x levels are lower (Cleveland et al., 1974; Pollack et al., 2012). Further, many commonly used air quality models under-predict high ozone events and their decrease with NO_x reductions. Possible causes for the discrepancies include the details of the chemical mechanism used, boundary conditions, meteorology, and long range transport (Appel et al., 2007; Gilliland et al., 2008; Im et al., 2015) Thus, the lack of consistent agreement between measured and modeled oxidants suggests an incomplete understanding of urban oxidation chemistry and the presence of other factors that control ozone amounts.

Aqueous Phase Oxidants

Elucidating the role of “reactive oxygen species” (ROS) in atmospheric chemistry and their impacts requires understanding their global abundance and spatial distribution as well as their transformations across a range of different environments. ROS is often used to describe particle components capable of catalytically generating oxidants in the condensed phase both in vivo and in vitro. ROS species include free radicals such as OH, HO_2, superoxide anion, peroxynitrites, H_2O_2, organic peroxides (Poschl and Shiraiwa, 2015), and transition metals (Charrier and Anastasio, 2011). These compounds are found both outdoors and indoors and there is significant recycling among the different oxidants and across the phases. As discussed in Chapter 4.2, ROS are thought to play a key role in inflammation, pulmonary oxidative stress, vascular dysfunction, atherosclerosis and lung cancer, so there are active research programs studying the health effects of ROS. In the atmospheric chemistry community, mechanistic studies of ROS in particles and on surfaces are in their infancy. A variety of analytical techniques have been developed to quantify PM-induced oxidative stress from different perspectives. Some systems mimic antioxidant loss (e.g., dithiothreitol assay; Cho et al., 2005), others measure cellular response such as macrophage ROS generation (Landreman et al., 2008), other stress expressions and cytokine activation (Wilson et al., 2010).

Oxidant Indoors

The most important oxidants in indoor environments are expected to be O_3, OH, and NO_3 (Gligorovski and Weschler, 2013; Waring and Wells, 2015). Of these, indoor O_3 has received the most attention and is best understood at present (Britigan et al., 2006; Weschler, 2000). O_3 is known to be lost by reaction on indoor surfaces, with VOCs and with human skin oils (Coleman et al., 2008; Singer et al., 2006; Wisthaler and Weschler, 2010). Some studies on the abundance, sources, and impact of OH indoors have been carried out (Gligorovski et al., 2014; Weschler and

Shields, 1997), where OH abundances are expected to be one to two orders of magnitude smaller than outdoor levels due to low OH production, but much remains to be learned. Even less is known about the abundance of NO_3 with only one published study of indoor measurements (Nøjgaard, 2010). It is not clear whether the Criegee intermediate or some other unidentified oxidants may be playing important roles indoors.

3.4 CHEMICAL PROCESSES INTERACT WITH ATMOSPHERIC DYNAMICS TO CONTROL THE DISTRIBUTION OF TRACE GASES AND PARTICLES

Overview

The distribution of atmospheric trace constituents reflects a balance between the effects of chemical sources and sinks, chemical transformations, and atmospheric transport. Dynamical processes in both the troposphere and stratosphere drive how quickly gases and particles are transported in these regions, as well as vertically between the two regions. Climate-related changes in these processes will drive changes in the distribution of the trace constituents, which can feed back on the dynamics, with implications for weather, air quality, and climate. In the troposphere, extratropical cyclones, which travel along the storm tracks, play an important role in the export of pollution from the continental planetary boundary layer to the global atmosphere. In the stratosphere, the dominant atmospheric transport process is the large-scale meridional overturning circulation, also known as the Brewer-Dobson circulation (Brewer, 1949; Dobson, 1956). Changes in this overturning circulation will influence the transport of pollution from the troposphere into the stratosphere, and the transport of ozone from the stratosphere to the troposphere. Furthermore, the dynamics of the troposphere and stratosphere are coupled, and changes in the stratospheric circulation can influence the storm tracks in the troposphere. Better understanding is needed for how climate-related changes in these transport processes affect the composition and chemistry of the atmosphere and how these chemical changes in turn feed back on atmospheric dynamics.

Recent Advances

Stratospheric Overturning Circulation

Recent advances have been driven in part by the use of chemistry-climate models with increasing spatial resolution and more detailed tropospheric and stratospheric chemistry. These models have robustly shown, for example, that warming due to increased greenhouse gases results in intensification of the Brewer-Dobson circulation (SPARC CCMVal, 2010), which has implications for the distribution of trace gases in the stratosphere and troposphere. Hegglin et al. (2014) reported a negative trend in water vapor in the lower stratosphere and a positive trend in the upper stratosphere, which they attributed to changes in the Brewer-Dobson circulation. Stratospheric water vapor, in addition to functioning as a greenhouse gas, also influences stratospheric chemistry by serving as a source of OH and contributing to the formation of polar stratospheric clouds that are key to important ozone-destroying heterogeneous chemistry (see Chapter 2). A positive trend in stratospheric water vapor that exceeded the increase expected from the oxidation of atmospheric methane has been suggested (Rosenlof et al., 2001; Rosenlof and Reid, 2008), but the more recent work by Hegglin et al. (2014) suggests that the trend inferred from balloon data may not be globally representative. This discrepancy highlights the need for better observational coverage—including satellite observations—to understand the changing composition of the atmosphere.

Changes in the Brewer-Dobson circulation also affect tropospheric ozone. Hegglin and Shepherd (2009) estimated that climate-induced strengthening of the Brewer-Dobson circulation could contribute to an increase in the flux of ozone into the troposphere of as much as 23 percent by 2095 (relative to 1965), although Neu et al. (2014) suggested a more modest increase of about 2 percent. While transport of ozone from the stratosphere represents only about 10 percent of the total budget of tropospheric ozone, it is an important source of ozone throughout the upper extratropical troposphere and may contribute significantly on an episodic basis to regional ozone budgets. For example, Lin et al. (2012) predicted that transport of ozone from the stratosphere accounted for 50–60 percent of surface ozone abundances in the western United States, two to three times greater than estimates from previous modeling studies.

Extratropical Jets

Another robust feature in climate models of increased greenhouse gases is a poleward shift of the midlatitude westerly jets (between 30–60 degrees latitude), associated with an expansion of the Hadley cells (low-latitude overturning circulations with rising air at the equator and sinking air at roughly 30° latitude) in the tropics (e.g., Lu et al., 2007; Yin, 2005). Comparisons of models with observations indicate that they generally underestimate the widening of the Hadley cells observed since 1979 (Johanson and Fu, 2009). This bias in the jet position in the models has implications for projections of changes in jet variability and thus midlatitude storm tracks. Barnes and Polvani (2013) suggested that changes in the variability of jets are linked to the mean latitudinal position of the jets. Consequently, models that have an equatorward bias in the jet position are biased in their projection of climate-related changes in jet variability and storm tracks. These models also incorrectly estimate climate-induced changes in summertime surface ozone variability since the maximum in surface ozone variability is linked to the position of the jets (Barnes and Fiore, 2013).

In the southern hemisphere, the shift in the jet is also linked to ozone depletion in the Antarctic stratosphere. Increases in greenhouse gases and stratospheric ozone loss have acted in concert to drive a positive trend in the southern annular mode, which is associated with the poleward shift of the westerly jet (Thompson and Solomon, 2002). The trend is robust in austral summer, reflecting the seasonal reduction in stratospheric ozone in austral spring and summer, and is accompanied by a summertime widening of the Hadley cell (Son et al., 2010). As the stratospheric ozone recovers, projected ozone-related changes to the circulation in the southern hemisphere will oppose the circulation response to increased greenhouse gases (Son et al., 2010); this change will complicate attempts to isolate the impact of increased greenhouse gases on storm tracks in the southern hemisphere.

Cyclones and Monsoons

Climate-related changes in the circulation of the troposphere have also influenced the frequency of extratropical cyclones, which play an important role in ventilating the planetary boundary layer. However, the impact on air quality of these changes in cyclone frequency is uncertain. Wu et al. (2008) suggested that a reduction in the frequency of these cyclones across the United States in the late 20th century may have reduced by 50 percent the air quality benefits expected from decreased anthropogenic emissions in the United States between 1980–2006. However, Turner et al. (2013) predict that there was little change in surface ozone exceedances, despite a decrease in the number of cyclones. These cyclones also influence the transport of continental pollution to the remote atmosphere. Much work has been done to examine intercontinental transport of pollution in the northern hemisphere (e.g., Auvray et al., 2007; Doherty et al., 2013; Fiore et al., 2009; Liang et al., 2004; Owen et al., 2006). Fewer studies (e.g., 2002; Staudt et al., 2001, 2002) have focused on the impact of pollution on the remote southern tropics and subtropics. For nutrients and pollutants of biogeochemical importance, long range transport plays a key role in the movement of phosphorus from North Africa to the Amazon (Swap et al., 1992) or iron into the southern ocean (Gasso et al., 2010); how these will change under climate change is an important area of future study. It is unclear how changes in storm tracks in the southern hemispheres, combined with increasing emissions from countries in South America and Africa, for example, affect the composition and chemistry of the remote atmosphere in the southern hemisphere.

Aerosol particles can have an important impact on the radiation budget and dynamics of the atmosphere. Studies have linked changes in rainfall in the South Asian and East Asian monsoons to the radiative effects of aerosol particles (e.g., Bollasina et al., 2013; Lee and Kim, 2010; Menon et al., 2002; Wang et al., 2015a). Surface cooling, driven largely by sulfate aerosol particles, tends to weaken the atmospheric monsoon circulation and results in reduced precipitation. In contrast, heating from black carbon enhances the circulation and leads to increased precipitation. Bollasina et al. (2014) argued that the influence of local aerosol particles on the dynamics of the atmosphere was the dominant driver for the late 20th century reduction in precipitation in South Asia. It has been suggested that anthropogenic aerosol particles in the northern hemisphere resulted in a southward shift in the Hadley circulation during the 20th century, producing a decrease in precipitation in the northern tropics and an increase in precipitation in the southern tropics (e.g., Hwang et al., 2013; Ming and Ramaswamy, 2011). Recent work by Allen (2015) suggested that future reductions in anthropogenic sulfate aerosol particles may drive a northward shift in precipitation in the tropics during the 21st century. However, Kirkby et al. (2016) showed the nucleation of aerosol particles with only highly oxygenated molecules, thus sulfate reduction may not reduce aerosol particle and CCN abundances as much as currently thought. Mechanisms for VOC oxidation and particle nucleation need to be included at the appropriate level of detail in climate models to better understand the influence of aerosol particles on the stability and large-scale circulation of the atmosphere.

3.5 PARTICLE CHEMICAL AND PHYSICAL PROPERTIES AFFECT CLOUD AND AEROSOL PARTICLE RADIATIVE PROPERTIES, CLOUD MICROPHYSICS, AND PRECIPITATION PROCESSES

Overview

Aerosol particles and clouds play a key role in the Earth's energy budget by directly scattering and absorbing radiation. Aerosol particles can also directly affect atmospheric stability and moisture fluxes that drive convective processes. Aerosol particles are the nuclei upon which clouds form and variations thereof can modulate cloud properties, the radiation budget, and the hydrological cycle. Most cloud condensation nuclei (CCN) grow from particles formed by condensation of molecules produced by atmospheric chemistry, but that are initially too small to act as CCN. Clouds in turn can also strongly affect aerosol particles through chemical and microphysical processes. The transformations that occur in aerosol particles during their interactions with water and clouds can affect ecosystems by modulating nutrient fluxes, and changes in precipitation and radiation can alter ocean thermohaline circulation. The interactions between aerosol particles and clouds are multiscale (spanning from spatial scales of meters to thousands of kilometers and timescales from minutes to months), multiphase (among water vapor, liquid water, ice, and all the components found in aerosol particles), and involve a multitude of dynamical and physical processes that couple them.

Inclusion of aerosol particle-cloud interactions in previous Intergovernmental Panel on Climate Change (IPCC) assessments completely changed the model responses to aerosol particles and revealed some fundamental problems with the treatment of cloud formation and cloud feedbacks in global models (Boucher et al., 2013). Combined with the large diversity of natural and anthropogenic sources of aerosol particles over space and time, aerosol-cloud-precipitation-radiation interactions constitute the largest source of uncertainty in assessments of climate sensitivity and anthropogenic climate change.

In terms of weather forecasts, there is a history of weather modification through cloud seeding with aerosol particles like silver iodide, but the scientific basis for its effectiveness has been difficult to ascertain (NRC, 2003). Nevertheless, with recent attention to the question of how particles affect precipitation, a combination of new measurements and chemically enhanced weather forecast models have opened up this field. For example, increases in aerosol particles have been linked to invigoration of convection and the occurrence of intense precipitation events (Koren et al., 2012). The use of interactive aerosol particles in weather forecast models is fairly recent, but is showing that interactive assimilated aerosol particles can increase the forecast accuracy (Kolusu et al., 2015; Sessions et al., 2015). For example, as part of the European Copernicus programme, atmospheric composition variables (greenhouse gases, aerosol particles, and chemical species) have been introduced in the ECMWF model to improve the radiative heating and the Numerical Weather Prediction (NWP) system itself, and other forecast models are engaged in similar experiments (Baklanov et al., 2014; Eskes et al., 2015; Kong et al., 2015; Pleim et al., 2014).

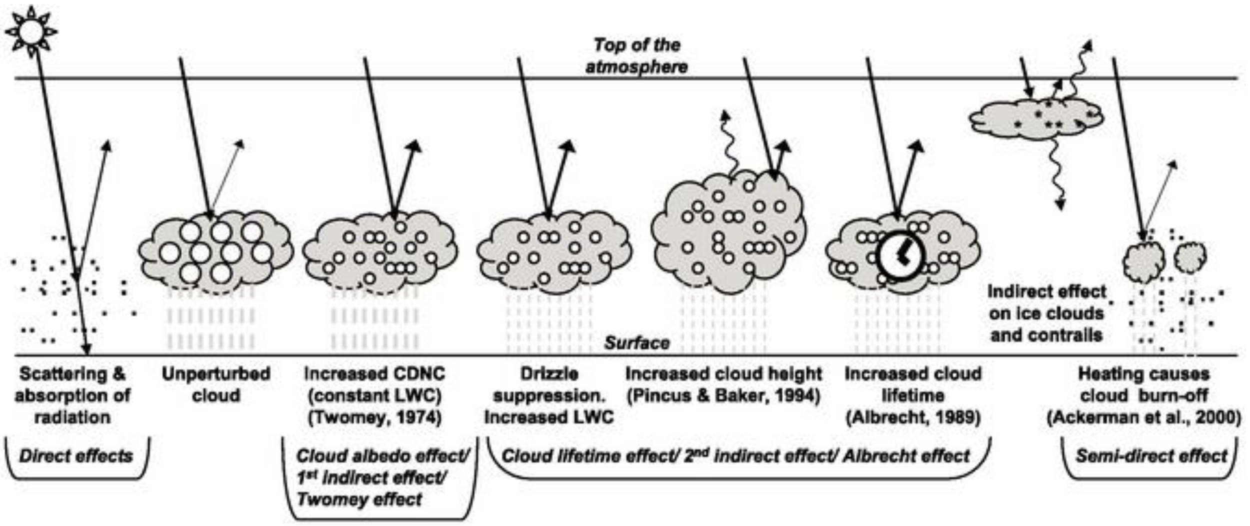

FIGURE 3.4 Schematic diagram showing the various radiative mechanisms associated with cloud effects that have been identified as significant in relation to aerosol particles (modified from Haywood and Boucher, 2000). The small black dots represent aerosol particles; the larger open circles cloud droplets. Straight lines represent the incident and reflected solar radiation, and wavy lines represent terrestrial radiation. The filled white circles indicate cloud droplet number concentration (CDNC). The unperturbed cloud contains larger cloud drops as only natural aerosol particles are available as cloud condensation nuclei, while the perturbed cloud contains a greater number of smaller cloud drops as both natural and anthropogenic aerosol particles are available as cloud condensation nuclei (CCN). The vertical grey dashes represent rainfall, and LWC refers to the liquid water content. SOURCE: IPCC, 2013.

Recent Advances

Atmospheric aerosol particles are emitted from a wide variety of sources including soil and deserts, the ocean, volcanoes, biogenic activity, biomass burning, burning of fossil fuels, and numerous other anthropogenic activities. As discussed in Chapter 3.2, reactions of gaseous precursors to form low volatility products also comprise a major source of airborne particles. Aerosol particle composition and size distribution are determined by their sources and are further modified in the atmosphere by chemical reactions with gases, photochemistry, and through interactions with clouds (Boucher et al., 2013). Cloud feedbacks and the resulting buffered response of clouds and cloud systems mitigate much of the small-scale responses of clouds and radiation to aerosol particles and challenge the understanding of aerosol particles in the Earth system (Stevens and Feingold, 2009). Nevertheless, considerable progress in understanding the links between aerosol particles, radiation, and clouds has been established; see Figure 3.4 for overview. The observational capacity for aerosols has advanced substantially in recent decades both via in situ instrumentation (e.g., Jayne et al., 2000) and satellite observations (e.g., Kaufman et al., 1997), thus improving our understanding of the distribution and composition of particles. Yet the heterogeneity and short lifetimes of these particles ensure that characterization of atmospheric aerosols remains an on going challenge.

Aerosol Direct Effect

Decades of research have established that aerosol particles directly scatter and absorb radiation and affect the planetary energy balance. Of all aerosol particle physical parameters that influence scattering efficiency, the uptake of water with increasing humidity, which is controlled by the composition of the particles, is most important (e.g., Pilinis et al., 1995; Seinfeld and Pandis, 2006; Wagner et al., 2015). The highly variable relative humidity across the scales, and the nonlinear response of aerosol water uptake to it, introduces aerosol direct radiative forcing uncertainty. Recent model intercomparisons suggest that estimates of the global all-sky radiative effect of anthropogenic aerosol remains quite uncertain (-0.58 Wm^{-2} to -0.02 Wm^{-2}) (Myhre et al., 2013). Furthermore, while estimates of aerosol direct radiative forcing are typically dominated by scattering aerosols, the degree of warming provided by absorbing aerosol has been an active area of research, particularly over the last decade.

The latest IPCC report ranked black carbon as the second most important climate warming agent, after CO_2 (IPCC, 2013). However, uncertainties surrounding the sources, mixing state, and optical properties of black carbon lead to large uncertainty in its ultimate impact on the absorption of solar radiation (Bond et al., 2013). The mechanism behind and prevalence of coated black carbon and the associated absorption enhancement is also an active research area (e.g., Cappa et al., 2012). Network absorption measurements and airborne measurements of black carbon over remote regions suggest that models typically overestimate its lifetime, supporting a downward revision of the climate forcing attributed to black carbon (Wang et al., 2014a,b). Quantifying the sources, transport, and removal of black carbon is therefore key to understanding its radiative forcing.

Brown carbon (organic carbon aerosol that absorbs at ultraviolet and visible wavelengths) may also make an important contribution to solar absorption and climate forcing. Initial modeling studies suggest that brown carbon could globally contribute 20–30 percent of the total aerosol absorption at visible wavelengths (e.g., Feng et al., 2013; Wang et al., 2014b). However, the sources and composition of brown carbon, its location throughout the atmosphere (especially in the vertical) as well as its absorption characteristics and how these might evolve in the atmosphere are not well known (e.g., Forrister et al., 2015; Laskin et al., 2015; Saleh et al., 2014).

The AR5 assessment of aerosol-radiation interactions was more uncertain than previous IPCC assessments, reflecting a growing understanding of aerosol sources and properties and, correspondingly, an increasingly complex treatment in models. Much work is needed to reduce the uncertainty on this key climate-relevant metric.

Aerosol Indirect Effect

Clouds can respond to aerosol perturbations through a large suite of complex interactions that affect cloud extent, precipitation rate, and radiative properties. Here we focus on those that are directly impacted by aerosol particle modulations, noting that the overall cloud responses to aerosol may be more complex than represented here owing to the nature of clouds themselves. Enhanced aerosol particle concentrations that act as CCN may affect the distribution and vertical extent of clouds with important feedbacks on the hydrological cycle (e.g., Andreae et al., 2004; Rosenfeld, 2006; Rosenfeld et al., 2008). Increased concentrations of large aerosol particles, termed giant CCN, may act as efficient collector drops and promote the formation of precipitation (e.g., Cheng et al., 2009; Levin and Cotton, 2009; Woodcock, 1950). The presence of large numbers of giant CCN (e.g., generated during storms) may also deplete water vapor availability in the early stages of cloud formation, strongly affecting the sensitivity of cloud droplets to aerosol particle variations (e.g., Ghan et al., 1998; Morales Betancourt and Nenes, 2014). Another effect of increased aerosol particles is enhancement of evaporation rates of droplets at cloud fringes, which may result in a negative buoyancy feedback from evaporative-entrainment and reduce cloudiness (e.g., Xue and Feingold, 2006). These complex dynamical interactions are neither well understood nor represented in large scale models.

Particles that act as ice nuclei (IN) are rare in comparison with CCN; about one in a million aerosol particles acts as an IN. It is known that this ability is strongly a function of temperature, and controlled by the physical characteristics and chemical composition of the aerosol particles in a fundamentally different way from CCN. A quantitative understanding of what makes an effective ice nucleating particle and a generally accepted theory for ice nucleation is lacking. Part of this difficulty in establishing a theory is related to the surface chemical and morphological complexity of the IN. The other difficulty lies in the multiple ways that IN can catalyze the formation of ice (the so-called “modes” of freezing, e.g., deposition, immersion, deliquescence, contact). The small number of particles that do act as IN makes measurement challenging and also has profound implications for clouds that contain ice. For example, if one IN or cloud droplet freezes in an existing cloud, it begins growing at the expense of the existing water droplets because the vapor pressure of water over ice is lower than over liquid water. Hence, the introduction of ice nuclei to a mixed phase cloud modulates cloud droplet size and concentration, which in turn may affect its precipitation efficiency, lifetime, shortwave reflectivity, and longwave emissivity (e.g., Lohmann and Diehl, 2006; Lohmann and Feichter, 2005; Rosenfeld, 2006; Rosenfeld et al., 2008). Increases of IN can profoundly affect pure ice (cirrus) clouds as well, as IN form ice before the more populous supercooled haze droplets. The ice from IN strongly competes for water vapor with the supercooled drops, potentially reducing or completely inhibiting their freezing, strongly affecting crystal number and size, hence cloud lifetime and longwave emissivity (e.g., Gettelman et al., 2012; Jensen et al., 2010; Liu et al., 2012; Lohmann and Feichter, 2005; Murray et al., 2010; Ren and Mackenzie, 2005).

An understanding of the behavior of and changes in ice and mixed-phase clouds requires identification of the number of IN in each location. A number of materials have been identified as ice nuclei, including mineral dust, soot, bacteria, fungal spores, pollen, crystalline soluble salts, glassy aqueous materials, and volcanic ash (Hoose and Möler, 2012; Moreno et al., 2013; Murray et al., 2012) with mineral dust being an especially important contributor to IN concentration (e.g., Atkinson et al., 2013; Baustian et al., 2012; Creamean et al., 2013; Cziczo et al., 2013; DeMott et al., 2003a,b; Kamphus et al., 2010; Pratt et al., 2009b). Highly variable contributions from bioparticles can regionally be very important for IN concentrations, especially in regions of agricultural activity (Tobo et al., 2013) and intermediate temperatures (Spracklen and Heald, 2014). Regionally and temporally, biomass burning can act as an IN source (McCluskey et al., 2014). Strong differences in the source and type of IN are expected to exist between land and ocean (Burrows et al., 2013), with potentially important impacts on cloud properties. Recently, biogenic organic material has been suggested to be an important source of IN in remote marine ocean environments (Wilson et al., 2015). While several modeling and empirical approaches describe heterogeneous ice nucleation (e.g., Barahona, 2012; Broadley et al., 2012; Connolly et al., 2009; DeMott et al., 2016; Herbert et al., 2014; Hoose et al., 2010;

Khvorostyanov and Curry, 2005; Niedermeier et al., 2011; Vali, 1994), the lack of an accepted mechanistic description for ice nucleation processes severely limits predictive capability in this area.

Compared to pure liquid and pure ice clouds, the effects of aerosol particles on mixed-phase clouds are more complex, owing to thermodynamic and microphysical interactions between cloud particle types throughout the cloud (Cheng et al., 2010; Fan et al., 2013; Lance et al., 2011; Lebo and Morrison, 2014; Lebo and Seinfeld, 2011; Rosenfeld et al., 2008; Saleeby et al., 2011; Storer et al., 2010; Van den Heever et al., 2006). The effects of particles on precipitation is a function of the cloud microphysics and thermodynamics, where particles can suppress warm rain processes and can enhance or suppress cold rain processes (Cheng et al., 2010). The end result of particle modulations on precipitation therefore strongly depend on the specific conditions of cloud formation examined. Precipitation enhancement has been explained by the phase change and release of latent heat during the transport of liquid mass to freezing levels that then fuels the updraft velocity (Lebo and Seinfeld, 2011). Microphysical features of deep convective clouds are sensitive to the type and size of hygroscopic aerosol particles (Storer et al., 2010). In deep convective systems, smaller drops resulting from CCN addition can be lofted higher in the atmosphere given the same updraft velocity (Rosenfeld et al., 2008). This process can invigorate convection, where the latent heat release from freezing enhances updrafts or secondary convection (Van den Heever et al., 2006). These changes in cloud microphysics affect precipitation of all hydrometeor types, driving thermodynamic and dynamic changes that can affect regional scale precipitation (Fan et al., 2013; Lebo and Morrison, 2014).

Connecting the effects of clouds on radiation and precipitation requires deep understanding of the relationship between clouds and aerosol particles. This relationship is directly controlled by atmospheric chemistry, and the potential responses that can prevail for each cloud type and state. The major mechanisms of interaction between aerosols and clouds have been evaluated on a microphysical level and estimated by cloud-resolving and large-scale models. Further research is needed to confirm aerosol particle influence on cloud-scale dynamics and to embed these influences in multi-scale models that can ultimately evaluate changes in radiative balance and precipitation on regional and global scales.

3.6 ATMOSPHERIC TRACE GASES AND AEROSOL PARTICLES COUPLE TO GLOBAL BIOGEOCHEMICAL CYCLES

Overview

Atmospheric chemistry plays a central role in the biogeochemical cycling of elements (e.g., carbon, nitrogen, sulfur) through the Earth’s system (Andreae and Crutzen, 1997; Duce et al., 2008; Nadelhoffer et al., 1999). Chemically reduced gases and particles are emitted from terrestrial and marine ecosystems, oxidized or otherwise transformed in the atmosphere, and deposited downwind. Biogeochemical cycles control the elements that are necessary for life on Earth. These cycles support the basic functioning and biodiversity of life (including humans), link biotic and abiotic systems, and ensure the continuous survival of ecosystems. Thus biogeochemical cycles also support the resource production associated with maintaining global food security (e.g., agriculture, fisheries) and certain energy sources (e.g., biofuels, wood). There is a critical need to understand the global distribution of fluxes of elements between the atmosphere and both the terrestrial and marine ecosystems, as well as how atmospheric chemical processes modulate the composition and bioavailability of these elements.

Furthermore, biogeochemical cycles are subject to the influence of ever-changing human activity and global climate and play an important role in the energy balance of the Earth system. The indirect climate forcing exerted by aerosol particles via biogeochemical cycles has recently been estimated to be as large as the direct impact of aerosol particles on radiation (Mahowald, 2011). This estimate largely reflects the enhanced carbon uptake associated with nitrogen, phosphorus, and iron deposition to the biosphere.

Recent Advances

New approaches and tools have largely enabled advances in connecting atmospheric chemistry processes with biogeochemical cycles. The advent of Earth system models, which dynamically couple reservoirs in the Earth system, has initiated new avenues of retrospective and predictive atmospheric chemistry modeling connected to biogeochemistry (e.g., investigation of the role of ozone on the land carbon sink [Sitch et al., 2007] and how climate change affects global dust sources [Mahowald, 2007]). Development and deployment of new analytical measurement approaches permit a more comprehensive characterization of atmospheric constituents relevant to biogeochemical cycling (e.g., new observations of atmospheric ammonia [Altieri et al., 2014; von Bobrutzki et al., 2010] and atmospheric trace metals such as divalent gaseous mercury [Landis et al., 2002; Park et al., 2013]). Global Earth observations from satellite platforms provide new constraints on the sources and transport of key nutrients over poorly observed regions (e.g., global ammonia emissions, constraints on NO_x emissions from soils, and transport of dust over the Atlantic [Clarisse et al., 2009; Jaegle et al., 2004; Kaufman et al., 2005]). Additionally, long term in situ concentration measurements allow the measurement of interannual fluctuations in important constituents, providing new information on the role of biogeochemical cycles at longer time scales (e.g., Prospero and Lamb, 2003).

The biosphere is reaching a state where human impacts are beginning to overwhelm natural ecosystems, which could lead to irreversible changes to the Earth system (Barnosky et al., 2012). Ecosystem changes can be abrupt and due to complex interactions and feedbacks and they are difficult to predict. Changes to the health of global ecosystems impact atmospheric composition by affecting biogenic emissions (see Section 3.1). For example, warming temperatures in the Arctic will result in reduced permafrost as well as increased emissions of methane and carbon dioxide from the land to the atmosphere (O'Connor et al., 2010). Land use and land cover change may play a role in controlling these surface-atmosphere exchange processes and the composition of the atmosphere (Heald and Spracklen, 2015; Unger, 2014). The ocean and atmosphere represent an important and strongly coupled biogeochemical system, where changes in atmospheric deposition of nutrients in the form of dust and other aerosols can induce phytoplankton blooms which change oceanic composition and emissions (Andreae and Crutzen, 1997; Boyd et al., 2007; Chien et al., 2016; Ito et al., 2016; Suntharalingam et al., 2012). Predictions of the atmospheric composition require a stronger understanding of these complex human-ecosystem interactions.

Nutrient Exchange with the Biosphere: Nitrogen

Humans, through the development of industrial ammonia fertilizer, as well as fossil fuel emissions of nitrogen oxides, have dramatically enhanced the nitrogen supply of the atmosphere (Erisman et al., 2008). Atmospheric deposition of this nitrogen can act as a beneficial external nutrient supply in both terrestrial and marine ecosystems. Over half of the global oceans are depleted in key macronutrients such as nitrate required for phytoplankton growth, and atmospheric deposition provides a key input for nitrogen in these regions (Guieu et al., 2014). However in the terrestrial environment, nitrogen inputs may lead to oversaturation, threatening biodiversity and leading to eutrophication of aquatic ecosystems (Beem et al., 2010; Erisman et al., 2007). Recent work suggests nitrogen deposition will decline world-wide as increases in ammonia emissions will be offset by large decreases in nitrogen oxide emissions (Lamarque et al., 2013). Despite this potential emission shift, other studies show that sensitive ecosystems may continue to be threatened by excessive nitrogen deposition (Ellis et al., 2013).

Nutrient Exchange with the Biosphere: Iron and Phosphorus

Mineral dust represents a major source of iron and other micronutrients which are essential for marine primary productivity (Maher et al., 2010). Iron is the limiting nutrient for over 25 percent of the surface ocean (Martin, 1990). There has been continuing interest in the relationship between atmospheric iron deposition and marine biological productivity given the proposed link between sea spray aerosols in biologically active regions and climate (Boyd et al., 2007). Changes in the atmospheric dust cycle could play a role in climate dynamics by altering the ocean-atmosphere carbon cycle and thus atmospheric carbon dioxide concentrations (Jickells et al., 2005; Mahowald et al., 2010; Parekh et al., 2006), although evidence for the importance of such a dust-ocean-climate feedback process is still limited (Schulz et al., 2012). Phosphorus, which is a limiting nutrient for many forest ecosystems including the Amazon, is supplied through the transport of smoke particles, bioparticles, and mineral dust (Mahowald et al., 2008a; Ridley et al., 2012). Because dust is a vector for nutrient transport, better constraints on global natural and anthropogenic dust source regions using satellite observations (Ginoux et al., 2012), and examining contemporary trends in dust transport (Ridley et al., 2014) are areas that need to be further developed.

Bioavailability is largely governed by the chemical speciation of a nutrient and, in general, insoluble species are not bioavailable. For iron and phosphorus (and perhaps other nutrient trace metals), solubility increases during transport through the atmosphere (Baker et al., 2006; Longo et al., 2016). The causes of this increase are complex and uncertain, but recent work suggests that the interactions of aerosol particles with acids plays an important role in the process (e.g., Baker and Croot, 2010; Kumar et al., 2010; Longo et al., 2016; Meskhidze et al., 2005; Nenes et al., 2011). These interactions of metals with atmospheric acidity, whether from sulfate, nitrate, or organic acids, lie at the intersection of atmospheric biogeochemistry and more traditional atmospheric chemistry. Recent studies have also highlighted the importance of the mineralogy of the dust for the amount of easily solubilized iron (e.g., Journet et al., 2008; Schroth et al., 2009) as well as the role of combustion sources of soluble iron (Chuang et al., 2005; Guieu et al., 2005).

Ozone Exchange with the Biosphere

Tropospheric ozone can damage vegetation, leading to reductions in the global land carbon sink (Sitch et al., 2007) and declines in global agricultural productivity (Avnery et al., 2011; Tai et al., 2014). Although uptake of ozone by vegetation represents a harmful ecological impact, it is also one of the primary removal mechanisms for surface ozone. However, estimations of this impact are highly dependent on the description of land use and vegetation phenology in models (Park et al., 2014; Val Martin et al., 2014). Efforts have been made to routinely estimate ozone dry deposition velocities from near-surface concentration measurements (e.g., Clarke et al., 1997). While direct measurements of ozone deposition flux are available from an array of specific sites for short-term studies, long-term measurements are limited (e.g., Fares et al., 2010; Munger et al., 1996).

Organic Compound Exchange with the Biosphere

The deposition of trace gases and aerosol particles to terrestrial and marine ecosystems plays a vital role in controlling the lifetime of atmospheric chemical species, while at the same time, dictating the location and extent of impacts described above. Goldstein and Galbally (2007) highlighted that a poor understanding of the sinks of organic species affects the ability to constrain the global budget of VOCs, yet almost no measurement constraints are available to quantify this sink. Hallquist et al. (2009) estimated that deposition of oxidized vapors is the dominant sink of VOCs from the atmosphere, further emphasizing the need for new observations. New analytical approaches have been applied to estimate the surface fluxes of many gas-phase organic species, allowing measurement of the location of deposition, and thus its impact (Karl et al., 2010; Nguyen et al., 2015a; Park et al., 2013). Beem et al. (2010) and Kanakidou et al. (2012) showed that the wet deposition of organic nitrogen can make up an important fraction of total nitrogen deposition to terrestrial and marine ecosystems, respectively. However, while network measurements regularly report the wet deposition flux of inorganic acids throughout much of North America and Europe (e.g., the National Atmospheric Deposition Program in the United States), the wet removal of organic species (including organic nitrogen and organic acids) is not routinely monitored. In addition, much of the deposition occurs in the form of dry deposition, for which there are very limited observations. Ecosystem damage from acid deposition (i.e., acid rain) has been quantified and addressed in some regions (see Chapter 2); yet, the way in which this type of damage translates to reductions in global carbon uptake is not well known.

Relative to terrestrial sources, far fewer measurements have characterized VOC emissions from the ocean. Recent studies have quantified marine emissions of species such as isoprene and monoterpenes, and these limited measurements indicate that these emissions are extremely low (Carpenter et al., 2012). Several decades ago, scientists speculated the ocean could act as a “planetary thermostat” and regulate climate through biogenic organosulfur (dimethyl sulfide) emissions which undergo processing to ultimately produce sulfate aerosols that can seed marine clouds and lead to changes in cloud properties, the so-called CLAW hypothesis (Charlson et al., 1987). After decades of conflicting results from marine field studies, the evidence for the CLAW hypothesis remains elusive, however the links between the biological productivity of the ocean, sea spray and gas phase emissions, and clouds are still under debate (Quinn et al., 2015).

Mercury Exchange with the Biosphere

The atmosphere transports and transforms compounds that damage ecosystem and human health (e.g., mercury, ozone, polycyclic aromatic hydrocarbons). For example, the understanding of the global biogeochemical cycling of mercury has advanced considerably over the last decade, in large part due to new measurements and modeling of atmospheric mercury and its transformations (Ariya et al., 2015; Selin, 2009). Recent advances include better modeling of atmosphere-ocean coupling (Soerensen et al., 2010), a better understanding of polar mercury depletion events (Steffen et al., 2008), the identification of bromine as a globally-important oxidant of elemental mercury (Ariya et al., 2015; Holmes et al., 2010; Obrist et al., 2011), especially in the high latitudes, and a better understanding of mercury sources (Pirrone et al., 2010). However, the effect of the deposition of mercury and other toxic constituents on ecosystem productivity is poorly understood. Limited efforts have explored the biogeochemical cycles and environmental impacts of other contaminants, such as persistent organic pollutants (e.g., Lammel et al., 2009).

3.7 CONCLUSION

The topics described in this chapter span the discipline of atmospheric chemistry research. This chapter has discussed a few examples that have recently been recognized as being important for understanding and predicting atmospheric processes and impacts, but for which key areas of uncertainty remain. These issues not only contribute to advancing fundamental scientific knowledge in a number of fields, including chemistry, physics, and meteorology, but—as described in the next chapter—they also lie at the core of understanding the basis of, and possible solutions to, many issues central to human health and welfare.

Chapter 4—Anticipating Tomorrow: Societal Challenges

As described in Chapter 2, atmospheric chemists have contributed to the response to many societal problems with use-inspired basic research. The first three sections (see Chapters 4.1–4.3) in this chapter focus on key areas where atmospheric chemistry research is currently contributing to the understanding of societally relevant issues and where the Committee believes there should be increased attention in the coming decade. These include the relationships of atmospheric chemistry to the weather and climate system, human health, and ecosystems, areas where rapid changes are occurring and where there are opportunities for important progress in the coming decade to better inform societal choices. The last section (Chapter 4.4) examines a number of areas where atmospheric chemistry affects society and where societal choices influence atmospheric composition, ultimately affecting people in different ways. The relationship among these areas is shown schematically in Figure 4.1.

4.1 ATMOSPHERIC GASES AND PARTICLES AFFECT CLIMATE AND WEATHER

The increase in the atmospheric abundance of greenhouse gases and particles due to human activity has altered climate with profound implications for society. Global mean temperature has increased by 0.8°C since the pre-industrial era, and 15 of the 16 warmest years since 1880 have occurred since 2000.[10] Despite the importance of this trend, the change in global mean temperature is just one indicator of the changing climate system. Other impacts of a changing climate include rising sea levels, increasing frequency of heavy precipitation, and significant seasonal ice loss in the Arctic. Some types of extreme events, which can incur large costs on the economy, are known to be exacerbated by climate change (NASEM, 2016). For the period of 1980–2011, it was estimated that droughts and heat waves cost the U.S. economy a total of $210 billion and floods cost $85 billion (Smith and Katz, 2013). These extreme climate events also have implications for global food security. The drought in the United States in 2012 resulted in a 21 percent reduction in the yield of maize, relative to the previous five non-drought years, which led to a 54 percent increase in the price of maize (Boyer et al., 2013; Gilbert, 2016). This alteration highlights the fact that regional climate impacts have global implications in the interconnected global economy.

These changes to the climate are driven by increases in greenhouse gases, including carbon dioxide, methane, nitrous oxide, and ozone. Greenhouse gases absorb infrared radiation, and the increase in their atmospheric abundance due to human activity has produced a radiation imbalance in the atmosphere, which is referred to as radiative forcing of the climate system. Aerosol particles scatter and absorb solar radiation, producing a direct effect on climate. Aerosol particles also have an indirect effect on climate through their influence on clouds, which can lead to either warming or cooling of the climate system, although the overall global impact is cooling. See Figure 4.2 for an overview of the role of atmospheric gases, aerosol particles, and clouds in the climate system.

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[10] NOAA global analysis (2015): https://www.ncdc.noaa.gov/sotc/global/201513.

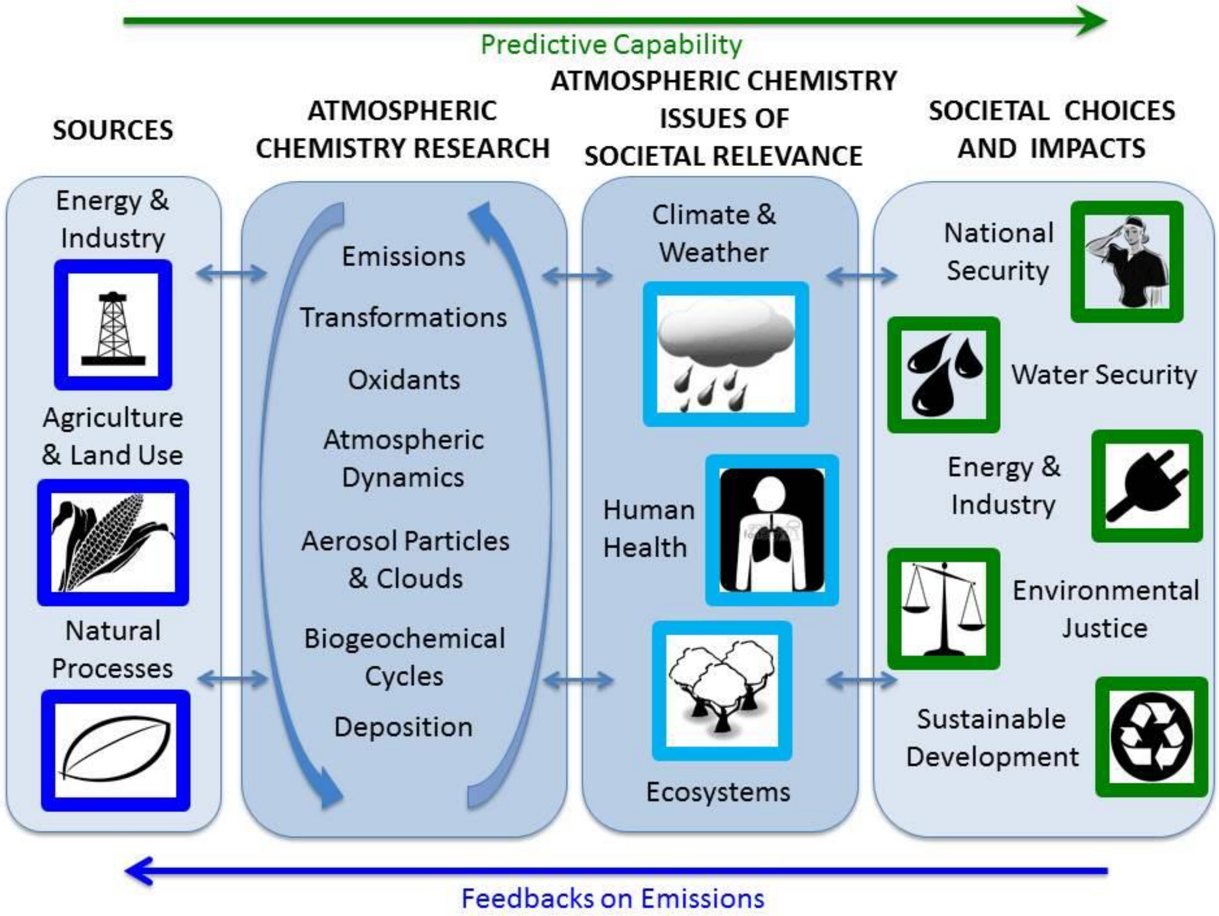

FIGURE 4.1 Schematic showing the connections among major emission sources to the atmosphere, atmospheric chemistry research, scientific issues of societal relevance, and subsequent societal choices and impact areas. Research in atmospheric chemistry helps to build a predictive capability for future environmental conditions that can indicate likely impacts to society of various policy choices.

Strong feedbacks exist between climate change and atmospheric composition. Changes in climate can impact ecosystems, resulting in changes in emission of gases and particles to the atmosphere (Bonan, 2008; Cramer et al., 2001; Hoegh-Guldberg and Bruno, 2010). A prime example is in the Arctic, which has experienced the greatest regional warming since the pre-industrial era. Continued warming in the Arctic could result in increased emissions of methane from the permafrost, which will feed back on the climate system directly, as methane is a greenhouse gas. Increases in atmospheric methane will increase tropospheric ozone and stratospheric water vapor, which are also greenhouse gases and play an important role in the chemistry of the troposphere and stratosphere, respectively (Akimoto et al., 2015; Isaksen et al., 2014; West et al., 2012). Many sources, such as wildfire and dust, respond to changes in climate and land use that may increase emissions of some gases and aerosol particles (Ginoux et al., 2012; Kloster et al., 2012; Knorr et al., 2016). For example, during the last glacial maximum, atmospheric dust concentrations were two to five times higher than interglacial periods (Maher et al., 2010). In addition, dust, black carbon (soot), and organic carbon deposited on snow and ice sheets reduce albedo, resulting in surface warming and faster melting (Yasunari et al., 2015). Obtaining an improved understanding of this coupling between atmospheric chemistry and

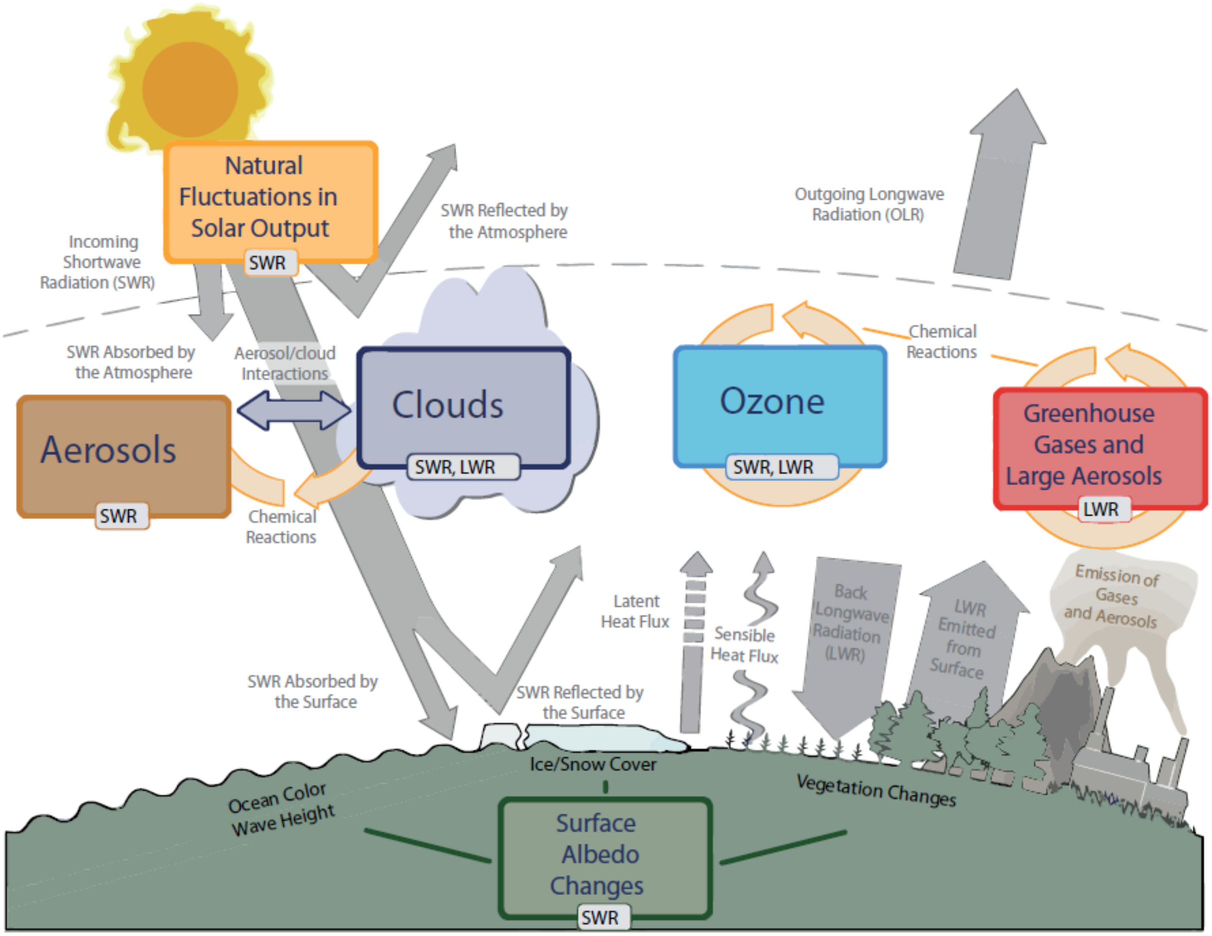

FIGURE 4.2 Schematic showing how atmospheric trace gases interact with the climate system. Atmospheric gases, aerosol particles, and clouds play a critical role in determining the energy balance of the planet. Abbreviations: shortwave radiation (SWR), longwave radiation (LWR). SOURCE: IPCC, AR5, 2013.

climate is essential for reliably predicting future changes in both atmospheric composition and climate.

The radiative forcing of climate on a global scale by well-mixed greenhouse gases is relatively well understood, and according to the IPCC (2013) the level of confidence in estimates of the radiative forcing is high for methane and halocarbons, and very high for carbon dioxide (CO_2) and nitrous oxide. However, projecting the regional response in climate to radiative forcing is challenging, and the coupling between the regional response and atmospheric composition and chemistry is a particular source of uncertainty. Climate-related changes in storm tracks will impact weather patterns as well as the transport of pollution, which has implications for air quality. Changes in the structure and intensity of convection influence the transport of pollution from the surface to the upper troposphere and lower stratosphere. These changes also impact the frequency of lightning, which is an important source of NO_x, a key precursor of tropospheric ozone (Schumann and Huntrieser, 2007; Seinfeld and Pandis, 2006). Changes in the lightning source of NO_x will have a dominant influence on ozone abundance in the upper troposphere, where ozone has the greatest capacity to affect climate.

BOX 4.1
Atmospheric Chemistry in the News: Methane Leak in California

From late 2015 to early 2016, a natural gas leak at the Aliso Canyon gas storage field, near Los Angeles, released almost 100,000 tons of methane into the atmosphere (Conley et al., 2016). The blowout occurred in October 2015 and, in the eighth attempt to control the leak, was finally sealed in February 2016, after surpassing any previously recorded methane leak in the United States. Methane is a potent greenhouse gas, and the warming anticipated from this leak will challenge California's ability to reach its greenhouse gas emission targets. Atmospheric chemists used instruments aboard a research aircraft to estimate the amount of methane emitted from Aliso Canyon. In addition to methane, they detected elevated atmospheric levels of other hazardous air pollutants, such as benzene, a known carcinogen (Conley et al., 2016). What was most evident to nearby residents was the odor due to organosulfur compounds added to the natural gas. Residents of a nearby community, Porter Ranch, complained of a range of physical ailments, and were evacuated in January 2016 when a state of emergency was declared in the region. Atmospheric chemists play a vital on going role in estimating the impacts of large-scale industrial accidents such as this and the Deepwater Horizon oil spill in 2010 (McGrath, 2016; Rich, 2016; Zielinski, 2016).

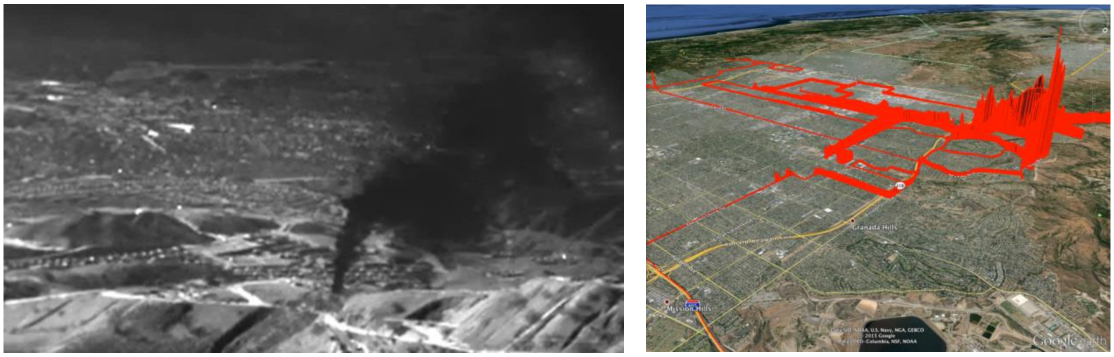

FIGURE A Right: Aerial footage from December 17, 2015, of the natural gas leak at a storage facility in California's Aliso Canyon. The methane plumes, which are naturally colorless, were made visible by a specialized infrared camera. SOURCE: Environmental Defense Fund. Left: Atmospheric measurements show the methane plume from the Aliso Canyon gas leak. SOURCE: http://www.greenmedinfo.com/blog/special-report-socalgas-natural-gas-leak-aliso-canyon.

There is less confidence in the radiative forcing associated with changes in the abundance of short-lived gases. There is also significant regional and seasonal variability in radiative forcing from these more chemically active gases (e.g., Bowman and Henze, 2012; Shindell et al., 2006). It has been suggested that controlling the abundance of short-lived climate pollutants, those with an atmospheric lifetime of less than 20 years, could provide a means of mitigating some of the expected changes in climate. For example, Shindell et al. (2012) showed that controlling methane and black carbon could reduce global mean warming by about 0.5°C by 2050. The initial focus for these pollutants has been on methane, tropospheric ozone, and black carbon, which are all warming agents. In addition, methane is a precursor of troposphere ozone, and ozone and black carbon are air pollutants. Consequently, reducing the atmospheric abundance of these short-lived pollutants would have the co-benefit of improving air quality (see Box 4.1). How radiatively-important short-lived pollutants have changed since preindustrial times is not well understood.

The impact of aerosol particles on the climate system is less well understood than the influence of well-mixed greenhouse gases, particularly their influence on clouds. Modeling, satellite remote sensing, laboratory, and field measurement studies over the last two decades have focused on understanding the impacts of aerosol particles on clouds and global climate. During this time, some scientific gaps have been closed, and additional processes have been identified that still elude quantification. As with many complex systems in intermediate stages of understanding, this progress has not yet reduced the overall magnitude of uncertainty, leaving major deficiencies in the ability to project future climate (Seinfeld et al., 2016). The root of this uncertainty is a lack of understanding of the contribution of natural and anthropogenic sources of global aerosol particles (Carslaw et al., 2013), their synergies and chemical transformations that affect how particles absorb and scatter light, as well as how particles take up water and their ability to serve as cloud and ice nuclei (e.g., Farmer et al., 2015; Fierce et al., 2013; Hoose and Mohler, 2012; McFiggans et al., 2006). The multi-scale and multi-phase coupling of aerosol particles with clouds and radiation further complicates its robust representation in models (Stevens and Feingold, 2009). Although particle radiative forcing (both direct and indirect) is still recognized as a leading source of uncertainty in current estimates of climate sensitivity, anthropogenic aerosol particle precursor emissions and the resulting radiative forcing are not expected to increase considerably compared to current levels. This expectation, together with the ever-increasing levels of greenhouse gases (and associated warming) has spawned one view in the climate community that continuing efforts on aerosol and aerosol-cloud interactions in climate studies may be of secondary importance.

However, this projection neglects the dominant contribution of natural aerosol particles, and how a changing climate may alter their abundance and impacts on the Earth's energy balance. Furthermore, describing the impacts of the changing aerosol abundances over previous decades is essential to the accurate estimation of climate sensitivity and the predictive capability of climate models. In addition, many studies point to the pivotal role that reactive gases, aerosol particles and their interactions with radiation and clouds can have on atmospheric phenomena that span the range from the weather to climate scale.

Aerosol particles impact radiative forcing on regional signals that are not reflected on the global scale (Shindell and Faluvegi, 2009), such as impacts on the Asian summer monsoon, rainfall patterns in Southeast Asia, North Atlantic variability, the evolution of tropical cyclones, and the invigoration of precipitation and intensification of precipitation. The changing abundance of atmospheric particles over Europe and North America has been linked to changes in climate over these regions (Mickley et al., 2012; Philipona et al., 2009). Leibensperger et al. (2012) estimated that between 1970 and 1990, atmospheric particles resulted in an annual mean net cooling of the central and eastern United States by 0.5–1.0°C, offsetting some of the warming due to greenhouse gases, and an annual mean reduction in precipitation of 0.2 mm/day along the U.S. east coast. Atmospheric particles have also been linked to changes in storm tracks over the North Pacific and North Atlantic (Booth et al., 2012; Dunstone et al., 2013; Wang et al., 2014c; Zhang et al., 2007) and to changes in Hadley circulation in the tropics (e.g., Allen, 2015; Hwang et al., 2013; Ming and Ramaswamy, 2011). In addition, Bell et al. (2008) argued that satellite observations of rainfall over the Unites States show weekly variation in cloud top heights and precipitation, with a midweek maximum in afternoon rainfall and storm intensity over the southeastern United States that are correlated with particle loading in the atmosphere, suggesting a strong connection between weather and anthropogenic pollution.

Many studies have concluded that changing anthropogenic aerosol particles can shift the tropical patterns of rainfall, contributing to the Sahel drought (Held et al., 2005) and threatening the Amazon rain forest (Cox et al., 2008). Large anthropogenic aerosol particle abundances over

Asia appear to delay the onset of summer monsoons and are spatially shifting rainfall patterns in SE Asia (Gautam et al., 2011; Kuhlmann and Quaas, 2010; Lee et al., 2014). Aerosol particle-cloud interactions have also been suggested to modulate North Atlantic variability (Booth et al., 2012; Dunstone et al., 2013) as well as the Pacific Decadal Oscillation (Allen et al., 2014). Saharan dust outbreaks may influence the organization and evolution of tropical cyclones (Reale et al., 2014).

The indirect effect of aerosol particles on climate and weather depends on the microphysical properties of the particles. An increase in cloud condensation nuclei (CCN) results in a greater concentration of smaller cloud droplets, which increases the reflection of solar radiation and reduces surface heating. The particles also alter the precipitation efficiency of clouds by reducing the rate at which cloud drops coalesce into raindrops, resulting in less warm rain production and a longer cloud lifetime (Albrecht, 1989). Although some modeling studies have shown that atmospheric particles can suppress downwind precipitation (Changnon, 1981; Schmid and Niyogi, 2013), other studies have seen little impact from the particles and have ascribed this lack of impact to changes in meteorology (Carrió et al., 2010). Still other studies have shown that biomass burning particles invigorate convection and enhance tornado formation (Saide et al., 2015). Dust and bioparticles can change cloud microphysics by promoting the formation of ice, which in turn may alter the precipitation efficiency of clouds (Creamean et al., 2013). The range of cloud changes from modeling studies highlights the poor understanding of the influence of the composition and size of particles on the interactions between atmospheric chemistry, climate, and weather. This weak understanding translates into high uncertainty in the consequences of geoengineering through deliberate introduction of particles into marine clouds.

Changes in climate since the pre-industrial era have been linked to changes in the frequency of extreme weather events, which have substantial societal costs. Robust statistical formulations to describe accurately attributable risk due to human perturbation of the climate system for a single observed extreme weather event are difficult, even when the studied event is unprecedented in the historical record (Hansen et al., 2014). However, it is possible to estimate the increased likelihood of the occurrence of extreme events, such as hurricanes (Webster et al., 2005), heat waves (Stott et al., 2004), heavy rainfall (Lau and Kim, 2006; Lau et al., 2006), tropical and landfall typhoons (Wang et al., 2015b), floods (Milly et al., 2002), and drought (Pongracz et al., 2014), due to the changing composition of the atmosphere (Stott, 2015). Probabilities for heat waves in western North America and central Asia, cold outbreaks in eastern North America, and droughts in central North America, Europe, and central Asia are predicted to increase in the future (Screen and Simmonds, 2014), and the societal costs for some of these events can be quite high. It was estimated that there were more than 70,000 additional deaths in Europe in 2003, compared to a reference period of the five years prior, due to the severe heat wave that year (Robine et al., 2008). The changing atmosphere likely played a role in the severe heat wave in the southwestern United States in 2013 that caused hospitalizations, cancellation of flights, and deadly wildfires (Shiogama et al., 2014).

4.2 ATMOSPHERIC CHEMISTRY AFFECTS HUMAN HEALTH

Many trace gases and particles emitted to or produced in the atmosphere impact human health. Air pollution has been reported to negatively impact human physical and mental health for at least 800 years (Evelyn, 1661; Finlayson-Pitts and Pitts, 2000; Goodhill, 1971), with one

FIGURE 4.3 A policeman wearing a mask for protection against the thick smog in London, England in 1952. SOURCE: Getty Images.

of the most dramatic episodes being an estimated 13,500 excess deaths in the London smog of 1952 (see Figure 4.3). As indicated in Chapter 1, one out of eight deaths on a global basis is currently attributed to outdoor and indoor air pollution combined (WHO, 2014), and a recent study estimates 3.3 million premature deaths annually due to ambient air pollution worldwide (Lelieveld et al., 2015). The total cost due to health effects of some air pollutants in the United States is estimated to be between $71 and $277 billion each year, the majority of that resulting from detrimental health impacts and pre-mature deaths (Muller and Mendelsohn, 2007). Children are particularly susceptible to the effects of air pollution due to increased dosage through more rapid breathing and spending more time outdoors; they also have more susceptible metabolic, immune, and lung systems (EPA, 2013; Office of Environmental Health Hazard Assessment, 2015; WHO, 2005).

The U.S. Environmental Protection Agency has designated a list of hazardous air pollutants found indoors and outdoors that are known to be carcinogenic or cause other serious health effects ranging from eye and skin irritation to impacts on reproductive, neurological, and respiratory systems (see Figure 4.4).[11] Three gaseous criteria pollutants[12]—carbon monoxide, nitrogen oxides, and sulfur dioxide—which are directly emitted to the atmosphere (particularly from fossil fuel combustion), cause a range of detrimental health impacts. These include asthma

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[11] EPA Health Effects Notebook for Hazardous Air Pollutants: http://www3.epa.gov/ttn/atw/hlthef/hapindex.html.

[12] U.S. EPA has designated six common pollutants as “criteria” pollutants for which national air quality standards are set to protect people and the environment: ground-level ozone, particle pollution, lead, sulfur dioxide, carbon monoxide, and nitrogen dioxide.

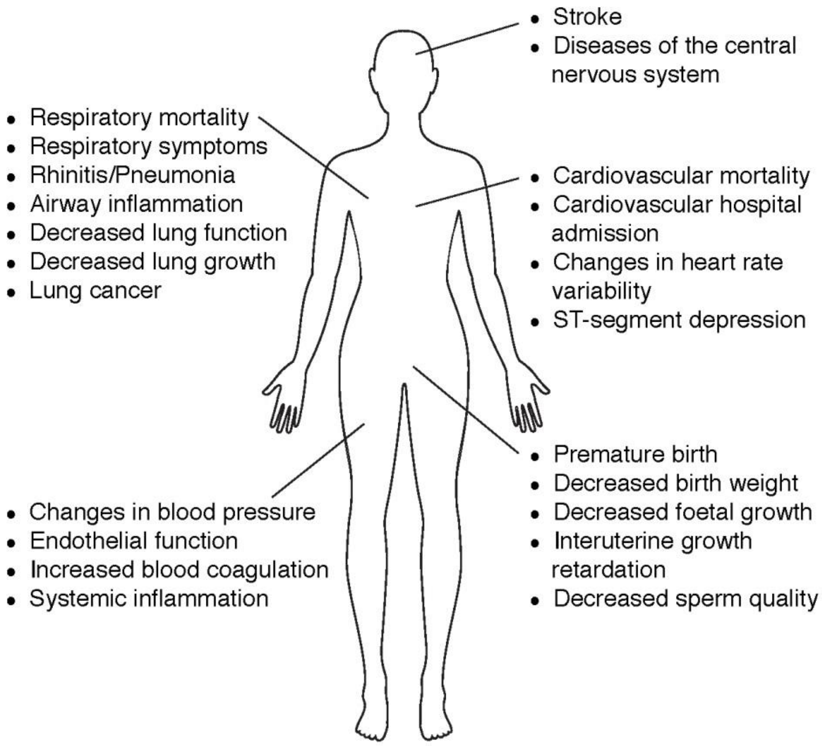

FIGURE 4.4 Diagram of the human organs that can be impacted by air pollution. Source: Peters et al., 2011.

symptoms, cough, cardiovascular, cardiac, and respiratory stress and mortality (Anderson et al., 2007; WHO, 2013). Ozone, which is created through the reactions of nitrogen oxides and volatile organic compounds in the presence of sunlight (see Chapter 2), also has well-known adverse health impacts (EPA, 2013), including premature cardiac and respiratory mortality (e.g., Jerrett et al., 2009b; Lipfert et al., 2006), increased hospital visits, asthma incidence and severity, decreased lung development in children (Gauderman, 2006; Office of Environmental Health Hazard Assessment, 2015; WHO, 2005, 2013), and possible impacts on reproductive (Olsson et al., 2013) and cognitive health (Chen and Schwartz, 2009; EPA, 2009).

Airborne particulate matter (PM) is also a criteria pollutant. Particles have a wide range of sizes and chemical composition depending on sources and removal processes from the atmosphere. Particulate matter has been linked to a range of health impacts, including asthma, cardiopulmonary disease, lung cancer, reproductive health problems (WHO, 2005), and increased mortality (Pope and Dockery, 2006; Pope et al., 2013; see Box 4.2). Recent studies suggest an association of atmospheric particles with neurodegenerative diseases such as Alzheimer’s and Parkinson’s, strokes, autism, and anxiety (Brauer, 2015; Calderon-Garciduenas and Torres-Jardon, 2015; Power et al., 2015; von Ehrenstein et al., 2014). It has been suggested that ultrafine particles (<100 nm in diameter) may have disproportionate impacts on health (Araujo, 2011a,b). Particles can have particularly adverse effects on children, including deficits in lung function development (Gauderman, 2006). Aggravation and increased incidence of asthma and airway inflammation and an increased risk of bronchitis and wheezing in children have been documented (Chen et al., 2015; WHO, 2005), and effects on neural development in children have also been suggested (Calderon-Garciduenas et al., 2014). Bioparticles also play a

BOX 4.2
Atmospheric Chemistry in the News: "Airpocalypse" in Beijing

Industrialization and urbanization have deteriorated air quality in China's major urban centers. In 2008 an air pollution monitor was installed atop the U.S. Embassy in Beijing; hourly fine particle (smaller than 2.5 micrometers in diameter, referred to as $PM_{2.5}$) concentrations were (and continue to be) tweeted from @BeijingAir (San Martini et al., 2015). This service was the first publicly available data reporting on air quality in China, revealing the severity of air pollution events in Beijing. Since 2010 air pollution events in China have been widely reported in the press. During these events, which have been described as "airpocalypses," fine particulate matter concentrations regularly exceed 200 $\mu g/m^3$—levels that put the population at elevated risk for cardiorespiratory disease. In 2012 China approved its first national environmental standard for $PM_{2.5}$, limiting 24-hour concentrations to below 75 $\mu g/m^3$, and has since invested billions of dollars to reduce pollution and achieve these targets (the World Health Organization recommends a maximum exposure of 25 $\mu g/m^3$ over 24 hours). The population of Beijing continues to regularly experience heavy smog events, including in December 2015 when Beijing issued the city's first "red alert" for air pollution, keeping school children and workers at home. Half of the city's vehicles were taken off the road and production at local factories was scaled back in an effort to cut emissions. The atmospheric chemistry research community continues to play a key role in understanding the specific sources and conditions that lead to these dramatic events and identifying how emission reductions will translate to improved air quality in Beijing and other growing urban centers.

FIGURE B Images of pollution in China. Top: the Forbidden City amid heavy smog in Beijing, China, 2005. SOURCE: Brian Jeffery Beggerly, Wikimedia Commons. Bottom: Smog in Chaoyang District, Beijing, January 2016. SOURCE: Suchi Chi, Wikimedia Commons.

major role in the transport of disease and have been linked with indoor health effects (Douwes et al., 2003; Fernstrom and Goldblatt, 2013). Recent studies have shown an association between atmospheric wind patterns and Kawasaki disease in three countries (Rodo et al., 2011). Thus, understanding the chemistry of atmospheric particles is central to understanding health effects.

A number of mechanisms have been proposed to link exposure to the health effects of particles. Reactive oxygen species (ROSs) are thought to play a key role in inflammation, pulmonary oxidative stress, vascular dysfunction, atherosclerosis and lung cancer (Araujo, 2011a; Hamra et al., 2014). ROS include free radicals such as OH, HO_2, superoxide anion, peroxynitrites, H_2O_2, and organic peroxides and can appear in both the gas and aerosol phases. The toxicity and health effects of particles thus also depend to a large and under-appreciated extent on atmospheric chemical transformations that depend strongly on aging processes in the atmosphere.

Exposures to higher levels of air pollutants can occur indoors, where people typically spend approximately 90 percent of their time (Nazaroff and Goldstein, 2015). A particular problem in the developing world is the emissions of gases and particles from biomass burning for cooking and heating (Lelieveld et al., 2015; Smith et al., 2014).

It is difficult to separate the effects of specific gases and particles on human health, and there are likely synergistic effects between gases and particles; in addition, some compounds are found in and move between the two phases. Further complexity arises from the fact that many of the gases and particles that cause significant health effects are not directly emitted, but rather are formed via atmospheric chemical reactions (Finlayson-Pitts and Pitts, 2000; Pandis and Seinfeld, 2006; Pöschl, 2005). Without knowledge of the nature and chemistry of gases and particles as well as how they are emitted, formed, and altered in the atmosphere from the atmospheric chemistry research community, reliable guidance for societal choices to minimize impacts on human health is not possible. These mechanisms place atmospheric chemistry at the fulcrum of connecting exposure to health effects and highlight the potential contributions of the atmospheric chemistry community to human health research.

4.3 ATMOSPHERIC CHEMISTRY INTERACTS WITH NATURAL AND MANAGED ECOSYSTEMS

Terrestrial and marine ecosystems support society through the production of food and energy, and play an important socio-cultural role in how we view our world. Ecosystems are unique, dynamic components of the Earth system that continually respond to changing conditions, including those in the atmosphere. Ecosystems rely on the atmosphere for uptake of life-sustaining elements such as carbon, oxygen, nitrogen, and trace elements through deposition processes and in turn contribute to the atmosphere and impact climate through the emission of gases and particles (Behrenfeld et al., 2006; Maher et al., 2010; Swap et al., 1992). As a result, changes in atmospheric chemistry affect key ecosystem services, including the health of forests, agricultural lands, and oceans. The interaction with atmospheric chemistry can influence the value of ecosystem services (de Groot et al., 2012) through the reliance on ecosystems for food production, the maintenance of wildlife and their habitats for biodiversity and sustainability, and the aesthetic effects of particulate matter on visibility in public lands (NRC, 1993).

Ecosystems are often described as natural or managed by human activity. Natural ecosystems support all life on Earth. Managed ecosystems such as croplands are manipulated by humans to increase and modify food and energy production, and they provide food, energy, and other essential and beneficial services (Millennium Ecosystem Assessment, 2005). Many gas and particle phase atmospheric compounds deposit from the atmosphere to the Earth's surface through wet or dry deposition. Ecosystem damage, for example through deposition of acidic compounds formed via fossil fuel combustion (e.g., Section 3.5) or the clearing of land for agriculture, can trigger a cascade of ecological effects through particle and trace gas exchange and atmospheric chemical reactions. Because natural and managed ecosystems are so closely tied to the atmosphere, atmospheric chemical reactions and chemical composition play a central role in understanding how ecosystems respond to global change and affect climate change. Therefore, there is a crucial need to understand how terrestrial and marine ecosystems contribute to, and are controlled by, the chemistry of the atmosphere.

While ecosystem productivity is dominantly a function of climate conditions and CO_2 concentrations, chemical reactions in the atmosphere create compounds that also influence ecosystem productivity. The exchange of these compounds (e.g., reactive nitrogen, organic compounds, and other inorganic compounds) between ecosystems and the atmosphere can potentially harm or benefit ecosystems, depending on the chemical compound. For example, oxidants such as ground-level ozone damage plant tissue and slow growth and other functions (Felzer et al., 2007); see Figure 4.5. Ozone can be taken up by plants, reducing the productivity of natural (Chappelka and Samuelson, 1998) and managed ecosystems such as agriculture (Booker et al., 2009; Fiscus et al., 2005). This can have significant impacts on crop production: for example, one study estimated that global cereal crop losses due to exposure to ozone in 2000 ranged from $11–$18 billion (Avnery et al., 2011).

Nutrients such as nitrogen and phosphorus deposited to the surface can benefit ecosystems, unless anthropogenic pollution drives an excess that may be harmful (e.g., acid rain deposition, eutrophication of aquatic systems). Nitrogen deposition can also alter ecosystem productivity by affecting biological diversity (Bobbink et al., 1998), nutrient cycling, and the rate

FIGURE 4.5 Comparison of healthy tall milkweed (*Asclepias exaltata* L.) leaves on the left and ozone-injured leaves on the right. Some plants species, such as tall milkweed, are especially sensitive to ozone. SOURCE: Bingham and Porter, 2015.

at which plants grow (Vitousek et al., 1997), all of which have substantial associated costs (Jones et al., 2014). A significant fraction of the global anthropogenic emissions of atmospheric nitrogen deposits on the ocean and alters new marine biological growth (Duce et al., 2008; Krishnamurthy, 2007), and the atmospheric deposition of mineral dust is an important source of the nutrients iron and phosphorus in many open ocean regions (Jickells et al., 2005; Mahowald et al.,2005, 2008a; see Figure 4.6). Toxic compounds, e.g., mercury, are known to deposit to terrestrial and marine ecosystems (Landis and Keeler, 2002). While much of the deposition of heavy metals such as mercury, lead, and cadmium does not directly affect productivity, it may influence the quality of food produced. Long range transport of dust also plays a role in adversely affecting the health of coral reefs due to the deposition of soil-associated fungal spores (Shinn et al., 2000).

To add complexity, many ecosystems play an active role in atmospheric chemical processes through their emissions of gases and particles, which can impact both tropospheric (e.g., volatile organic compounds [VOCs]) and stratospheric (e.g., nitrous oxide) composition and chemistry. Understanding this complex two-way exchange of constituents between the atmospheric reservoir and ecosystems is necessary to determine how atmospheric chemistry affects, and is affected by, ecosystems. As an example of this complexity, organic forms of carbon and nitrogen that are oxidized, reactive, and thermally labile are thought to be important in understanding the fluxes of carbon and nitrogen at the biosphere (soil, plant, ocean)/atmosphere interface. Measurement of these species is often difficult, but necessary to close the reactive carbon and nitrogen budgets.

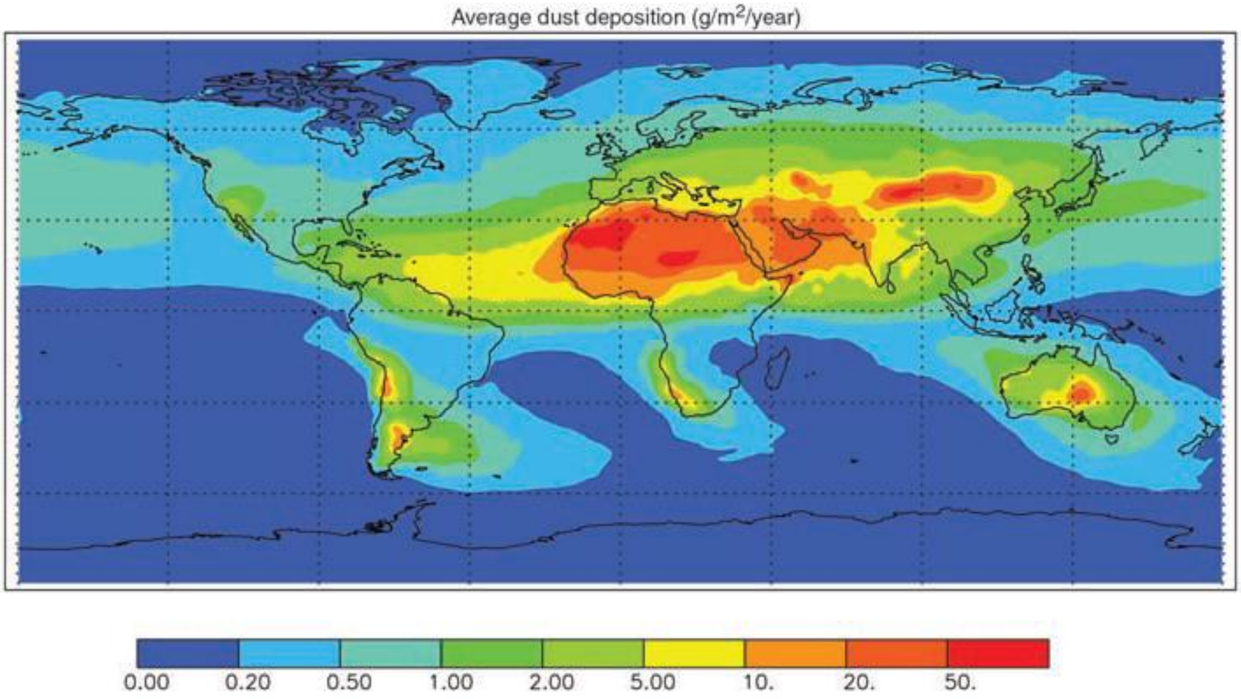

FIGURE 4.6 The long range atmospheric transport of mineral dust over and into the ocean provides an important source for the nutrients iron and phosphorus in many marine areas. SOURCE: Jickells et al., 2005.

In many regions of the world, atmospheric composition and chemistry are driven by terrestrial and marine ecosystems. Terrestrial vegetation emits a significant amount of VOCs (Guenther et al., 1995). Biologically produced VOCs are key contributors to tropospheric ozone formation and, depending on the oxidation pathway, VOC oxidation products form or grow secondary organic aerosol particles. Ecosystems also produce many types of bioparticles, ranging from plant debris, pollen, fungi, and bacteria that can oxide (NO) and these emissions comprise 15–25 percent of the industrial era NO_x in the global atmosphere (Holland et al., 1999). Terrestrial and marine ecosystems also emit nitrous oxide, an important greenhouse gas. Concentrations of nitrous oxide have been increasing in the industrial era due to fertilizer applications in managed ecosystems, and food production is estimated to account for 80 percent of the increase in atmospheric nitrous oxide (Ciais et al., 2013). The burning of terrestrial vegetation, either occurring naturally or ignited by humans, can inject large quantities of NO_x, carbon monoxide, and aerosol particles to the atmosphere, and these also play an important role in climate and air quality.

Similar to terrestrial ecosystems, the ocean is an important natural source of many trace gases and particles that can influence atmospheric chemistry. Marine emissions include greenhouse gases (e.g., nitrous oxide), halocarbons, VOCs, sea spray, bioparticles, primary organic carbon aerosol particles, and sulfur species such as dimethyl sulfide and organosulfates (Quinn et al., 2015). The impacts of ocean acidification on the processes that control the production of these species are potentially important but still uncertain (Gruber, 2011). It is generally believed that marine ecosystem emissions are less than terrestrial emissions, however far fewer measurements of reactive organic species have been made over the oceans so the uncertainties in marine emissions remain quite large (Carpenter and Nightingale, 2015; Shaw et al., 2010). Because these emissions occur in remote regions that might not be strongly influenced by terrestrial emissions, they can have an important impact on gas-phase chemistry and aerosol formation in these regions.

Ecosystems are increasingly affected by numerous perturbations and stressors, including land use changes, water pollution, and climate change. Habitat destruction, competition from invasive species, unsustainable exploitation of species for economic gain, and climate change have already increased extinction rates and affected species ranges and migratory patterns (Barnosky et al., 2012; Lenton, 2011; NRC, 2013). Because the impact of air pollution and deposition of nutrients and contaminants can be substantial, improved understanding of the interactions of atmospheric chemistry on natural and managed ecosystems is urgently needed.

4.4 SOCIETAL CHOICES AND IMPACTS

Societal needs involving health and welfare, economic and intellectual development, and many other aspects of lifestyle (beyond just food, clothing, and shelter) benefit from basic science, including atmospheric chemistry. Actions taken to achieve certain benefits have repercussions on the environment, and often result in changes in atmospheric composition. Atmospheric chemistry research can benefit societal development by attributing changes to specific aspects of development. Optimally, this basic scientific knowledge can point the way to alternatives and less damaging technologies. Following are several areas where the relationship between atmospheric chemistry and societal choices and impacts are particularly important

National Security

Threats to a nation's security that are potentially destabilizing can impact the U.S. military or diplomatic missions and thus present a concern for U.S. national security. The U.S. governmental entities interested in national security include the military, intelligence, international development, and diplomatic corps. Their concerns are global and include, *inter alia*, food and water shortages, pandemic disease, refugees, clashes over resources, and devastation by natural disasters. Most of these crises involve humanitarian emergencies with a strong health and welfare component. They may be exacerbated by slow environmental degradation from poor air or water quality, or they may involve destruction of societal function and governance by natural disasters such as earthquakes, tsunamis, volcanoes, floods, or hurricanes. A changing atmosphere not only directly affects human health, agricultural productivity, and the state of the environment, but is also a substantial contributor to climate change. Climate change may be shifting water resources as well as the frequency and strength of extreme weather such as hurricanes, storm surges and heat waves. The rapid changes in the Arctic are an example where climate change presents a challenge to U.S. national security (see Box 4.3). All of these changes can have important effects on national and global stability and security. As discussed above (see Chapter 4.1), research in atmospheric chemistry can improve the fidelity of climate models, which can improve the ability to anticipate these changes.

The science connecting national security and climate change is a growing research area (e.g., Brown, 1989; Busby, 2007, 2008; Smith, 2007). For example, a university-based program in Climate Change and African Political Stability was initiated in 2009 with a Department of Defense grant. The Center for Naval Analyses' Military Advisory Board of retired generals and admirals continues to point out the increasing risks for national security (CNA MAB, 2014). The Center for Climate & Security (also including ex-military and government leaders) publishes reports like *The Arab Spring and Climate Change* (Werrell and Femia, 2013), including essays such as "Chinese Drought, Wheat, and the Egyptian Uprising: How a Localized Hazard Became Globalized."

The U.S. government's recent interest in the significance of future climate change on national security began with the setting of new priorities for the intelligence community that specifically included climate change. To highlight this, a recent National Intelligence Assessment relied on the Intergovernmental Panel on Climate Change 4th Assessment Report as its primary source for climate science, which it then linked to the security aspects (Fingar, 2008). The importance of climate change for national security is treated in a number of recent reports and assessments, including a White House summary (2015) "The National Security Implications of a Changing Climate." Similarly, the National Intelligence Council (NIC, 2015) acts as a source of reports on water availability and climate. The Secretary of Defense recently released a Climate Change Adaptation Roadmap (DOD, 2014). There are a number of efforts to address these concerns. For example, the State Department initiated the Climate and Clean Air Coalition to Reduce Short-Lived Climate Pollutants[13] as an effort to support international development goals as well as to gain international partners, and the EPA recently announced the Clean Power Plan, which establishes final emission guidelines for states to follow in developing plans to reduce CO_2 emissions from existing fossil fuel-fired electric generating units. (However, this plan does not address other greenhouse gases such as nitrous oxide and methane.) All of these reports highlight

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[13] http://www.ccacoalition.org/2012.

BOX 4.3
The Rapidly Changing Arctic

The Arctic presents a clear example of where climate change—driven by increasing carbon dioxide as well as chemically controlled greenhouse gases like methane and ozone—presents a challenge to U.S. national security. The rapid retreat of sea ice to date, which is projected to continue to change in the future (see Figure C), and the overall atmospheric warming has damaged coastal infrastructures and made re-supply of bases via ice roads more difficult. An ice-free summer Arctic Ocean presents new opportunities for major shipping and exploitation of natural resources plus military incursions, not only by the Arctic Council nations but by other countries as well. The important role of chemically controlled black carbon and methane in Arctic change[14] has been recognized by the Arctic Council as well.

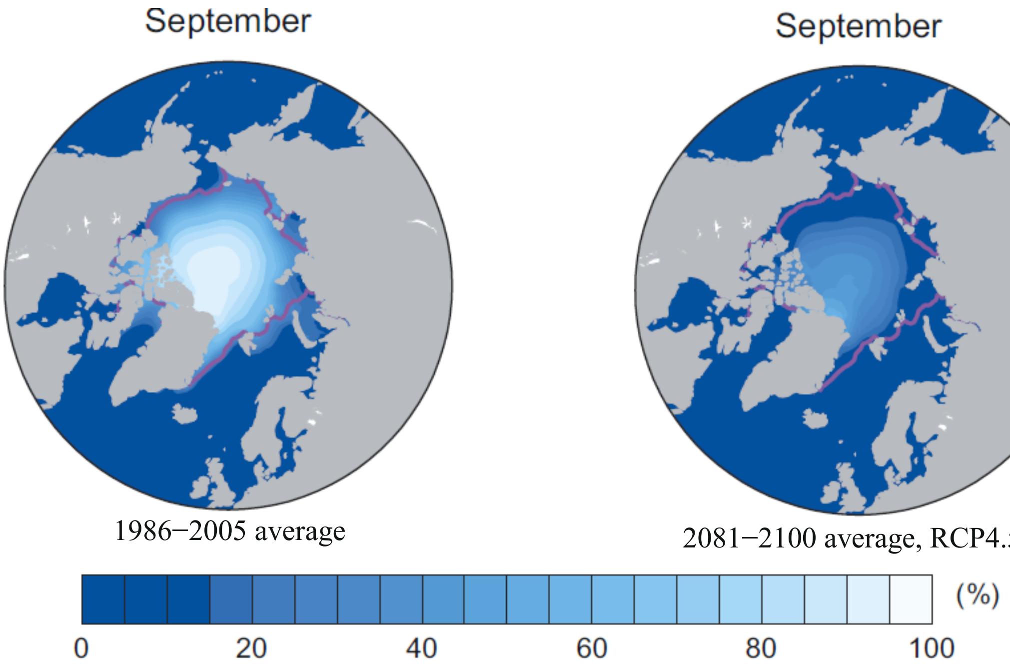

FIGURE C Present and future Arctic sea ice extent during the September minimum under the IPCC RCP4.5 future scenario (IPCC, 2013, Fig. 12.29).

the growing interest and concern of the U.S. government about the related issues of climate change and national security.

In addition to the climate-national security connection, atmospheric chemistry research is important for understanding the fates of chemical or radioactive contaminants on populations. For example, the dispersion of the Chernobyl fallout required accurate chemistry-transport modeling, and the chemical properties of warfare agents will determine their degradation and dispersion. It is important that the best current knowledge of atmospheric science is included in assessments of complex issues of national security, and as new research is identified that is vital

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[14] *Arctic Council Observer States 2015 National Reports on Enhanced Black Carbon and Methane Emissions Reductions:* According to the framework for “Enhanced Black Carbon and Methane Emissions Reductions” agreed on at the Arctic Council Ministerial in Iqaluit April 2015, each Arctic Council Observer State was invited to join the actions described in the framework including sharing a national report according to the guidance to be found in the framework. See http://hdl.handle.net/11374/1169.

to the understanding of national security, it is included as a central part of the atmospheric chemistry research portfolio.

Water Security

The availability of fresh water is central to human health and welfare, including food and energy production. In many regions of the world insufficient rainfall exists, which means irrigation must be used to water crops. Currently, 70 percent of global fresh water consumption is used for agriculture, and a 20 percent increase in water consumption for agriculture is expected by mid-century. Increased demand for fresh water for agriculture and livestock, together with other uses such as energy production, will add substantially to the current stress on non-renewable groundwater sources. The water cycle is already changing due to changes in Earth's climate driven by increasing greenhouse gas concentrations. For example, the amount of rain that falls during the most intense 1 percent of storms has increased by almost 20 percent over the past 50 years (USGCRP, 2009). Some regions of the world are in the midst of historic droughts. In 2015 alone, ten weather and climate disaster events occurred with losses exceeding $1 billion each across the United States, including drought and flooding events.[15]

Atmospheric chemistry plays a key role in impacting the water cycle largely through serving as the conduit for the formation and transport of aerosol particles which act as cloud condensation and ice nuclei (CCN and IN) (see Figure 4.7). The dynamics of atmospheric transport, cloud formation, weather patterns, and precipitation processes, all play a role in controlling the delivery of fresh water to the Earth's surface.

Through their ability to heat or cool the atmosphere and seed clouds, atmospheric aerosol particles have been shown to play a critical role in affecting the amount, type, and location where precipitation occurs (see Chapter 3.5). It has been suggested that in regions with high levels of pollution, the amount of precipitation can be reduced due to the presence of too many cloud nuclei that form drops small enough to inhibit rain formation (Rosenfeld et al., 2008). In contrast, certain aerosol particles can form ice in mixed phase clouds which can lead to increased amounts of precipitation (Ault et al., 2011; Muhlbauer and Lohmann, 2009). Increases in particles that act as ice nuclei may have a proportionally larger effect on precipitation than corresponding changes in cloud condensation nuclei (Rosenfeld et al., 2008), although this largely depends on the thermodynamic and dynamic state of the clouds in question. Only a small portion of the available moisture in clouds is transformed into precipitation that reaches the surface (Bruintjes, 1999). Thus, depending on their sources and composition, aerosol particles can lead to a re-distribution of water resources with more extreme events such as flooding and drought. This shift translates into less water in regions where reservoirs can capture the water with more runoff in other regions which lead to poor water quality. The overall effects of aerosol particles on precipitation formation and redistribution are highly uncertain (Huntington, 2006), highlighting the need for improved understanding of the sources and properties of aerosol particles that impact precipitation (see Figure 4.8).

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[15] Billion-Dollar Weather and Climate Disasters: http://www.ncdc.noaa.gov/billions/events.

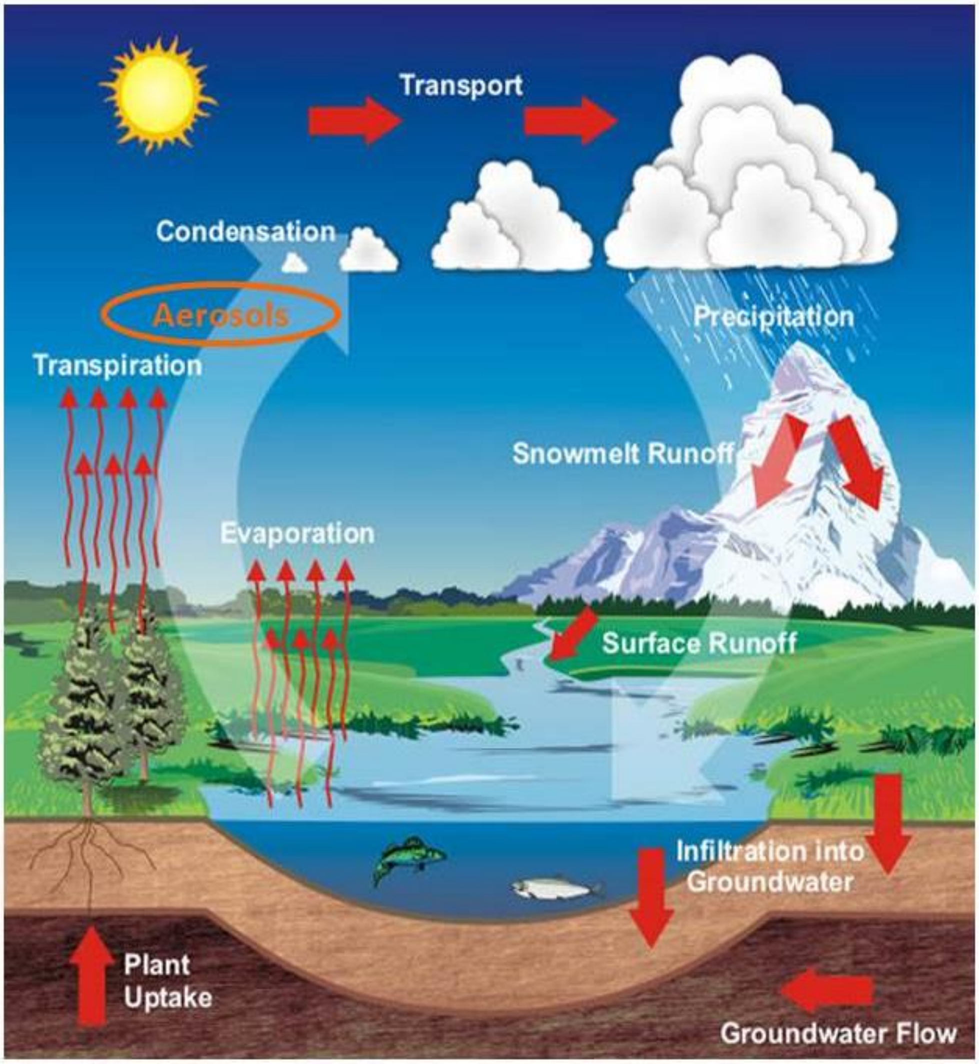

FIGURE 4.7 Schematic of the water cycle highlighting aerosol particles as a key step affecting cloud formation, lifetime, and precipitation efficiency. SOURCE Modified from http://www.srh.noaa.gov/crp/?n=education-watercycle.

Energy and Industry

Energy and industry are a core part of societal growth and development. The world's energy demands have continually evolved with expanding population and advancing technology. Energy sources, from biofuel to fossil fuels to alternative energies, have broadened as the global energy demand has expanded. The exploration and development of energy sources usually creates waste, both in the atmosphere and in water, that degrades the human and natural environment. Further, the generation of energy, particularly from fossil fuels, has been notoriously polluting, thus inspiring development of systems that are designed to result in clean air and clean water (e.g., U.S. Clean Air Act of 1963 [P.L. 88-206, 77 Stat. 392]; U.S. Clean Water Act of 1972 [P.L. 92-500, 86 Stat. 816]). Since the Los Angeles experience with photochemical smog (Haagen-Smit, 1952; see Chapter 2), society has come to realize that the products of combustion, as well as solvents and other chemicals released by industry, can rapidly pollute the air, making it unhealthy, degrading visibility, and in general reducing urban and even rural amenities.

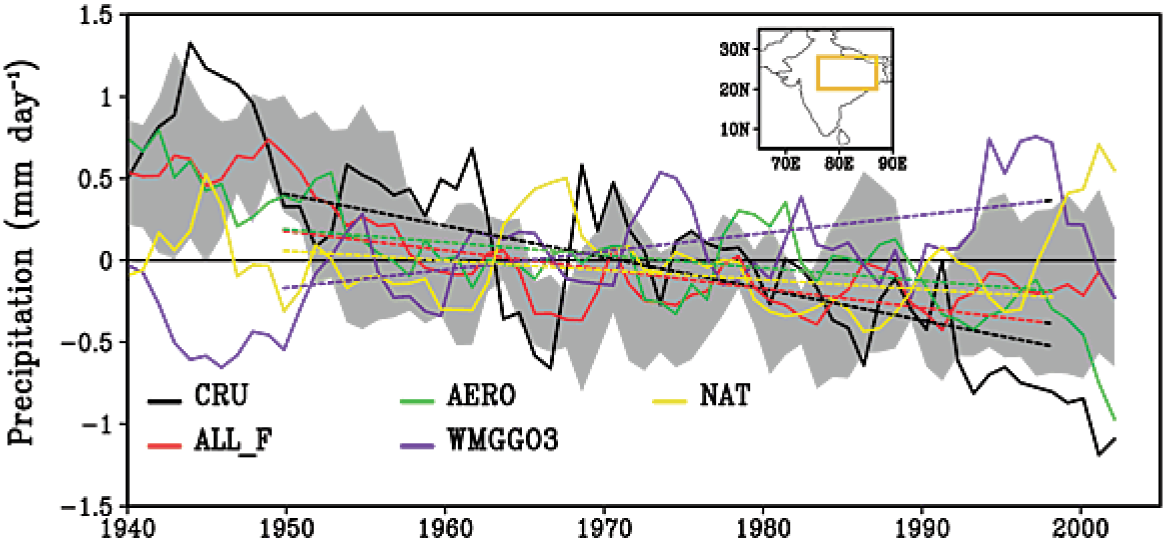

FIGURE 4.8 Five-year running mean June-September average precipitation anomalies (mm day-1) over central-northern India (orange box in the map). Anomalies are calculated as deviations from the 1940-2005 climatology. The black line is based on an observational dataset (Climate Research Unit TS 3.0 [CRU]). The red, green, blue, and yellow lines are the ensemble-mean for all-forcing (ALL_F), aerosol-only (AERO), greenhouse gases and ozone-only (WMGGO3), and natural forcing-only (NAT) CM3 historical integrations, respectively. The grey shades represent the standard deviation of the 5-member all-forcing ensemble. The least-squares linear trends during 1950–1999 are plotted as dashed lines in the respective colors. The observed precipitation decrease can be attributed mainly to human-influenced aerosol emissions. The drying is a robust outcome of a slowdown of the tropical meridional overturning circulation, which compensates for the aerosol-induced energy imbalance between the Northern and Southern Hemispheres. These results provide compelling evidence of the prominent role of aerosol particles in shaping regional climate change over South Asia. SOURCE: Bollasina et al., 2011.

Fortunately, an understanding of energy and industrial emissions combined with research on atmospheric chemical reactions and removal processes have enabled society to clean up the air in many urban areas (see Chapter 2). Increased future demands for energy and the associated industrial activities will result in increased requirements for the control of emissions of gases and particles from these processes.

The buildup of CO_2 and methane from fossil fuels is a major contributor to anthropogenic climate change (IPCC, 2014). For example, Paulik et al. (2015) have shown increased emissions of methane and other gases such as ethane (see Figure 3.1) as well as increased levels of atmospheric polycyclic aromatic hydrocarbons in the vicinity of hydraulic fracturing (“fracking”) activities. As discussed in Chapter 3.1 research in atmospheric chemistry can improve the understanding of trends in emissions.

The chemical industry has developed elegant technologies like the synthetic chlorofluorocarbons that appeared at first benign but were later tied to stratospheric ozone loss with associated increases in skin cancer, and more recently, climate change (IPCC, 2014; Seinfeld and Pandis, 2006). Research in atmospheric chemistry must keep pace with the magnitude of development and new technologies in the energy sector and other industrial sectors in order to best protect environmental services.

Environmental Justice

Environmental Justice is defined by the U.S. Environmental Protection Agency as the "*fair treatment* and *meaningful involvement* of all people regardless of race, color, national origin, or income with respect to the development, implementation, and enforcement of environmental laws, regulations, and policies."[16] Systematic integration of perspectives from all communities will enable management of the Earth system in a way that is beneficial for all its inhabitants. A closer linkage of atmospheric chemistry research to those using the information and directly affected by the pollution will strengthen the research program and make it more useful to policymakers and communities. Some important principles follow.

The issue of fair treatment is based on the fact that societal impacts of changing atmospheric composition like climate change are not distributed equally. Studies of these distributional effects find that negative environmental impacts tend to disproportionately fall on low-income populations (Brown, 1995; Jerrett et al., 2001; Morello-Frosch and Jesdale, 2006). Environmental justice studies examine the physical, social, economic, and equity implications of impact distribution, rather than just average effects across an entire population. Atmospheric chemistry research can contribute to a greater understanding of environmental justice by employing low-cost, widespread monitoring and high-resolution modeling to characterize the spatial distribution of pollution and its impacts, and by integrating this analysis with socioeconomic characterization. The unequal distribution of poor air quality should be a key part of assessments of future development and climate change.

The issue of meaningful involvement is based on a growing recognition that involvement and integration of local knowledge and perspectives is a key component of sustainable development (e.g., Cash et al., 2003). Some approaches to broadening perspectives are termed community-based participatory research (Minkler et al., 2010) or working with indigenous knowledge (Grenier, 1998). These approaches seek to identify and integrate principles and practices embedded in local context and culture, rather than those defined more globally by the research community. For example, communities may be aware of impacts of degraded air quality on populations and ecosystems, or how local actions alter emissions or exposure, in ways not apparent to the research community (Gonzalez et al., 2011; Nasir et al., 2014). Community engagement may provide key knowledge about indoor air pollution, which is affected by individual or community practices of smoking, cooking, and related activities. Indigenous knowledge can also describe the conditions and practices that govern tropical deforestation and biomass burning. Integration of scientific and community knowledge can provide improvements in both communities' quality of life and in scientific understanding of atmospheric chemistry.

To understand community-scale pollution, impact, and response, and hence facilitate advances, key information is found in non-academic knowledge systems. An open question is—how can such collaborations be fostered? While collaborative research is often motivated by knowledge generation and scientific reputation, such benefits may not be attractive to participants outside the traditional research community. Guidance for ensuring collaborations are beneficial to both sides is needed. Furthermore, innovation and advances may be found in disciplines such as psychology, sociology, and ethics, which are farther removed from atmospheric chemistry than typical partnerships such as Earth system modeling.

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[16] Environmental Justice: https://www3.epa.gov/environmentaljustice/.

Sustainable Development

Development almost always comes with industrialization, transportation, and the expansion of agriculture that pollute air and water, bringing ill health and poor living conditions. An understanding of the chemistry of this pollution has proven essential to designing healthy and sustainable communities in a changing world. Sustainable development thus involves atmospheric chemistry.

In September 2015, the United Nations released a list of 17 Sustainable Development Goals (SDGs), each supported by quantitative and qualitative targets to be met by 2030 (United Nations, 2015a). The primary obstacles to attaining the SDGs are implementation, funding and commitment rather than a lack of research. Nevertheless, many of the goals were developed from intricate geoscience knowledge in general, and atmospheric chemistry in particular. SDGs in which atmospheric chemistry plays a role are summarized in Box 4.4. The specific contributions of atmospheric chemistry research to each goal are discussed in the text below, where the targets relevant to both atmospheric chemistry and the overarching Sustainable Development Goals are identified.

Atmospheric chemistry research has an important role to play in sustainable development through minimizing environmental risks from exposure, preparing for extreme events, demonstrating lower-impact societal pathways and through international cooperation.

Minimizing environmental risks from exposures: Understanding the sources, fate and transport of chemicals in the atmosphere can lead to better identification of hazards and management of releases (Goal 12) and a reduction in deaths and illnesses from hazardous chemicals and air pollution (Goal 3). Indoor air pollution is a major cause of preventable deaths of children under five years old (Goal 3), a hazard that will be reduced by providing modern energy (Goal 7).

Preparing for extreme events: The SDGs call for reducing the exposure of vulnerable populations to extreme events, which is often related to poverty (Goal 1), developing food production systems resilient under climate change (Goal 2), and strengthening resilience to climate-related hazards (Goal 13). They also promote managing national and global health risks with early-warning systems (Goal 3). Atmospheric chemistry, as part of Earth system models, can provide predictive understanding of how energy consumption and agricultural activity affect the pollution of the atmosphere, land, coastal and blue waters.

Demonstrating lower-impact societal pathways: The SDGs recommend several pathways that will affect human environments and the Earth system. These include better air quality and safer public transport in cities (Goal 11), where concentrated pollutants are now generated. They also include greater emphasis on renewable energy and energy efficiency (Goal 7), and industry retrofits to achieve efficiency (Goal 9). These trends can be included in future scenarios of air pollutants and greenhouse gases to demonstrate environmental costs and benefits of each course of action. Scientific characterization of the Earth system is part of ensuring "that people everywhere have the relevant information and awareness for sustainable development and lifestyles in harmony with nature" (Goal 12).

International cooperation: In order for scientific research to proceed in tandem with and be useful for sustainable development, scientific partnerships must cross national boundaries, and scientists, practitioners, and policy-makers must integrate their disparate forms of knowledge. Participatory decision making (Goal 16), enhanced knowledge sharing, and capacity building (Goal 17) are all part of cooperative international initiatives such as Future Earth.

The ambitious goals charted in "Transforming Our World—The 2030 Agenda for Sustainable Development" require social, economic and political support. Progress and cooperation in science—including atmospheric chemistry—are also integral to success.

BOX 4.4
Sustainable Development Goals (United Nations, 2015) Involving Disciplinary Knowledge of Atmospheric Chemistry

1. End poverty in all its forms everywhere
2. End hunger, achieve food security and improved nutrition and promote sustainable agriculture
3. Ensure healthy lives and promote well-being for all at all ages
7. Ensure access to affordable, reliable, sustainable, and modern energy for all
9. Build resilient infrastructure, promote inclusive and sustainable industrialization and foster innovation
11. Make cities and human settlements inclusive, safe, resilient and sustainable
12. Ensure sustainable consumption and production patterns
13. Take urgent action to combat climate change and its impacts
16. Promote peaceful and inclusive societies for sustainable development
17. Strengthen the means of implementation and revitalize the global partnership for sustainable development

4.5 CONCLUSION

Human choices are affecting the chemistry of the atmosphere now and are certain to do so in the future. As a scientific community, we are beginning to understand the impacts that changes in atmospheric chemistry have on pressing societal issues such as climate and weather, human health, and natural and managed ecosystems. These impacts have resulted not only in environmental degradation, but also the loss of billions of dollars to the global economy. As society makes choices that will affect the atmospheric environment, preparing for future quality of life needs to include a predictive framework that is sufficiently robust to generate predictions about the environmental impacts of those choices—whether local, regional, or global. In turn, those predictions need to be based on fundamental knowledge of atmospheric chemistry. Refining such an effective predictive capability for tomorrow is an ultimate goal of atmospheric chemistry research.

Chapter 5—Anticipating Tomorrow: Research Priorities in Atmospheric Chemistry

As discussed in the preceding chapters, atmospheric chemistry has become a robust scientific discipline. The fundamental science of understanding atmospheric chemistry can contribute to both core knowledge of the way the planet works as well as to many issues that directly relate to societal challenges. Thus, in Chapter 4, the Committee identified several issues central to human health and welfare where atmospheric chemistry research can make important contributions in the coming decade. The goal of such research is improved predictive capability to be able to inform policy decisions about these issues.

In addressing its task, the Committee has identified priority areas of atmospheric chemistry research that need intensive attention in the coming decade. The next section (see Chapter 5.1) describes the Committee's process for developing these priorities, and the following section (see Chapter 5.2) lists five Priority Science Areas identified by the Committee.

5.1 DEVELOPING PRIORITIES

As described in Appendix B, the Committee engaged in an expansive process for gathering input from the atmospheric chemistry community. Using this community input as its underlying basis, the Committee deliberated extensively and ultimately chose five Priority Science Areas that it believes will drive atmospheric chemistry research over the next decade. In choosing these areas, the Committee prioritized potential research areas based on two criteria—scientific imperative and societal relevance. Scientific imperative describes research that is central to enabling substantial progress towards addressing important fundamental questions in atmospheric chemistry. This includes laboratory experiments, theory, field and modeling research that advances frontier knowledge in atmospheric chemistry. This criterion incorporates important issues within atmospheric chemistry that have already been identified but where substantial further research is needed.

Much of the current research in atmospheric chemistry relates in various ways to the health and well-being of human society and the natural world, including understanding impacts on ecosystems, agriculture, human health and the economy. In an increasingly globalized world, research that can address current and project future societal and ecological problems is increasingly valued as an essential component of the development of effective public policies by a broad segment of society, including Congress and the public. The Committee therefore considered societal relevance to be an important criterion in choosing their Priority Science Areas.

The Committee has given priority to closing key science gaps that are impeding the development of atmospheric chemistry predictive capability and to building collaborations with other disciplines to solve urgent societal and environmental issues. The emphasis is on the science of atmospheric chemistry, not the engineering applications of the science. While the Committee recognizes the breadth of science that is important in addressing these issues, we focus on just five Priority Science Areas. Within each Priority Science Area, the Committee identified key scientific gaps, chosen to be at a level of effort that could be addressed with funding through the National Science Foundation (NSF) proposal process. These key scientific gaps were chosen based on their necessity for addressing the Priority Science Area under which they fall and on their transformative potential, which is a central tenet of research supported by the NSF. By transformative potential, the Committee refers to higher-risk and higher-reward research areas that have the potential to dramatically change the understanding of atmospheric processes. There is, by the nature of the problems, some overlap of key scientific gaps from one Priority Science Area to another, particularly between the foundational and societally relevant areas.

5.2 PRIORITY SCIENCE AREAS

Following the process described in the preceding section (see Chapter 5.1), the Committee identified five priority areas for scientific research in atmospheric chemistry. The first two Priority Science Areas are necessary for building the foundation of atmospheric chemistry, aimed at providing further growth in understanding of how the atmosphere works. The next three Priority Science Areas directly address major challenges facing society, for which advances in atmospheric chemistry are required to make progress.

In total, these Priority Science Areas cover a broad range of research questions. More specific areas that represent key scientific gaps are described within each area, along with examples of actions needed to address those gaps. All examples are intended to be illustrative, and not exclusionary or indicative of priorities within each area.

Priority Science Area 1

Advance the fundamental atmospheric chemistry knowledge that enables predictive capability for the distribution, reactions, and lifetimes of gases and particles.

Motivation

Predictive capability starts with a fundamental understanding of the atmospheric chemistry occurring now. While some predictions can be made with confidence using current understanding, there remain major gaps and inconsistencies in the understanding of fundamental atmospheric chemical processes. These major gaps need to be closed and inconsistencies resolved.

Fortunately, current advances in atmospheric chemistry are enabling atmospheric chemists to identify and begin to narrow the gaps and to resolve discrepancies in understanding the basis of today’s atmospheric composition. This progress is being driven by a combination of laboratory experiments, theory, modeling, and observational capabilities. As climate and other conditions continue to change, developing new and quantitative understanding of the fundamental underlying chemistry will be essential for proactively exploring and defining future chemical and dynamical regimes.

Actions to Address Key Scientific Gaps

A. Quantify reaction rates and understand detailed chemical mechanisms in multi-pollutant and multi-phase environments that cover the chemical and dynamical regimes from polluted urban to natural remote regions.

    There are many important problems in atmospheric chemistry that fall under this category, the background for which is summarized in Chapter 3. A few illustrative (but not exclusive) examples out of many important needs include quantum chemistry studies of reactions of organic radicals that may not be possible to address with experimental methods (see Chapter 3.2); chemical species and factors controlling the formation, growth and aging of aerosol particles (see Chapters 3.5, 3.6, 4.1, 4.2, and 4.3); and the role of water in mediating chemistry in the gas phase, in particles and on surfaces (see Chapter 3.2).

B. Identify and quantify important atmospheric oxidants or other reactants that lead to transformation and removal of chemical species from the atmosphere across broad spatial and temporal scales.

    Examples include quantification of the gas phase oxidative capacity across broad spatial and temporal scales (see Chapter 3.3); developing improved understanding of what controls OH reactivity in different environments (see Chapters 3.2 and 3.3), understanding instrumental biases in measurements of oxidants (see Chapter 3.3), and identifying and quantifying important individual oxidants in the condensed phase and their role in determining the composition and chemistry of particles, fogs, and clouds (see Chapters 3.3 and 4.2);

C. Develop a stronger understanding of the influences that heterogeneous chemistry exerts on tropospheric composition.

    Examples include identifying and quantifying changes in the spectroscopy and photochemistry of species (organic and inorganic as well as neutral species and ions) in surface films compared to the gas or bulk phases; elucidating reaction mechanisms and products of organic species in thin films under different conditions and mixtures of co-pollutants, including trace metals; and quantifying the partitioning and exchange of species between the gas phase and surface films due to physical and chemical processes.

D. Understand and quantify the influence of the coupling between chemical and meteorological processes on the distribution of trace constituents in the troposphere.

    As discussed in Chapter 3.4, atmospheric composition and chemistry is the result of chemical, physical and meteorological processes whose coupling needs to be understood to develop predictive capabilities. Examples include obtaining a better understanding of the transformation and transport of chemical constituents in convective systems; quantifying the role of meteorology on the frequency of air pollution stagnation events; and quantifying how different forms of convection affect lightning-generated NO_x production and subsequent ozone production.

E. Understand and quantify the influence of the coupling between chemical, dynamical, and radiative processes involving stratospheric chemistry.

As discussed in Chapters 2.1 and 3.4, coupling between the troposphere and stratosphere has consequences for atmospheric chemistry and composition in both regions of the atmosphere. Examples include understanding how changes in the Brewer-Dobson circulation, driven by changes in ozone and the well mixed greenhouse gases, will affect the distribution of stratospheric species; quantifying the impact of deep convective transport of tropospheric water vapor and other tropospheric constituents on the budgets of chemically and radiatively important species in the stratosphere; and quantifying how transport of stratospheric ozone will impact tropospheric ozone in a changing climate.

Approaches and Support Needed

- Develop the next generation of accurate, sensitive, and specific measurement capabilities for atmospheric constituents in single-phase and multi-phase environments and on surfaces. This includes development and interpretation of observations from miniaturized, low-power sensors for deployment in networks and on a variety of platforms for sampling a wide range of chemical and dynamical regimes. An important part of this effort is instrument and technique validation.
- Enhance the integration of laboratory, computational chemistry, field and chamber studies, and photochemical and multiphase modeling.
- Mine existing high-quality atmospheric chemistry field and environmental chamber data sets to probe chemical mechanisms, to examine trends in time, and to compare the chemistry across different chemical and dynamical regimes.
- Accelerate the trend of developing and including more realistic atmospheric gas-phase, aqueous-phase, and multi-phase chemistry in next-generation models.
- Explore the interconnection of stratosphere-troposphere atmospheric chemistry using a combination of in situ and remote sensing techniques in combination with models.

Priority Science Area 2

Quantify emissions and deposition of gases and particles in a changing Earth system.

Motivation

Emission and deposition processes govern concentrations and spatial distributions of gases and particles in the atmosphere. A quantitative understanding of these distributions is key for assessing the impacts of atmospheric processes on human and ecosystem health, weather and climate. Although recent advances have reduced uncertainties, current shortcomings in predicting emissions from both natural and anthropogenic sources remain an issue for developing a predictive capability for atmospheric chemistry. Research is needed to both reduce these uncertainties for known sources and constrain emissions of poorly understood constituents (e.g., bioparticles) (Burrows et al., 2009a; Despres et al., 2012). Similarly, deposition processes for many species are not well understood or quantified, which is important not only for understanding atmospheric composition but also for elucidating atmospheric impacts on ecosystems. The net flux of species to or from the atmosphere is the combination of emissions and deposition. To develop a truly predictive understanding of the net flux, the factors controlling emissions and deposition must both be known.

The global environment is rapidly changing. Sources of atmospheric constituents change annually as humans make new decisions about technology, energy systems, pollution control, and land use. The global geographical distribution of human-derived emissions is also changing, with increases in the developing world and decreases in response to regulatory policies in developed nations. Natural sources respond to meteorological conditions and longer-term changes in climate. Processes that remove atmospheric constituents are also subject to such changes with time. Understanding emission and removal processes that determine atmospheric composition is a key component of modeling not only today's Earth system, but also a future system that reflects the outcome of policy choices. Quantification of these processes and the feedbacks that occur, for example in response to changing climate, is thus central to the development of a predictive capability.

Actions to Address Key Scientific Gaps

A. Better determine emissions from both anthropogenic and natural sources and their spatial and temporal variations and trends.

As discussed in Chapters 3.1 and 3.5, there are many important issues that need to be addressed to better define various aspects of emissions into the atmosphere. Examples include determining the impacts of pollutants on emissions from the terrestrial and marine biosphere (see Chapter 3.5); quantifying trends in emissions from different regions of the world and their impacts on atmospheric composition and chemistry over scales from local to global (see Chapter 1.1); and developing reliable approaches to integrating "top down" and "bottom up" results for emission inventories (see Chapter 3.1).

B. Identify mechanisms and measure rates by which wet and dry deposition removes aerosol particles and trace gases from the atmosphere.

Examples include determining deposition mechanisms and rates for species where data are sparse but where deposition could have a significant impact on atmospheric composition and ecosystems (see Chapters 3.6 and 4.3); determining synergistic interactions that can change deposition processes for a species from that measured for the single compound alone, and the mechanisms involved (see Chapters 3.6 and 4.3); measuring and characterizing how trace gases are taken up by vegetation, including canopy scale processes; and characterizing how hydrometeors scavenge aerosol particles.

C. Determine the role of meteorology, including temperature, precipitation, and extreme events, on emissions and removal of atmospheric species.

As discussed in Chapter 3.4, there are close couplings and synergies between atmospheric composition and chemistry, and atmospheric dynamics and meteorology that impact human health and welfare (see Chapters 4.1 and 4.2). Examples in this area include evaluating how changes in precipitation rates and locations alter the removal of species from the atmosphere (see Chapters 3.4 and 3.5); determining how dry deposition is affected by changing wind speed, temperature, and humidity in a changing climate (see Chapter 3.6); and measuring how natural emissions such as bioparticles, BVOC, soil NO_x, and methane are impacted by changing meteorology.

D. Determine the role of global change and societal choices (including changes in climate, energy choices, and land use) on the emissions and removal of atmospheric species.

As discussed in Chapters 1.1, 3.1, and 4.4, atmospheric composition and chemistry is inextricably intertwined with choices made by society and by global change. Examples include understanding how agricultural activities impact emissions and removal of atmospheric constituents following conversion of lands to agricultural use; and determining how energy choices influence extraction methods and the resulting emissions.

Approaches and Support Needed

- Develop instrumentation and measurement strategies to quantify fluxes.
- Support long-term measurements, especially those of biosphere-atmosphere exchange, including over the oceans, which remain relatively under-studied compared to land.
- Develop integrated strategies that employ both models and observations to attribute atmospheric concentrations to particular sources.
- Data mine measurements (e.g., concentration ratios) that can help constrain emissions and trends. Connected to this is the need to further develop data archiving resources.
- Develop and test model parameterizations for deposition processes.
- Coordinate integrated assessment with agricultural and ecological communities.

Priority Science Area 3

Advance the integration of atmospheric chemistry within weather and climate models to improve forecasting in a changing Earth system.

Motivation

As described in Chapter 4.1, all greenhouse gases and atmospheric particles impact the Earth's radiation budget and dynamics of the atmosphere, affecting weather in such ways as changing precipitation patterns and monsoon circulations. Aerosol particles, in particular, play a critical role through their influence on the growth and formation of clouds and precipitation. In global climate models, the effect of an increase in atmospheric aerosol particle concentrations on radiation and the distribution and radiative properties of the Earth's clouds is the most uncertain component of the overall global radiative forcing. Changes in atmospheric dynamics and circulation that are linked to changes in atmospheric composition (e.g., precipitation patterns, monsoon circulations) are even more uncertain than radiative forcing. During the past decade of intensive aerosol-cloud-climate research, some scientific gaps have been closed, and additional processes have been identified that still elude quantification. As with many complex systems in intermediate stages of understanding, this progress has not yet reduced the overall magnitude of uncertainty, leaving major deficiencies in the ability to project future climate (Seinfeld et al., 2016).

The chemical reactions involving aerosol particles and gases determine not only particle formation but also the processes by which climate-relevant trace species are removed from the atmosphere. Atmospheric chemistry, therefore, remains the crucial component that allows estimates of the atmospheric lifetimes of many species, and consequently the ability of pollution to accumulate in the atmosphere and thus influence climate and weather.

Some studies of regional climate change (seasonal-to-interannual or longer-term projections) or weather forecasting have included heterogeneously distributed aerosol particles or ozone in their models, but many have not. Many climate models have adopted a cloud-aerosol particle microphysical model without using the proper aerosol mixing state or chemical processes that create and alter aerosol particles. Most anthropogenic aerosol particles are initiated with gas-phase emissions followed by thermal and photochemical reactions in either the gas phase or in clouds or aerosol particles (Bauer et al., 2013; Seinfeld et al., 2016).

The fundamental understanding of atmospheric chemistry impacts on climate need to be improved to allow predictions to be made of those aspects of the changing climate structure that most urgently threaten human health, security, economic opportunity, social stability, and confidence in the future. Thus the atmospheric chemistry community needs to continue to work with the climate and weather research community in several major areas so that knowledge of the many roles that atmospheric composition plays in climate and weather can be built into climate models.

Actions to Address Key Scientific Gaps

A. Determine the global distributions and variability of atmospheric trace gases and aerosol particles, and better understand their climate-relevant properties.

   Some examples of the many important research areas to be explored to better understand how greenhouse gases and aerosol particles impact climate include quantifying how aerosol particle composition, size, mixing state, and morphology determine their radiative properties (see Chapter 3.5); and determining the temporal, regional, and vertical distribution of short-lived climate forcers (see Chapter 1.2).

B. Understand the role of aerosol particles as a modulator of cloud microphysics and precipitation efficiency in natural and anthropogenically-perturbed environments.

   As discussed in Chapter 3.5, there are numerous aspects of aerosol-cloud interactions which need to be better understood for constraining the impacts of aerosol particles on weather and on climate via their impacts on clouds. Examples include determining which aerosol particle sources most effectively seed clouds and lead to enhanced precipitation; and determining how the vertical structure of different aerosol particle sources influences clouds and precipitation processes.

C. Develop accurate descriptions of the complex chemical and physical evolution of atmospheric constituents that can be implemented in models for robust prediction of the impact of the chemical state of the atmosphere on climate and weather.

> As discussed throughout this report, developing a predictive understanding of atmospheric chemistry impacts on climate and weather will rely on developing accurate model descriptions of observed processes. Examples include further developing methods to chemically describe a population of aerosol particles and predict their influence on cloud microphysics (see Chapter 3.5); quantifying how uncertainty in aerosol particle formation processes and properties impacts estimates of climate forcing (see Chapter 3.5); and characterizing the pre-industrial baseline atmospheric chemistry of gases and particles relevant for climate forcing.

Approaches and Support Needed

- Continuously measure aerosol particle properties, precursors, and climate parameters (to unravel natural versus anthropogenic influences on climate) over a range of representative environments (e.g., marine, urban, remote) as a function of altitude.
- Measure cloud properties, including precipitation efficiencies, as a function of relevant gas and particle sources, compositions, and concentrations.
- Improve the predictive capability of models across scales through the integration of laboratory measurements, in situ measurements, and satellite observations with detailed process evaluation.
- Develop a measurement-based modeling framework for atmospheric chemistry that can be readily implemented in Earth system models to augment and improve forecasting of weather and regional climate.
- Develop and improve chemical data assimilation techniques with weather and climate forecasts to enable more accurate air pollution forecasting capabilities.

Priority Science Area 4

Understand the sources and atmospheric processes controlling the species most deleterious to human health.

Motivation

As discussed in Chapters 1 and 4, air pollution has documented adverse health outcomes, including chronic and acute effects that can lead to increased mortality and impacts on cardiovascular and pulmonary functioning, and possibly on reproductive and neurological systems as well. It is estimated that air pollution is responsible for 1 out of 8 premature deaths (more than 7 million annually) worldwide.

While particulate matter has been identified as a major contributor to human health risks, the specific chemical species in both the gas and particle phases that cause these various effects and potential synergisms among them are not well understood. The fate and transport of persistent organic pollutants, toxic metals such as mercury, allergens, and pathogens in the atmosphere are also central to understanding atmospheric impacts on human health (e.g., Kellogg and Griffin, 2006). Atmospheric chemistry is a vital component that connects emissions to atmospheric composition and ultimately to human health. Advances in atmospheric chemistry provide the data at the core of understanding the identities, sources, and fates of health-related atmospheric gases and particles on individual, local, regional, and/or global scales.

Actions to Address Key Scientific Gaps

A. Develop mechanistic understanding to predict the composition and transformations of atmospheric trace species that contribute to impacts on human health.

   A few examples to improve understanding of how air pollutants impact human health include: identifying the source of toxic components in primary and secondary particles and how they depend on precursors and reactions in the atmosphere (see Chapters 3.1, 3.2, and 4.2); identifying and quantifying the important individual oxidants in the condensed phase that contribute to toxicity (see Chapters 3.3 and 4.2); and understanding how ozone chemistry and the associated health risks will evolve with a changed climate (Chapter 3.2, 3.3, 4.2).

B. Quantify the distribution of atmospheric constituents that impact human health.

   Examples include determining which atmospheric constituents need to be measured and on what scale to quantify human exposure (see Chapter 4.2); improving the characterization of both individual and population exposure to air pollutants (see Chapter 4.2); and characterizing urban-scale air quality where a growing fraction of global populations live and breathe (see Chapters 1.1 and 4.2).

C. Determine what unique sources and chemical reactions occur in indoor environments that have implications for atmospheric chemistry and human health.

   The indoor environment represents a key scientific gap in atmospheric chemistry as discussed in Chapter 4.2. Examples of unresolved problems include: better understanding of the interplay between indoor and outdoor air quality; characterizing how human emissions and human activities influence indoor atmospheric chemistry; and determining the dominant fate of chemical species indoors, including gas-phase reactions, surface reactions, surface partitioning, and ventilation.

Approaches and Support Needed

- Develop tools to characterize particle composition over a broad size range from nanometer to micrometer scales, with the ability to differentiate bulk from the surface composition, which may determine bioavailability.
- Develop measurement techniques that facilitate high temporal and spatial resolution measurements of a wide variety of gases and particles that may either have direct health impacts or be precursors to those that do, including highly oxidized, multifunctional, and toxic species. This should also include the development of reliable, inexpensive sensors that can be widely deployed for air pollution exposure assessment.
- Integrate observational and modeling outputs to provide accurate assessment of pollutant concentrations for exposure estimates on the appropriate spatial and temporal scales. These should include sources, transformations, and fates of key precursors.

- Develop methods for effective communication of real time air quality data to non-practitioners.
- Coordinate research with toxicologists and epidemiologists.

Priority Science Area 5

Understand the feedbacks between atmospheric chemistry and the biogeochemistry of natural and managed ecosystems.

Motivation

Biogeochemical cycles control the elements that are necessary for life and couple chemistry in the atmosphere with oceans, the solid earth and the terrestrial and marine biospheres. This exchange of compounds supports resource production and maintenance associated with global food security (e.g., agriculture, fisheries) and certain energy sources (e.g., biofuels, wood).

There is a need to understand the spatial and temporal fluxes of elements between the atmosphere and terrestrial and marine ecosystems, as well as how atmospheric chemistry modulates the composition and bioavailability of these elements. These exchange processes are influenced by human activity and global climate, and are directly tied to the societal issue of natural and managed ecosystem health. In addition, biogeochemical cycles and ecosystem health play a central role in climate by regulating carbon uptake by the biosphere and the exchange of greenhouse gases and aerosol particle precursors. A better understanding of how atmospheric chemistry impacts the land and marine carbon sink is key to understand the ability of the Earth to temper the impacts of current and future emissions of CO_2 and other greenhouse gases to the atmosphere. Furthermore, the biogeochemical cycling of toxic constituents (e.g., mercury) directly affects ecosystems, as well as human health. Finally, given the growing pressure on agricultural systems in an era of rising demand for food, it is vitally important to evaluate the impact of atmospheric composition and deposition on crop growth, livestock and seafood stocks, and aquaculture. In addition, these managed ecosystems are also a significant source for a number of important atmospheric species, including methane, nitrous oxide and ammonia.

Understanding biogeochemical cycles is tightly coupled to fundamentally important atmospheric chemistry questions, including the sources, sinks, and oxidation processes associated with gas and condensed phase chemistry. However, addressing these inherently interdisciplinary scientific challenges will require the collaboration of atmospheric chemists with scientists from other relevant disciplines. Major scientific goals include understanding the cycling of elements through the various components of the Earth system, the impacts of deposition of atmospheric nutrients and contaminants to natural and managed ecosystems, and the feedbacks of ecosystems onto the atmosphere. New laboratory and field studies are needed to characterize these atmospheric chemistry processes, which are fundamentally important to the atmospheric chemistry community and can be incorporated into predictive models.

Actions to Address Key Scientific Gaps

A. Quantify the full suite of trace gases and particles deposited from the atmosphere and connect these to ecosystem responses.

> As discussed in Chapter 3.6, much work is needed to address this key scientific gap (which is connected with PSA2). Examples of such research include determining how the deposition of nitrogen, phosphorus, iron, and other nutrients impact terrestrial and marine carbon uptake; and determining how crop productivity is impacted by ozone and other chemical species.

B. Identify and quantify the chemical composition, transformations, bioavailability, and transport of nutrients and contaminants in the global atmosphere and their interactions with the biosphere.

> As discussed in Chapter 3.6, atmospheric constituents and their transformations and bioavailability can significantly impact ecosystem health. Examples in this area include understanding how atmospheric oxidation processes alter the distribution and form of nitrogen deposited to the biosphere; determining what processes control the bioavailability of iron in the atmosphere and its assimilation into the marine carbon cycle; and quantifying the exchange of organic species between ecosystems and the atmosphere.

C. Identify important feedbacks between atmospheric chemistry and the biosphere under global change.

> As discussed in Chapter 3.6, biosphere-atmosphere interactions respond to global change. Examples of open research questions in this area include: understanding how land use change and urbanization will impact biosphere-atmosphere fluxes; identifying the impact of changing biological processes and atmospheric and oceanic acidity on the air/sea exchange of nutrients and radiatively active species; and understanding how temperature and other environmental changes affect biogenic emissions and the resulting formation of atmospheric pollutants.

Approaches and Support Needed

- Develop a more comprehensive treatment of coupled atmospheric chemistry and natural and managed ecosystems in Earth System Models.
- Develop complementary atmospheric chemistry measurements at existing long-term terrestrial (e.g., NSF Long-Term Ecological Research [LTER], Ameriflux, NEON in the U.S. and international sites) and marine (e.g., Bermuda Institute of Ocean Sciences; Mace Head, Ireland; American Samoa) locations.
- Develop instruments to quantify total reactive carbon and nitrogen.
- Develop measurement techniques that facilitate high time and spatial resolution measurements of a wide variety of gases and particles that impact ecosystems, including highly oxidized, multifunctional, and toxic species.
- Initiate research coordination with the ecology, plant physiology, agriculture, and marine biogeochemistry communities.

Chapter 6—Recommended Supporting Programmatic and Infrastructure Priorities for Advancing Atmospheric Chemistry Research

Thus far, this report has discussed the importance of the field of atmospheric chemistry (Chapters 1–4) and recommended Priority Science Areas and more focused key gaps that the Committee believes will drive the field forward (see Chapter 5). In this chapter, the Committee discusses important practical recommendations over the next decade for enabling the research priorities described in Chapter 5. The focus here is on how to support the research identified above. We take as a given that the funding of high-risk/high-reward projects by NSF is necessary to advance the field of atmospheric chemistry. As per the statement of task, these are directed primarily to areas of interest for the atmospheric chemistry program at the National Science Foundation (NSF).

As with the scientific areas, the Committee gathered ideas from the atmospheric community for possible logistical and programmatic recommendations. In particular, in order to understand the trends that are currently occurring in the field of atmospheric chemistry and the research emphases of different federal groups supporting work in this area, the Committee requested information from five government agencies. The full results are presented in Chapter 6.1 and Appendix C.

During the Committee's interactions with members of the atmospheric chemistry research community (see Appendix B), the great majority of participants indicated their appreciation for the NSF staff who have managed this area. There is a sense that proposals have generally been dealt with efficiently and fairly. Appreciation was expressed for the system of using a combination of independent peer review and Program Officers' judgment, rather than relying heavily on panel reviews as is the case increasingly in other parts of NSF. Having a base of permanent staff, with some rotating visitors, has provided continuity of interaction with the community.

The Committee recognizes that an important fraction of the NSF-funded research involving atmospheric chemistry research occurs through support to the National Center for Atmospheric Research (NCAR) from the Facilities Section of NSF. While the Statement of Task did not explicitly include an assessment of NCAR's role, NCAR is part of the infrastructure of atmospheric chemistry in the United States and the peer-reviewed Atmospheric Chemistry program at NSF relies on partnering with NCAR. We thus also comment on NCAR's role in the field.

To enable the recommended science priorities in Chapter 5, support of various activities that include the development of "tools;" the collection, analysis, and archiving of data; and collaborations within atmospheric chemistry and with other communities are all important. These are shown schematically in Figure 6.1 and discussed in more detail below in Sections 6.2 through 6.5.

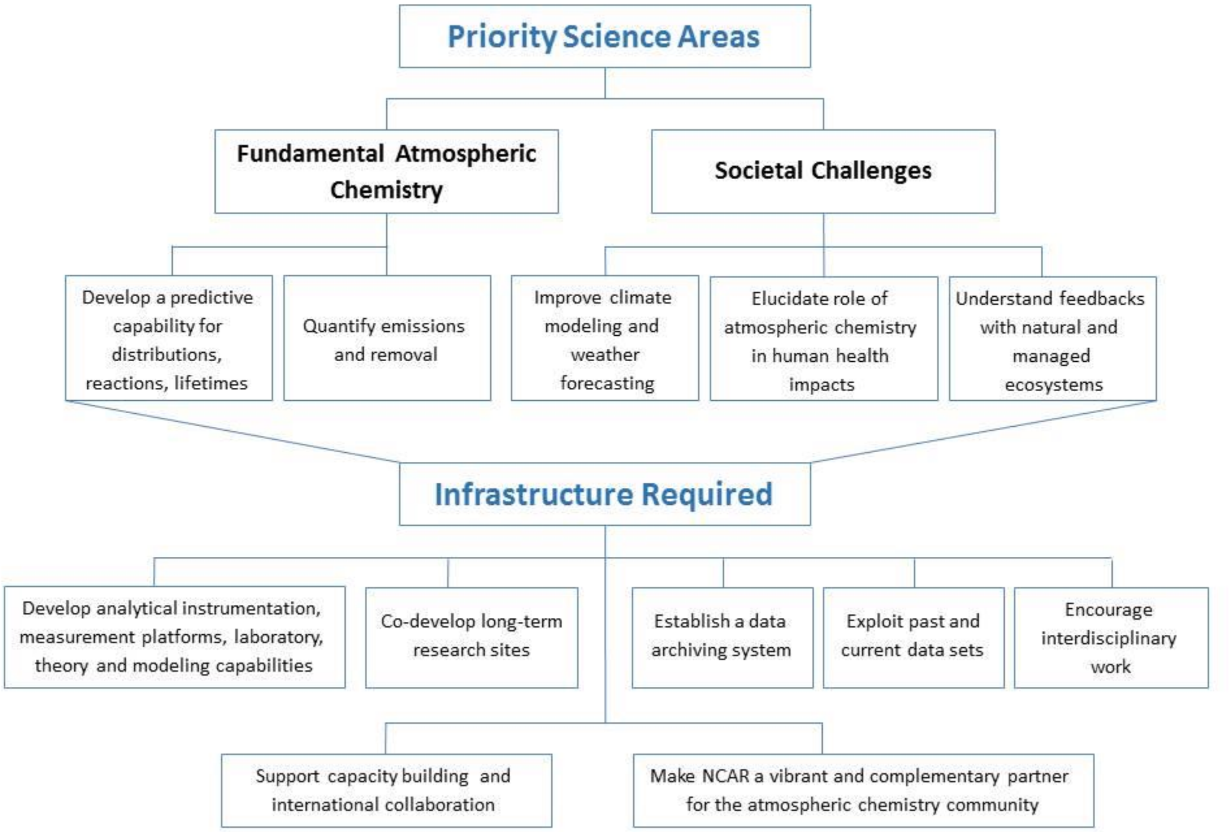

FIGURE 6.1 Summary of the Committee's Priority Science Areas (see Chapter 5) and recommended infrastructure/programmatic actions (Chapter 6). The National Center for Atmospheric Research (NCAR) is part of the infrastructure of atmospheric chemistry in the United States, and the peer-reviewed Atmospheric Chemistry program at NSF relies on partnering with NCAR; thus, the Committee also comments on NCAR's role.

6.1. DISCUSSION OF TRENDS IN ATMOSPHERIC CHEMISTRY

The field of atmospheric sciences as a whole has been growing over the past several decades. One way to illustrate this trend is by tracking the number of graduate students in the field. NSF reports that a range of 140 –170 PhDs in Atmospheric Science and Meteorology are conferred each year in the United States and that this number is slightly increasing through time.[17] Although it was not possible to discern what fraction of the total is atmospheric chemistry due to the interdisciplinary nature of the field, and this number does not capture the atmospheric chemists graduating with PhDs in other fields (e.g., Chemistry, Environmental Engineering), it is the sense of the Committee that atmospheric chemistry is also growing in terms of numbers of

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[17] In the 1990s, the number of PhDs awarded in atmospheric sciences and meteorology was approximately 145 annually, and in 2013 the number was 167 graduates. Information was obtained through NSF's Survey of Doctorate Recipients under the Scientists and Engineers Statistical Data System (NSF SESTAT) Metadata Explorer: http://ncsedata.nsf.gov/metadataexplorer/metadataexplorer.html. Accessed: September 10, 2015.

researchers. While workforce issues were not within the scope of the Statement of Task, the Committee notes that the engagement, recruitment and retention of traditionally underrepresented and underserved groups continues to be an important responsibility of the atmospheric chemistry community. More detailed data gathering on the atmospheric chemistry related PhD degrees awarded and the career paths of atmospheric chemistry PhDs would allow for better characterization of the challenges facing the field as a whole.

A related question is whether the resources for the field of atmospheric chemistry have also been growing. Recent trends in spending by the major atmospheric chemistry research funding agencies, including NSF, National Oceanic and Atmospheric Administration (NOAA), Environmental Protection Agency (EPA), Department of Energy (DOE), and National Aeronautics and Space Administration (NASA) are shown in Figure 6.2. Overall, most of the agency funding shown here has been relatively flat over the past decade (with the possible exception of DOE and NOAA Oceanic and Atmospheric Research, OAR), especially when inflation is taken into account (dashed lines in Figure 6.2 show funds adjusted to 2015 dollars[18]). For NSF, the total number of atmospheric chemistry research projects funded, the inflation-adjusted combined award amount, and the median award amount have not changed substantially over the past two decades (see Appendix C; Figures C.2a and C.2b).

The Committee also examined how support from the various agencies is divided among types of research approaches, including laboratory, theory, field, and modeling studies. Although there is often overlap of types of research within a given study, estimates over the past decade from the NSF Atmospheric Chemistry program show that it has typically spent approximately half of its budget on field projects, 25 percent on lab studies, 15 percent on modeling, 5 percent on instrument development, and 5 percent on “other” projects (see Figure 6.3). The larger proportion used to support field work is likely due to the nature of field work, which generally is personnel intensive and requires resources for instrumentation and personnel deployment. Data from EPA and NOAA also indicate that the largest fractions of their support are spent on field studies, with variable smaller amounts on lab and modeling studies (see Figures C.3a and C.3b). For example, NOAA and EPA external funding appears to spend relatively more on modeling compared to laboratory studies.

In summary, the judgement of the Committee is that the field of atmospheric chemistry has been expanding for the past several decades, but the amount of funding has not increased substantially. This has put pressure on the field as a whole to do substantially more with less, while maintaining a balanced portfolio of research approaches required by the breadth of research described in Chapters 3 and 5. While the Committee does not recommend major changes to this balance now, the appropriate balance in the future will depend on the nature of the issues being addressed, which is likely to change with time. It is therefore important to have flexibility in the distribution of efforts and support, with the recognition that any initiatives to provide more support in one area will have negative effects in another unless additional support is found.

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[18] The funding amounts from each agency for each year available were converted into 2015 dollars using the U.S. Inflation Calculator, based on Consumer Price Index data from the Bureau of Labor Statistics: http://www.usinflationcalculator.com/) (accessed November 12, 2015).

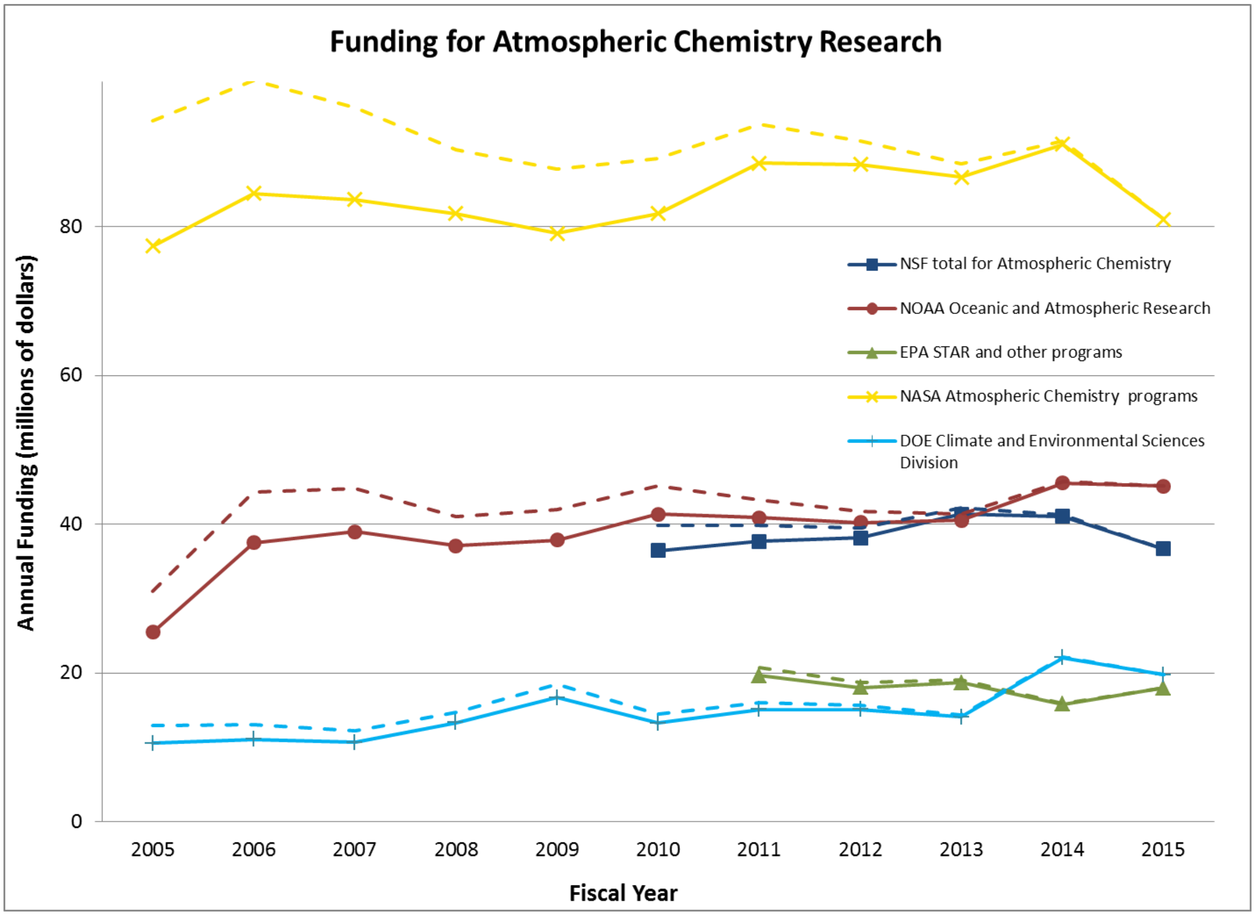

FIGURE 6.2 Millions of U.S. dollars spent on atmospheric chemistry research each year by NSF through the Atmospheric Chemistry program (ATC) and other sources[19] (dark blue squares), NOAA Office of Oceanic and Atmospheric Research (red circles), EPA (a sum of Science to Achieve Results [STAR] and other programs; green triangles), NASA Atmospheric Chemistry programs[20] (yellow x marks), and DOE Climate and Environmental Sciences Division (light blue plusses) between fiscal years 2005 –2015.[21] Corresponding dashed lines indicate funding adjusted for inflation (shown in 2015 dollars). Aggregate budget and funding information were provided through personal communication with representatives from NSF, EPA, NOAA, DOE, and NASA, and data were subject to availability.

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[19] Total NSF funding prior to 2010 was not available in a consistent format that could be displayed here. Other sources include the NSF Deployment Pool (https://www.eol.ucar.edu/facilities-instruments); Atmospheric Chemistry Observations & Modeling (ACOM, part of the National Center for Atmospheric Research [NCAR]); and funding for atmospheric chemistry research from other NSF directorates, including the Directorate for Mathematical & Physical Sciences (Environmental Chemical Sciences program, and Division of Chemistry through the Centers for Chemical Innovation program); the Directorate for Engineering (Environmental Engineering and Environmental Sustainability programs); and the Directorate for Geosciences (Chemical Oceanography, Arctic Natural Sciences, and Antarctic Ocean and Atmospheric Sciences programs).

[20] NASA Earth science combines a number of areas of atmospheric science into atmospheric composition along with atmospheric chemistry; for example research on the physical and optical properties of clouds and aerosol particles is in the same category as global measurements of ozone; often, atmospheric physics is part of projects that are primarily atmospheric chemistry. Funding amounts vary with the life cycle of satellite missions, so care should be taken when interpreting temporal trends. Also, comparisons are complicated by changes in accounting personnel and systems during this period.

[21] Data received from one agency may not include specifications of whether any funds were directed to another agency; therefore the amounts shown may reflect some double counting.

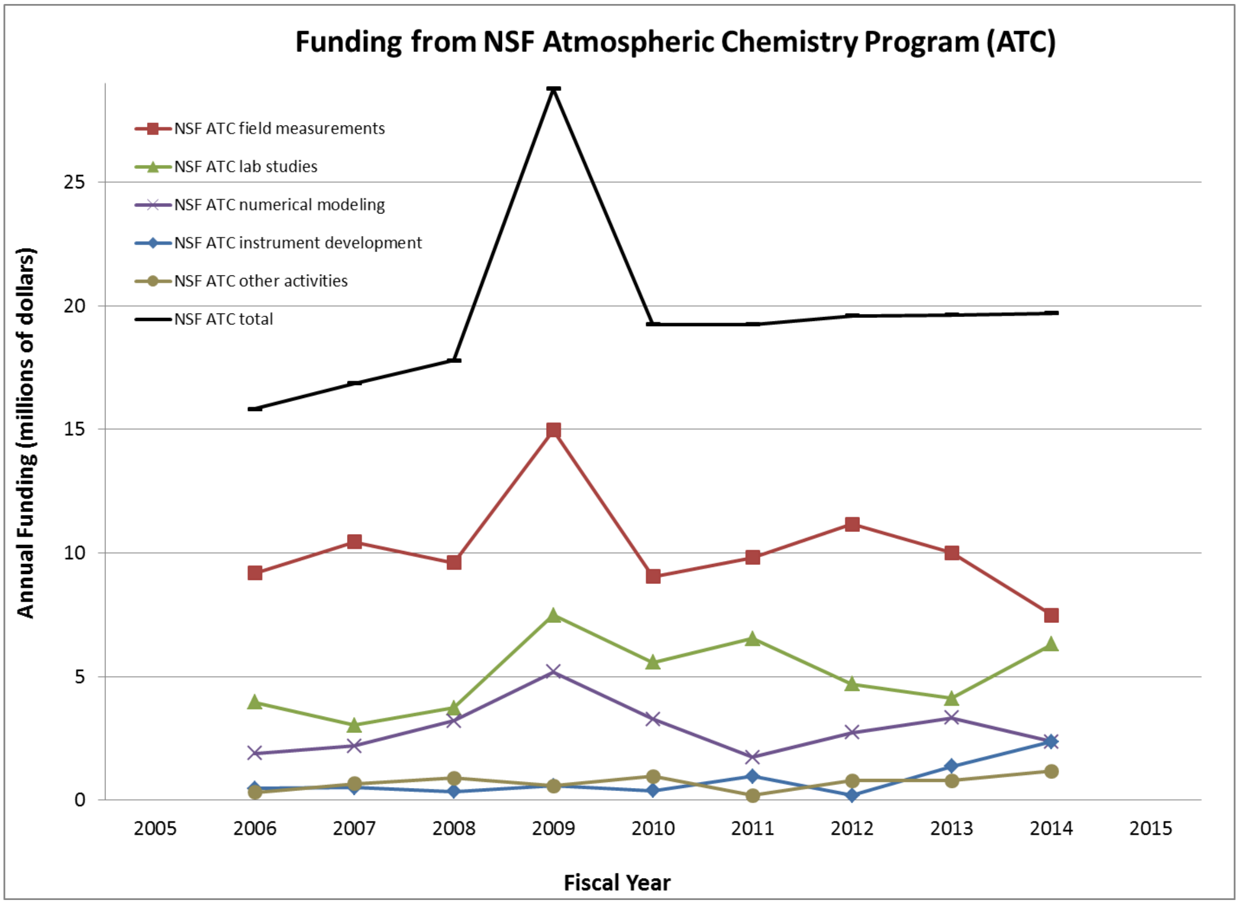

FIGURE 6.3 Annual funding in millions of U.S. dollars for the NSF Atmospheric Chemistry (ATC) program that has been allocated to field measurements (red line), laboratory research (green line), numerical modeling studies (purple line), instrument development or facilities and infrastructure (dark blue line), and "other" projects[22] (brown line) between fiscal years 2005 –2015. Note the peak in NSF ATC total funding in 2009 corresponds to a one-year increase provided by Recovery Act stimulus funding. Aggregate budget and funding information were provided through personal communication with representatives from NSF and data were subject to availability.

6.2 DEVELOPMENT OF TOOLS FOR ATMOSPHERIC CHEMISTRY RESEARCH

As discussed throughout this report, building toward a predictive capability for understanding the chemistry of the atmosphere requires contributions from laboratory studies, theory, field research, satellite measurements, and modeling approaches. And as described in the previous section, the current support from the federal agencies, including NSF, generally supports a balance of the various types of approaches. These are the fundamental tools of the field of atmospheric chemistry, and the Committee believes that NSF plays an essential role in

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[22] The "other" category includes support for workshops, conferences, symposium, summer student programs, the International Global Atmospheric Chemistry (IGAC) core office, plans and costs for proposed field campaigns, University Corporation for Atmospheric Research (UCAR) educational and outreach programs, and Atmospheric Chemistry Center for Observational Research and Data (ACCORD) activities at NCAR.

fostering the development of the next generation of many of these tools. Contributions to this by industry and other government funding agencies could also be important.

Laboratory Experiments and Theoretical Studies

Many of the tools needed to advance laboratory studies overlap with those needed for field studies, such as development of new instrumental approaches discussed in the following section. However, some needed tools are sufficiently complex and hardware intensive that they will be useful, at least initially, primarily in laboratory situations where power, size, and space are not as limited as in field deployments. Ultimately such techniques may be adapted to field projects to provide important insights into unrecognized atmospheric constituents and processes. Laboratory studies could also benefit from implementing new techniques as they arise from other areas such as surface, materials, and pharmaceutical sciences with the long-term goal of interrogating complex systems under atmospherically relevant conditions.

One example of needed techniques is that for the analysis and identification of individual organic species and their location in complex milieus such as secondary organic aerosol particles (SOA) and surface films, in real time without sample collection and workup that can introduce artifacts. Techniques to probe interfaces of soft materials such as SOA and bioparticles as well as species on solid surfaces such as dust particles or the built environment are also needed in order to develop accurate mechanisms and models that faithfully represent the fundamental underlying principles and explain and guide laboratory and field studies.

Computational chemistry has undergone remarkable advances in the last decade and is able to address based on first principles simpler model systems relevant to the atmosphere. There continues to be a need to develop theoretical approaches that can be applied in practical terms to systems that encompass thousands of chemical species and cover a range of particle sizes and complexity from several to hundreds of millions of molecules, and that encompass both physical and chemical processes. Theoretical advances, supported and evaluated by both laboratory and field investigations, are required and essential to improving predictive capability.

Instrument and Instrument Platform Development

Accurate and specific measurements of trace gases and particles are central to atmospheric chemistry understanding and to developing predictive capabilities that inform societal choices (see Chapter 4). Many of the advances in the field of atmospheric chemistry that have been made over the last decades as described in Chapter 3 have arisen largely because of the ability to measure more species in air and at ever decreasing concentrations. Identification of new species and lowering detection limits for known species are key to testing the applicability of results from laboratory and theoretical studies to ambient air as well as providing essential data for testing model predictions.

As discussed in Chapter 5, new analytical techniques, instruments, and instrument platforms are needed to support the Priority Science Areas (PSAs) of the next decade. For example, new analytical techniques are needed to identify sources and sinks of compounds and radical species in both gas and condensed phases that cause detrimental health outcomes, and to quantify their concentrations and distributions (see PSA 4). Further, understanding the role of aerosol particles on climate and weather will require tools to characterize particle composition, morphology and phase over a broad size range from nanometer to micrometer scales, preferably with the ability to differentiate bulk from the surface (see PSA 3). Similarly, understanding feedbacks between the atmosphere and the biogeochemistry of natural and managed ecosystems would benefit from the development of instrumentation and measurement strategies to quantify fluxes of many different gases and particles (see PSA5).

The development of instruments and instrument platforms is a challenging area, since there is a wide range of spatial and temporal scales that need to be covered, with specific requirements depending on the science questions being addressed. Being able to make measurements in three dimensions of many different chemical species over varying temporal and spatial scales is key, including defining the vertical heterogeneity in composition. Concentrations can change significantly with altitude, resulting in non-linearities in the chemistry, for example the rate of the RO_2 radical self-reactions, which varies with the square of the RO_2 concentration, becomes competitive with the first order reactions of RO_2 with NO as the latter concentrations decrease. In addition, sources and concentrations of both gases and particles are often inhomogeneous with altitude. Interpreting surface measurements correctly under conditions of strong vertical mixing also requires understanding these heterogeneities and non-linearities. Similarly, testing model predictions would be greatly aided by the availability of atmospheric chemical observations in three dimensions over as wide an area and over many different times and meteorological conditions as possible. As a third example, the connection of atmospheric chemistry to issues of societal relevance often involves measurements of atmospheric constituents on scales relevant to a specific problem. In many cases, atmospheric composition is not directly measured at or close to the site of interest, but rather interpolated or extrapolated from fixed site monitors that can be located some distance away. The latter often have a limited suite of measurements and fixed sampling times and durations that may not be suited to addressing the specific scientific questions.

The development of new instrument platforms is also important. While the use of aircraft, balloons, and blimps has provided, and will continue to provide, important atmospheric chemistry data in three dimensions, by their nature they are expensive and generally limited to planned field campaigns carried out over limited spatial and temporal regimes. The rise of unmanned aerial vehicles (UAVs or “drones”) provides increased opportunities for atmospheric chemistry measurements (see Figure 6.4; Everts and Davenport, 2016).[23] They have the potential to cover a wider spatial region than fixed sites or towers, and provide a nimble capability that can be relatively easily moved to different locations. This would be particularly useful in the case of unforeseen events such as the Deepwater Horizon explosion and aftermath, or volcanic eruptions that require rapid response with a variety of instruments. Transmission of data to a central facility that provides quality control and standardization of formats, as well as conversion to user-friendly (including the public) formats, would help take full advantage of the data generated.

In this light, a key to taking full advantage of UAVs is the development of robust, light, small, accurate, and specific sensors that would provide data traceable to standardized methods. There are currently a number of excellent instruments that are now flown on aircraft, balloons, and blimps, but the use of UAVs will likely require lighter instruments with low power demands, and hence new approaches to measurement techniques. Furthermore, there is substantial interest in developing distributed sensor networks for estimating air pollution exposure for health studies

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[23] See also https://eos.org/project-updates/drone-squadron-to-take-earth-monitoring-to-new-heights.

Figure permission pending

FIGURE 6.4 Various sizes of unmanned aerial vehicles (UAVs) can be useful in atmospheric chemistry research. Left: Photograph of coordinated measurement campaign using three smaller UAVs to make measurements below, within, and above a cloud layer. Right: The NASA Global Hawk UAV—with a 1,500-pound payload, 8,500-nautical-mile range, and 24-hour endurance—has been used in longer-duration measurement projects. SOURCES: Henson, 2005, and http://www.nasa.gov/centers/armstrong/news/FactSheets/FS-098-DFRC.html.

(PSA 4). Advances in solid-state technology and materials science may be helpful in the development of such methodologies. The next step in the development of miniaturized instruments and instrument platforms is vitally important and NSF could play a central role in fostering that development.

Low-power, lightweight sensors would also be useful in widespread networks, perhaps involving citizens. Although low cost distributed sampling networks provide an exciting opportunity to characterize concentrations of gases and particles with high time resolution and fine spatial granularity, the quality of many existing approaches and data sets have not been thoroughly vetted. Even established techniques for measuring atmospheric trace species can be subject to a variety of uncertainties related to representativeness, precision, bias, detection limits and accuracy. It is thus important that new methodologies are rigorously tested and the uncertainties in the data they generate clearly defined. Poor data quality hinders continued advancement in atmospheric chemistry and can misinform the public. Therefore, robust instrumentation with excellent precision is necessary for this testing and determination of uncertainties.

NSF currently provides instrument development support through several programs outside of its Atmospheric Chemistry program, such as the Major Research Instrumentation (MRI) program, which has an instrument development component, and the NSF Small Business Innovation Research (SBIR) and Small Business Technology Transfer Research (STTR) programs. These programs have produced valuable breakthrough technologies that have enabled improvements in atmospheric chemistry research (NASAEM, 2015; NRC, 2005). However, there are limitations with respect to atmospheric chemistry needs. For example, only two proposals per campus are allowed to be submitted to the MRI program for larger universities, and these proposals cover all areas of science. SBIR topics have been described as being narrow as a means to pare down the number of applications (NASEM, 2015). As a result, there are generally few opportunities for high-risk, high-reward proposals for instrument development in atmospheric chemistry at NSF.

The Committee believes that there will be continuing instrument development needs related to atmospheric chemistry in the future. We encourage the Atmospheric Chemistry program to consider mechanisms for providing more support for instrument development work.

For example, the Atmospheric Chemistry program at NSF could work more closely with other NSF programs, such as the MRI and SBIR programs as well as the Chemistry Division in the Mathematical and Physical Sciences Directorate at NSF and the Chemical, Bioengineering, Environmental, and Transport Systems Division in the Engineering Directorate, to ensure that atmospheric chemistry research needs are fully integrated into the priorities of these other programs. In addition, it is important that viable mechanisms be available within the Atmospheric and Geospace Sciences Division (AGS) to submit proposals for new instruments and techniques that often take substantial amounts of time to adequately develop and test before they are ready for use in the field or lab. Many other federal agencies have historically been reluctant to fund such instrument development outside of their own laboratories unless there is a specific set of “deliverable” measurements by the end of the grant, which is typically three years. Given the challenges for instrument development, it may be that cross-directorate support or advancing a cross-directorate initiative, for example, with the Mathematical and Physical Sciences and/or Engineering Directorates, would be beneficial. Another potential mechanism might be collaboration with scientists at a national center (see Section 6.5).

Model Development

There is a wide variety of available atmospheric chemistry modeling tools that range in spatial scale, temporal scale, and technical approach. The utilization of model type depends on the scientific application, and a diversity of approaches is needed to develop a broad toolbox to understand the complex problems described in Chapters 4 and 5. From the spatial perspective, tools range from point-based approaches such as box models and one-dimensional column models to three-dimensional models that range from very high resolution (on the order of meters; e.g., large eddy simulation models) to regional (e.g., continental scale) and global scales. From the temporal perspective, box models typically run on the scales of hours, whereas coupled chemistry-climate models run on the scale of decades to centuries. Methodological approaches also vary, including using Lagrangian plume models to fixed grid Eulerian models. Within fixed grid models, there are a number of new approaches that can help to answer key scientific questions. For example, data assimilation techniques can help integrate models and observations and improve predictability (e.g., Bocquet et al., 2015), adjoint model approaches can assess model sensitivities (Chai et al., 2006; Vautard et al., 2000), inverse modeling can improve understanding of sources and sinks (Kasibhatla et al., 2013), and reduced-form models can operate with reasonable verisimilitude and efficiency to be incorporated in socio-economic or decision-making tools (Kerl et al., 2015).

Modeling is the key element for developing a predictive capability for atmospheric chemistry and addressing the areas of priority science identified by the Committee in Chapter 5. For this predictive capability to develop, substantial advancements are required, including (1) acquiring more observations of chemical species across temporal and spatial scales for chemical data assimilation and model evaluation, as well as taking advantage of existing datasets (see below), (2) dedicating additional resources to support the development of modeling software, and (3) continuing investment in high performance computing (HPC) resources. New predictive modeling approaches can build on techniques developed by the numerical weather and seasonal to decadal forecasting communities. On shorter timescales (e.g., days), a key element influencing model skill is the organization and assimilation of observations for initial conditions. As time scales lengthen, the uncertainty in the knowledge of emissions becomes dominant. However, atmospheric chemistry models will have additional needs beyond what has been developed for the weather forecasting community, as the range of chemical lifetimes of important species provides a unique data assimilation problem.

Currently, the broad toolbox of modeling software is supported by a wide variety of federal agencies, including NSF. Despite substantial progress over the past two decades in model tools, continued investment in atmospheric chemistry model development is needed to make accurate, predictive atmospheric chemistry modeling a reality in addressing the interdisciplinary issues described in this report. However, similar to instrument development, model development faces substantial challenges in obtaining funding. A major obstacle for the atmospheric modeling community is the disparate spatial and temporal scales of atmospheric chemistry, and the resulting difficulty in building consistent modeling tools and methodological approaches that can work together and integrate across these scales. An additional and perhaps greater challenge is integrating models to develop a predictive capacity for atmospheric chemistry. NSF needs to continue its investments in model development and applications across scales, ranging from developing and incorporating theoretical chemistry to predicting global composition. There is a clear opportunity for NSF to initiate a new focus for modeling across scales to develop a predictive capacity for atmospheric chemistry. Such a research initiative could promote collaboration and coordination across agencies and across different NSF programs (e.g., theoretical chemistry and atmospheric chemistry, environmental engineering, and climate).

Currently, most three-dimensional numerical models used in atmospheric chemistry require some level of HPC, whether at a university center or a national center. Computing resource limitations and complexity of parallel architectures can present barriers for future research in atmospheric chemistry that more often than before requires HPC modeling. NSF provides the atmospheric chemistry community with vital HPC capability at NCAR, directly through HPC computing services at centers like Yellowstone[24] or indirectly through joint projects using the Community Atmosphere Model with Chemistry or the Whole Atmosphere Community Climate Model. Because atmospheric chemistry plays a key role in climate, the urgency of these simulations needs to be understood as part of the Earth system community's research into global change (Garcia et al., 2012; Mahlman et al., 1980; Pinto et al., 1983; Prather et al., 1987; Wild et al., 2003). In this context, NSF should ensure that principal investigators have the availability of resources to advance the science.

Summary of Development of Tools for Atmospheric Chemistry Research

The Atmospheric Chemistry program at NSF is well positioned to lead efforts to improve the tools needed for laboratory and theory studies as well as for instrument and model development in collaboration with other directorates and programs within NSF and at other agencies. Overall, improved support for these basic tools of atmospheric chemistry research is essential for advancing the science over the next decade, and will allow the scientific community to observe, measure, and predict atmospheric chemistry processes and their interactions with other physical, biological, and human systems.

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[24] https://www2.cisl.ucar.edu/resources/computational-systems/yellowstone.

Recommendation 1: NSF should ensure adequate support for the development of the tools necessary to accomplish the scientific goals for the atmospheric chemistry community, including the development of new laboratory and analytical instrumentation, measurement platforms, and modeling capabilities.

6.3 INFORMATION COLLECTION, ANALYSIS, AND ARCHIVING IN THE ERA OF "BIG DATA"

Data are an essential element of research in all fields of science, and the field of atmospheric chemistry is no exception. In particular, there are several issues related to the collection and analysis as well as sharing and communication of data that the Committee chose to focus on in this report. First, the collection of measurement data over long periods of time allows for the discernment of trends that are not possible to see in one-time field projects. Second, the answers to research questions are often apparent only after intensive data analysis or with the analysis of data from multiple projects. Third, the management of large volumes of "big data" is becoming more ubiquitous in atmospheric chemistry research (and scientific research more broadly) and mechanisms for effectively and efficiently archiving, sharing and mining data, including making it available to the broad scientific community and the public, are needed.

Long-Term Research Sites

The Committee recognizes the central importance of long-term research sites for comprehensive atmospheric chemistry research. The need for such sites was also clearly identified by the 2001 NRC report *Global Air Quality: An Imperative for Long-Term Observational Strategies* (NRC, 2001). A classic example is the Mauna Loa data set that clearly revealed long-term trends in CO_2 many decades ago. Such sites are beneficial for addressing important scientific research questions, in particular those related to the spatial and temporal fluxes of elements between the atmosphere and terrestrial and marine ecosystems (see PSA 2) and understanding feedbacks between the atmosphere and natural and managed ecosystems (see PSA 5). For example, how does atmospheric chemistry modulate trends in the composition and bioavailability of nutrients and contaminants? What are the emissions from natural and anthropogenic sources across sectors and environments, and how do those emissions change with climate change, land use change, and other global changes? How efficiently do wet and dry deposition remove particles and trace gases from the atmosphere, and how are these changing?

The vision for these long-term sites is not simply to develop a network of monitoring stations, but rather to exploit research sites with core measurement capabilities and long-term knowledge about regional photochemistry, meteorology, ecosystem properties, and biosphere-atmosphere exchange processes that the atmospheric chemistry community can also use as a resource for making and interpreting new measurements. Long-term sites gain scientific value with time and provide a rich environment in which to understand changes in atmospheric chemistry driven by changing emissions, land use, climate, or other factors of societal relevance. Because the atmosphere is a global commons, it is important that sites be spatially distributed in a representative way, and that necessarily involves international cooperation and collaboration. For many scientific questions in atmospheric chemistry, research sites do not necessarily need to be created in new locations, in fact there would be great value in leveraging from existing monitoring locations. Indeed, there are several sites where such long-term research has enabled progress on important scientific questions and involve international cooperation by scientists as well as funding by governmental organizations from different countries and the UN World Meteorological Organization's Global Atmospheric Watch.[25] For example, NOAA GMD's background monitoring sites (Mauna Loa, HI; American Samoa; Barrow, AK; the South Pole, etc.) were set up primarily for monitoring greenhouse gases, but they have been expanded to make many additional measurements and have been used extensively by non-NOAA scientists to conduct process-oriented research.[26] Another example is the AGAGE-NASA[27] network which includes 11 sites measuring over 50 gases with in situ instrumentation over the past 38 years. Beyond the existing sites of opportunity managed by international, federal, and state partners, scientific research using existing data and atmospheric modeling tools should be used to optimally site any additional measurement locations as defined by the needs of the atmospheric chemistry community.

Long-term sites have some distinct advantages over individual, one-time field projects. Developing field sites for specific campaigns is often necessary to address critical regional and/or unique problems. However, setting up new field sites for specific campaigns is typically a substantial fraction of the cost and effort associated with the experiment, and it requires a large scale coordinated effort and commitment by the research community and funding agencies. Large scale individual field campaigns are not always the most cost-effective approach for making scientific progress. Furthermore, it is often difficult to place the results from a single field project in a larger context unless it is built around longer term observations and understanding. Supporting long-term sites and encouraging proposals for research at those sites enables regular introduction of new observational approaches, testing and inter-comparison of new measurement capabilities, and evaluation of new scientific understanding of emissions, transformation mechanisms, and deposition, without the need to develop novel infrastructure for costly field campaigns as frequently as is often done in the atmospheric chemistry community.

Specifically for issues of understanding biosphere-atmosphere exchange and its evolution, the atmospheric chemistry community generally lacks long term infrastructure in an appropriate array of representative environments in the United States. NSF and other U.S. scientific agencies have developed long-term sites to understand ecological processes and the carbon cycle (e.g., NSF NEON, DOE AmeriFlux, NSF Long-Term Ecological Research [LTER]). There is potential synergy to be gained by developing infrastructure in collaboration with one or more of these existing networks, or specific long-term sites with a history of atmospheric chemistry research on which additional research facilities could be built. If sites were needed in additional locations, they may provide an appropriate focus for a recently announced initiative at NSF for mid-scale infrastructure.[28] Natural emissions and deposition, and therefore atmospheric chemistry and composition, are highly dependent on phenology, meteorology, season, climate, and anthropogenic air pollution, and are extremely variable across different ecosystems. Thus a distributed set of research sites within the context of existing

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[25] http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html.
[26] http://www.esrl.noaa.gov/gmd/.
[27] http://agage.mit.edu.

[28] http://www.sciencemag.org/news/2016/05/nsf-director-unveils-big-ideas-eye-next-president-and-congress.

knowledge and infrastructure that relates to current and future priorities in atmospheric chemistry research would be most cost-effective.

With several different agencies currently supporting various kinds of long-term sites, it would be most useful if representatives from the various agencies worked in collaboration with academic researchers to determine how existing sites could be more useful to the atmospheric chemistry research community and to decide if/where and how many new sites might be needed. An interagency panel could help prioritize the long-term sites and determine the required infrastructure, whether the sites would be centrally managed (e.g., using resources associated with NCAR) or managed by individual principal investigators (PIs) (like AmeriFlux or LTER sites), core measurements to be included with each site, procedures for the archiving of the samples collected at these sites, and criteria and a review process to support funding decisions. Further, this panel can investigate the potential to contribute to international programs, such as the Global Atmospheric Watch program of the United Nations World Meteorological Organization that focus on long term observations of atmospheric chemistry around the globe.

If existing infrastructure can be used to form the backbone of a network of long-term research sites, the cost to NSF could be reasonable. In addition, NSF would be able to individually support research at these long-term sites in response to proposals from single or small groups of PIs. It could be beneficial to fund such proposals for longer than the standard three-year period that most awards are given.

Recommendation 2: NSF should take the lead in coordinating with other agencies to identify the scientific need for long-term measurements and to establish synergies with existing sites that could provide core support for long-term atmospheric chemistry measurements, including biosphere-atmosphere exchange of trace gases and aerosol particles.

Resources for Data Analysis

Many observations of atmospheric chemical composition and processes, especially those from measurement networks and field campaigns, are not used to their full potential. Scientists historically have obtained three years of funding at a time, which often allows for collection of the data and publication of the most obvious trends and findings. Often the funding is insufficient to mine the data deeply for thorough analysis. In addition, questions raised by future discoveries may be answered with reanalysis of existing data sets.

Although funded research endeavors have produced important results, data streams created from these efforts present a rich resource that could be used for additional analysis to help answer evolving atmospheric chemistry and composition questions and to provide guidance for future field studies. There is considerable opportunity to exploit the treasure trove of existing measurements that represent the collective atmospheric chemistry knowledge from decades of investment in atmospheric chemistry research. For example, existing high-quality atmospheric chemistry field and environmental chamber data sets could be mined to examine chemical mechanisms, to look for trends in time, and to compare the chemistry across different chemical and dynamical regimes (see PSA 1). In addition, data mining of measurements (e.g., concentration ratios) can help constrain emissions and trends (see PSAs 2 and 5). The synthesis and analysis of existing datasets can be applied to guide future research directions and help to test models across various regimes. All previous data sets are not equally valuable, and new field campaigns that take advantage of the rapid advances in measurement technology can provide scientific information that supersedes anything already available. Nor is the analysis of previous observations a replacement for new field campaigns to address contemporary research questions. However, in the face of tighter budgets, taking advantage of existing datasets for new analysis may be a cost-effective way to continue to advance the atmospheric chemistry research agenda.

Funding is required for researchers to perform analysis on collected datasets and for collaborations that take knowledge from the lab or field into model applications. Longer grant periods or perhaps supplemental installments to afford PIs the time and effort to continue analyses may be needed to accomplish these efforts. NSF should also encourage and support new projects that use data mining to advance the science. For a fraction of the cost of another field study, NSF could dedicate some amount of funds to encourage atmospheric chemists, possibly in collaboration with computer scientists, to mine data from previous studies, performing detailed intercomparisons with satellites, measurements, and models.

Recommendation 3: NSF should encourage mining and integration of measurements and model results that can merge and exploit past datasets to provide insight into atmospheric processes, as well as guide planning for future studies.

Data Management and "Big Data"

Establishing a predictive understanding of chemical processes in the atmosphere and their impacts as described in this report has been possible to date only through large investments in laboratory and field observations, theory, and modeling. During this process, vast and multidimensional datasets are continuously generated, particularly in field studies that require increasingly larger resources to manage. Data need to be archived in suitable formats to be useable. In particular, data archives from field measurements in atmospheric chemistry are a fundamentally important resource for the atmospheric chemistry and broader scientific and regulatory communities, and ultimately for the public. They serve as benchmarks for models, provide a rich set of observational constraints to understand atmospheric chemical processes, document changes in composition due to both natural and anthropogenic activities, and assess societally relevant impacts such as human and agricultural exposure.

Currently, the availability of these datasets varies substantially. Some are archived at data centers (with online or offline access) while others are available only upon request from individual scientists. A crucial part of facilitating the comparison and synthesis of data sets is to ensure that there is a common data formatting that allows integration between models and measurements, especially by scientists who were involved in the studies, but also by those who were not. However, the format of much of the currently archived data and the information content associated with them can vary considerably. Critical information, including expert interpretation and data quality aspects, are often not documented sufficiently. Model simulation outputs of importance for atmospheric chemistry research are often not archived or available to the general community. Other datasets generated from models or post-processing of raw data may involve codes that are insufficiently archived or documented. This diversity, together with the increasing volume and complexity of data poses great challenges for research in atmospheric chemistry.

Part of this variability in the availability of datasets is due to a lack of coordination among the federal agencies. Archives of atmospheric chemistry field campaigns and long-term measurements from research and regulatory efforts are currently maintained separately by multiple federal agencies including DOE, NOAA, EPA, NASA, NCAR, and statewide agencies (e.g., California Air Resources Board, among others), as well as private, non profit, and citizen scientist organizations. The atmospheric chemistry community could leverage some of these sources of data, depending on data quality. Atmospheric chemistry-related data archives are not currently coordinated at a national level between different agencies that support or conduct research and measurements in this area.

Syntheses of these large and diverse datasets may provide opportunities for breakthroughs and transformative science over the next decade. It is therefore vitally important that an effective and visionary management approach is established to facilitate the maximum use and impact of datasets, in particular those generated under NSF support. The scientific community and NSF at large have long been aware of the need for effective data management and have established guidance principles and requirements for each research PI to follow towards that goal. However, much of the support and management of datasets still rely on individual research teams, even long after funding has expired.

NSF already engages in data management efforts. Federally funded research through NSF does require data management plans, but no central coordinated data archive and sharing system exists to serve as a repository and resource for the atmospheric chemistry community and other users who need data for related societally relevant work. The use of common cyber-based infrastructures for handling and managing diverse data and facilitating information extraction and knowledge discovery could provide part of the solution to the data management problem. One such initiative established within NSF, joint between the Directorate for Geosciences (GEO) and the Division of Advanced Cyberinfrastructure (ACI), is EarthCube. EarthCube was initiated in 2011 with anticipated support until at least 2022; it is currently a community-based initiative where members can “influence how data will be collected, accessed, analyzed, visualized, shared, and archived; facilitate and participate in interdisciplinary research; and help educate scientists in the emerging practices of digital scholarship, data and software stewardship and open science” (Gil et al., 2014).[29]

NSF should require that future NSF-funded datasets be handled in a manner that allows ready access and comparison with previous datasets. The Committee envisions a centralized system for providing and supporting data management for atmospheric chemistry. Apart from providing facilities for data archiving, accessibility and transparency, a centralized responsibility could assure that datasets are managed with expert preparation and fostering, accompanied with sufficient and standard documentation and metadata (such as codes for generating derivative products and links to source data). It would also ensure longevity in the storage and accessibility of data sets. A centralized system can ensure that its data resources are coordinated with other data management initiatives, like EarthCube, and also ensure that atmospheric chemistry is an integrated discipline within these efforts. Management and financial support for such a system would have to be integrated into long-term planning and resource allocation.

Another possible end goal of these efforts would be an archiving tool that would provide a user-friendly interface for accessing and manipulating (e.g., plotting) global data. This could enable the design of future field campaigns to fill the gaps identified in the more detailed

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[29] EarthCube: http://www.nsf.gov/geo/earthcube/; http://earthcube.org.

analysis and comparison efforts, and also provide a highly visible outreach activity directed to the public at all levels from K –12 to interested citizens.

Recommendation 4: NSF should establish a data archiving system for NSF-supported atmospheric chemistry research and take the lead in coordinating with other federal and possibly state agencies to create a comprehensive, compatible, and accessible data archive system.

6.4 IMPERATIVE FOR COLLABORATIONS

The scope of problems in atmospheric chemistry that need to be addressed is broad, from those that are suitable for study by a single PI to those that require a much larger breadth and depth of expertise than a single PI or single community has. Addressing more complex problems may optimally be undertaken by collaborations that are of different sizes and include scientists from a mixture of academia, government, and the private sector. This will clearly require working across disciplines and thus across directorates at NSF, across government agencies, and across international boundaries.

Interdisciplinary Work

Developing the comprehensive understanding of the Earth system that will allow enhanced predictive capability relies on close integration of knowledge from multiple disciplines or approaches across the Earth sciences. In addition, many of the important problems in atmospheric chemistry require interdisciplinary solutions. Atmospheric chemists need to engage with scientists from other disciplines to solve problems at these disciplinary interfaces, such as collaborations between chemists and cloud physicists to determine aerosol particle effects on clouds and radiation or chemists and toxicologists to better understand the health impacts of particulate exposure. Interdisciplinary work will be integral in addressing many of the scientific questions related to atmospheric chemistry and climate/weather (see PSA 3), human health (see PSA 4), and exchange between the atmosphere and natural and managed ecosystems (see PSA 5). For example, cooperative efforts will be required between atmospheric chemists and physical and chemical oceanographers as well as ecologists to understand the fluxes and impacts of the air/sea exchange of chemicals.

Atmospheric chemistry should also play important collaborative roles where other disciplines lead. For example, joint studies will be necessary between atmospheric chemists and the health science community to understand the key species that need to be measured and on what time and spatial scales to address the needs of the epidemiology and toxicology communities. Collaborations with specific industries or regional air-quality managers can provide better characterizations of emission fluxes to the atmosphere. To contribute to the solution of problems that span multiple disciplines, atmospheric chemists need to (1) identify the science within atmospheric chemistry required to address a particular problem; and (2) build collaborations with the other disciplines whose knowledge is required. Each scientific component required to address the problem need to have either a disciplinary or cross-disciplinary “home.” A collective assumption that a particular component is handled elsewhere may lead to persistent gaps at critical interfaces. The Committee identifies here the scientific priorities within the field of atmospheric chemistry as well as those related to disciplines with which collaboration is required.

Ultimately, the predictive capabilities developed by the atmospheric chemistry community will be used by others such as economists and social scientists in order to translate the science into policy assessments. Included in this discussion of interdisciplinary work is the need to acknowledge and reward integration within a discipline in addition to obvious cross-disciplinary work. Examples are scales of modeling from process-level to global-level, or connection of laboratory, theory, field, and model studies.

NSF sponsors a broad range of research, including social and behavioral studies that provide potential partnerships with the atmospheric chemistry research community to address environmental degradation and human impacts on community scales. The Committee commends NSF for initiatives that motivate collaboration among disciplines and NSF directorates, such as Science and Technology Centers, and programs such as the Dynamics of Coupled Natural and Human Systems. However, the Committee is concerned that mechanisms to support interdisciplinary work submitted outside such directed and relatively short-term vehicles, which have specific goals and structures, may encounter barriers due to NSF institutional and review structures. There are emerging examples of cross-directorate and interagency investment areas such as the SEES (Science, Engineering and Education for Sustainability) and INFEWS (Innovations at the Nexus of Food, Energy and Water Systems) initiatives. It is noteworthy that the INFEWS initiative "enables interagency cooperation ... and allows the partner agencies—National Science Foundation (NSF) and the United States Department of Agriculture National Institute of Food and Agriculture (USDA/NIFA) and others—to combine resources to identify and fund the most meritorious and highest-impact projects that support their respective missions, while eliminating duplication of effort and fostering collaboration between agencies and the investigators they support."[30] The Committee encourages exploration of the possibility of such interagency collaborations.

Funding larger interdisciplinary collaborations is often too expensive for individual programs, such as atmospheric chemistry, so program managers need to leverage their funds by partnering with other directorates in NSF or at other funding agencies. Another challenge is that interdisciplinary research often requires sustained long-term funding, which can be difficult to achieve using a series of the typical three-year NSF grants. In some NSF directorates, an effective approach has been to fund, in five-year increments, centers that draw together scientists who have different expertise and are often geographically dispersed. Such centers could be real, on-the-ground centers, or they could be virtual centers that connect multiple groups to work on a common problem.

Other options for NSF include developing funding mechanisms whereby small, focused teams can integrate expertise defined by the team across multiple disciplines, which would allow an investigator-driven, creative and competitive approach to integration. In addition, NSF could provide funding that supports short-term training of excellent young researchers in another research group with a very different focus. NSF could also identify and alter incentive structures that discourage integration across disciplines. These factors may include the lack of rewards for individual programs to fund work that crosses boundaries, with the appearance of a "zero-sum game" in which resources devoted to interdisciplinary work are perceived as flowing outside the

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[30] Innovations at the Nexus of Food, Energy and Water Systems: http://www.nsf.gov/pubs/2016/nsf16524/nsf16524.htm.

sphere of influence of a particular program. Fixed grant sizes and reviewer pools that evaluate an entire proposal from the perspective of a single discipline can also discourage inclusion of multiple disciplines. Finally, NSF could encourage a "Cross-Disciplinary Integration" component of proposal evaluation, thereby acknowledging the potential and substantial contributions that lie between the traditional ranking categories of "Intellectual Merit" and "Broader Impacts."

Overall, the Committee believes that NSF should explore these various options to develop mechanisms that facilitate integration of expertise across disciplines and across academia, institutes, government, and industry. These mechanisms need to allow an investigator-driven, creative, and competitive approach that can complement the funding of individual PI research. The Committee further encourages NSF's Atmospheric Chemistry program to help lead these interdisciplinary efforts to address problems that better integrate research within the atmospheric chemistry community and help connect atmospheric chemistry research with other disciplines.

In addition to working across a single agency, cross agency efforts are particularly important, and NSF assistance in obtaining support for partners may be needed when funding from a single program or even research agency is not possible. There are existing mechanisms for coordination and collaboration among the various federal agencies, including the Air Quality Research Subcommittee, which is an interagency subcommittee of the National Science and Technology Council in the White House. Interagency planning needs to first identify the topical components required for progress, and then agree upon agency homes for each component, including knowledge gaps that have persistently hindered advances. The overall goal should be to incentivize close coupling of knowledge from multiple disciplines and approaches across government agencies. Cross-agency integration should focus on identifying and removing barriers in targeted subjects such as planning for long-term research sites (Recommendation 2), effective management of data archives (Recommendation 4), or broad research themes as discussed below. In contrast, persistent meetings that serve primarily administrative purposes are not encouraged.

As examples of interagency evaluation, NSF could more proactively partner with agencies like the National Institutes of Health (NIH; including the National Institute of Environmental Health Sciences [NIEHS]) and the Environmental Protection Agency (EPA) on cross-agency initiatives, such as centers focusing on the connection between air quality and health. As discussed in PSA 4, the atmospheric chemistry community can bring important research to this inherently interdisciplinary topic including the chemical characterization of air pollution that leads to adverse health effects, improved quantification of exposures to pollutants, and the characterization of indoor environments. Closer connections of the atmospheric chemistry community with the epidemiology and toxicology communities could allow for enhanced progress. Examples of previous interdisciplinary work in this area show the enormous potential for progress. For example, meningitis outbreaks in sub-Saharan Africa have been tied to local weather, air quality and pollution sources, and climatic conditions, suggesting complex interactions between the atmosphere and human impacts (e.g., Deroubaix et al., 2013; Dukic et al., 2012; Hodgson et al., 2001).

Support of cross-cutting scientific research will require participation of multiple groups, such as from different directorates within NSF and/or a variety of government agencies (e.g., NASA, NOAA, EPA, DOE, NIH, Centers for Disease Control and Prevention [CDC]), as well as cooperation with international entities. Additionally, given the societal relevance of many areas of atmospheric chemistry, the Committee believes that it is important to consider public engagement with atmospheric chemistry research. Examples of public engagement include participation in atmospheric chemistry measurements, use of air quality indicators, and making air quality data readily accessible in a user-friendly format.

Recommendation 5: NSF should improve opportunities that encourage interdisciplinary work in atmospheric chemistry and facilitate integration of expertise across disciplines and across academia, institutes, government, and industry. This improvement may include support of focused teams and virtual or physical centers of sizes appropriate to the problem at hand.

Capacity Building and International Collaboration

Many of the scientific challenges discussed in Chapters 3 and 5 are global in nature and affect individuals from a wide array of backgrounds. Data from a diversity of regimes or representative environments within the United States and around the globe are critical to addressing these challenges, from quantifying the flow of atmospheric carbon and nitrogen between ecosystems and the atmosphere to understanding aerosol particle properties and precursors to unravel anthropogenic influences on climate.

The international atmospheric chemistry community is strong and committed to developing a global understanding of atmospheric chemistry and its impacts on human activities. This has become increasingly essential as the importance of long-range transport is becoming evident. International cooperative efforts are necessary to make major progress in attaining the objectives of all the Priority Science Areas described previously. For example, working with the international community will help to identify locations and develop the sites for long-term measurement programs for many global scale studies (Recommendation 2). These range from individual PI-PI interactions to large-scale international research programs (see Box 6.1).

The success in understanding and applying atmospheric chemistry to improve air quality in the United States has effectively translated into success in improving air quality in some other regions, such as Mexico City. However, this success does not extend to many other areas such as parts of Africa, where indoor and urban air quality remain poor and the depth of air quality problems is essentially unknown. Even some underserved areas within the United States may be poorly characterized. While the basic principles of atmospheric chemistry and its implications for air quality in well-studied regions in the United States and other developed countries also apply in principle to other situations, each urban area has unique environmental conditions, meteorology, and emissions. As a result, cost-effective solutions to poor air quality vary among populated regions around the globe. Scientists from developed countries have studied the atmospheric chemistry of some regions, but for many regions the understanding of the regional atmospheric chemistry is too poor to devise cost-effective solutions. A more sustainable approach is to build the expert human capacity and observational and modeling capability at the regional level through collaborations between U.S. atmospheric chemists and scientists who focus on these regions. This approach also has the greatest chance of improving the air quality in these regions because regional governments are more likely to listen to their own atmospheric chemists than to U.S. scientists, and communities are more likely to trust and communicate with individuals who are familiar with their situations.

BOX 6.1
International Atmospheric Chemistry

Global programs that engage scientists from around the world to focus on atmospheric chemistry are vital for cooperation and capacity building to address societal needs. The **International Global Atmospheric Chemistry (IGAC)** project has been jointly supported by NSF, NASA, and NOAA at a modest level over several decades to promote international cooperation in research and scientific capacity building in developing regions. Since its founding in 1990, IGAC has been jointly sponsored by the International Geosphere-Biosphere Programme (IGBP) and the international Commission on Atmospheric Chemistry and Global Pollution (iCACGP). IGAC facilitates international scientific collaborations through research and community activities focused on deepening scientific understanding of atmospheric chemistry, the interactions of the atmosphere with other components in the Earth system including the biosphere, cryosphere, and oceans, and understanding the impacts of humans on atmospheric composition and chemistry. An important focus for IGAC is building scientific capacity in developing regions and fostering cooperation in the field of atmospheric chemistry around the globe, particularly through its biennial science conferences and young scientists programs.

Two sister IGBP programs to IGAC with significant atmospheric chemistry components are the **Surface Ocean-Lower Atmosphere Study (SOLAS)** and the **Integrated Land Ecosystem Atmospheric Processes Study (LEAPS)**. SOLAS focuses on understanding the key biogeochemical-physical interactions and feedbacks between the ocean and atmosphere that are central to the role that ocean-atmosphere interactions play in the regulation of climate and global change. ILEAPS focuses on enhancing understanding of interacting biological, chemical and physical processes that transport energy and matter through the land—atmosphere interface, with particular emphasis on the anthropogenic influence on these processes. Support of IGAC, SOLAS, and iLEAPS is one mechanism by which NSF should continue to engage scientists from around the world with the U.S. atmospheric chemistry community, and specifically promoting collaboration with atmospheric chemists in developing nations. As IGBP ends, a major new international research effort, Future Earth, has been developed that focuses on the development of a sustainable world, with three major themes: Dynamic Planet, Global Sustainable Development, and Transformations toward Sustainability. Sponsors include the International Council for Science (ICSU), the International Social Sciences Council (ISSC), and several United Nations bodies. Many of the former IGBP programs, including IGAC, SOLAS, and iLEAPS are currently transitioning to Future Earth.

A complementary approach to building global atmospheric chemistry capacity is supporting peer-to-peer relationships between scientists from the United States and those in developing countries to advance and implement measurement or modeling plans in developing countries. One possibility is establishing network sites for global scientific measurements, such as the AERONET (AErosol RObotic NETwork) program,[31] in developing countries or taking advantage of the Global Atmosphere Watch (GAW[32]) and its Integrated Global Atmospheric Chemistry Observations (IGACO) strategy under the World Meteorological Organization. Another is the transfer of measurement and modeling capability that is common in the United States for assessing air quality to scientists in developing nations where such capability is uncommon, recognizing that this may be determined in part by export controls and other restrictions. While NSF may be less prone to such restrictions, there is a need to develop a

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[31] AERONET: http://aeronet.gsfc.nasa.gov/.

[32] GAW is considered the atmospheric chemistry component of the Global Climate Observing System (GCOS); http://www.wmo.int/pages/prog/arep/gaw/gaw_home_en.html.

strategy to support collaborative relationships in the face of laws in both the United States and abroad that have the potential to hinder progress. Such activities both within NSF and across multiple U.S. agencies are very important for fostering global research programs and building global capacity in atmospheric chemistry.These collaborative plans should include joint participation in maintenance, quality control, and data analysis in addition to implementation of the tools, and they can involve training students from both countries. Maintaining this capacity beyond the initial development phase is vital in producing long-term observations and modeling data sets in regions for which the air quality and atmospheric composition can be rapidly changing.

NSF already does a substantial amount to promote international collaborations, for example, through the foundation-wide PIRE (Partnerships for International Research and Education) program or the USAID PEER (Partnerships for Engaged Enhancements in Research) program. PIRE supports international partnerships to address important science and engineering problems that require international collaboration, while helping to develop an internationally-engaged workforce. PEER grants are developed by scientists in developing countries to propose research with NSF-funded scientists to support research and capacity building activities on topics with strong potential development impacts.

International cooperative activities include participation in existing and emerging international coalitions, frequent international exchange and training of students, peer-to-peer collaborations between scientists in developing countries and the United States, and sharing of atmospheric chemistry expertise from the United States with peer scientists in developing countries.

A recent NSF Geosciences solicitation states that the geosciences “continue to lag other science, technology, engineering, and mathematics disciplines in the engagement, recruitment and retention of traditionally underrepresented and underserved minorities.”[33] Engaging U.S. researchers with a diverse range of backgrounds could foster innovation by incorporating a greater understanding of local contexts into the workforce. NSF is encouraged to support activities designed to engage underrepresented groups, either through nontraditional partnerships or through recruiting and retention of minority individuals.

Recommendation 6: NSF, in coordination with other agencies, should continue to encourage and support U.S. scientists involved in atmospheric chemistry research to engage with underserved groups, in capacity building activities, and in international collaborations.

6.5 ROLE OF A NATIONAL CENTER

The scope of the scientific problems facing atmospheric chemistry and the rest of atmospheric science are broad. As a result, the research required to solve these problems ranges from single PI investigations to large, multi-investigator, multi-discipline collaborations. In addition, the required observational research capability ranges from individual PI’s instruments in their laboratories to sophisticated field deployable research platforms, such as aircraft, containing several instruments that are engineered to meet the high standards required for field deployment. Few individual PIs have the resources or expertise to develop, maintain, and operate

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[33] http://www.nsf.gov/pubs/2016/nsf16516/nsf16516.htm.

an array of instruments and platforms over a long term that is required to answer today's (and tomorrow's) science questions. Similarly, few individual PIs are able to develop complex weather, chemistry, and climate models or to have access to high performance computational resources on which to run these models. Yet without these research platforms, models, and computational resources, the science described in this report cannot be accomplished.

A national center can be an optimal approach for providing these more complex and costly observational and computational capabilities because (1) dedicated center staff with expertise are most efficient at maintaining these complex capabilities; (2) NSF competitive processes are suitable for making support of these abilities available to individual or groups of PIs; and (3) the center can help foster the collaborative research needed to identify and solve critical science and societal problems. A national center can provide these resources and expertise to investigators in the broader community, primarily those served directly by NSF, while at the same time contributing directly to scientific advancements. This service to the community can be more than providing upon request atmospheric measurement capabilities, Earth system models, laboratory tools, or technical and scientific expertise that are difficult to maintain within universities or private companies. It can also be attracting and retaining intellectually leading scientists to the national center who are fully engaged with university faculty and their students by contributing to their research as well as involving them in research initiated within the center. At the same time, these center scientists can have opportunities to pursue their own research directions part of the time, and thus maintain their intellectual contributions at the cutting edge of the field. In this ideal center, service and science leadership are tightly connected. Rewarding staff scientists for their service to the atmospheric science community as much as their scientific and technical excellence provides visible recognition of that connection. In addition, a national center can bring the community together—by hosting a large steady flow of visitors of all stages in their careers, facilitating collaborations, holding conferences and workshops, and playing a leading role in the production of scientific assessments. This kind of partnership between the broader community and a national center that has essential, unique capabilities can be a critical component for advancing atmospheric chemistry and addressing the scientific gaps that are exposed in this report.

The National Center for Atmospheric Research (NCAR) is currently providing some of these capabilities that are needed from a national center. NCAR was established as a federally funded national center dedicated to achieving excellence in atmospheric science research (UCAR, 1959). NCAR's mission is "to understand the behavior of the atmosphere and related Earth and Geospace systems; to support, enhance, and extend the capabilities of the university community and the broader scientific community, nationally and internationally; and to foster the transfer of knowledge and technology for the betterment of life on Earth."[34]

Atmospheric chemistry is an essential part of understanding the behavior of the atmosphere, so that a national center for the atmospheric sciences such as NCAR must include a vibrant program in atmospheric chemistry research. Atmospheric chemistry research occurs within many divisions of NCAR, especially the Earth Observing Laboratory (EOL) and the Atmospheric Chemistry Observations and Modeling (ACOM) Laboratory. EOL has aircraft and ground systems for field studies and expertise in using these systems and interpreting their data. The Committee notes that an important fraction of the NSF-funded atmospheric chemistry research relies on NSF facilities administered at NCAR. NCAR's computational capability has supported many in the NSF Atmospheric Chemistry program. ACOM has strengths in modeling

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[34] https://ncar.ucar.edu/about-ncar.

across scales, in situ and satellite observations, instrumentation, and laboratory kinetics. Scientists within ACOM and its predecessor, the Atmospheric Chemistry Division, have historically provided prominent leadership roles within the broader atmospheric chemistry community and have been an integral part of the science research successes achieved in this field over the past five decades. The ACOM Advisory Committee[35] provides input from the broader academic community to NCAR. The atmospheric chemistry community regularly questions how well ACOM (as well as its predecessors) has been fulfilling this complete vision of its role.

Many on the Committee have observed that ACOM's capabilities have diminished in the past decade or so—including departures of some prominent atmospheric chemists. This situation occurred at the same time that NCAR ACOM scientists have been pushed to provide instruments, measurements, laboratory studies, and models for numerous projects, reducing the time for them to pursue their own research and/or development interests.

Nevertheless, the Committee believes that NCAR can be an even stronger partner with the atmospheric chemistry community by continuing to move its strategic vision closer to the Committee's vision of the roles of a national center given above and the original founding charter of NCAR. Some steps have been made within NCAR and ACOM in this direction, but in order to be the partner that the atmospheric chemistry community needs, NCAR must find its unique role in atmospheric chemistry research, one that complements and enhances the research being conducted by the broader atmospheric chemistry community and engages individual PIs from universities, federal labs, and the private sector. Visionary intellectual scientific leadership and a strategic allocation of resources will be needed from within NCAR to bring about necessary changes to fulfill this role.

In summary, the partnership between the competitively funded NSF atmospheric chemistry program and the facility-funded NSF programs at NCAR needs to be improved; doing so would facilitate greater scientific advances to close the science gaps presented in this report. As such, NSF and NCAR will need to work together to develop a strategy for improving this partnership and making NCAR a vibrant and complementary partner within the atmospheric chemistry community as a whole.

Recommendation 7: NCAR, in conjunction with NSF, should develop and implement a strategy to make NCAR a vibrant and complementary partner within the atmospheric chemistry community. This strategy should ensure that scientific leadership at NCAR has the latitude to set an energizing vision with appropriate personnel, infrastructure, and allocation of resources; and that the research capabilities and facilities at NCAR serve a unique and essential role to the NSF atmospheric chemistry community.

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[35] https://www2.acom.ucar.edu/about/advisory-committee.

Chapter 7—Final Thoughts

The field of atmospheric chemistry was born out of the need to respond to a series of societal challenges that were threatening human health and welfare—air pollution in urban centers, widely dispersed acid deposition, and a thinning stratospheric ozone layer. While atmospheric chemistry research has its foundations in the traditional disciplines of chemistry, physics, biology, geology, oceanography, engineering, and meteorology, as the field has grown it has become a robust area of basic science in and of itself.

As described in this report, atmospheric chemistry research is expanding into new frontiers. Atmospheric chemistry research today covers a wide swath of topics that integrate understanding the fundamental chemistry of the atmosphere, including emissions and transformations of gases and particles and how that chemistry affects and interacts with climate and weather, ecosystems, and people. The future challenges for atmospheric chemistry involve nothing less than the health of the planet's climate, the health of ecosystems, and the health of humans everywhere. As the Committee looks to the future, we see a deliberate shift in the field of atmospheric chemistry. This shift is to a full embrace of this dual role—on the one hand to observe, learn, and discover for the sake of fundamentally understanding the Earth system and the underlying chemical, physical and biological processes, and on the other hand to advance research to address those challenges that directly affect society. Atmospheric chemistry research alone cannot solve these major problems, but it is also true that they cannot be solved without it.

Moving in both of these directions will take effort, investment, and a willingness to adapt by the National Science Foundation, other agencies, and the atmospheric chemistry community as a whole. But after conversations with many members of our community during the course of this study, the Committee is convinced that we are ready for these challenges. The future of atmospheric chemistry research relies on the community to continue advancing our scientific knowledge and applying these findings to improve the world around us.

References

Abbatt, J. P. D., J. L. Thomas, K. Abrahamsson, C. Boxe, A. Granfors, A. E. Jones, M. D. King, A. Saiz-Lopez, P. B. Shepson, J. Sodeau, D. W. Toohey, C. Toubin, R. von Glasow, S. N. Wren, and X. Yang. 2012. Halogen activation via interactions with environmental ice and snow in the polar lower troposphere and other regions. *Atmospheric Chemistry and Physics* 12(14):6237-6271. DOI: 10.5194/acp-12-6237-2012.

Ahmadov, R., S. McKeen, M. Trainer, R. Banta, A. Brewer, S. Brown, P. M. Edwards, J. A. de Gouw, G. J. Frost, J. Gilman, D. Helmig, B. Johnson, A. Karion, A. Koss, A. Langford, B. Lerner, J. Olson, S. Oltmans, J. Peischl, G. Pétron, Y. Pichugina, J. M. Roberts, T. Ryerson, R. Schnell, C. Senff, C. Sweeney, C. Thompson, P. R. Veres, C. Warneke, R. Wild, E. J. Williams, B. Yuan, and R. J. Zamora. 2015. Understanding high wintertime ozone pollution events in an oil- and natural gas-producing region of the western US *Atmospheric Chemistry and Physics* 15:411-429. DOI: 10.5194/acp-15-411-2015.

Akimoto, H., J. Kurokawa, K. Sudo, T. Nagashima, T. Takemura, Z. Klimont, M. Amann, and K. Suzuki. 2015. SLCP co-control approach in East Asia: Tropospheric ozone reduction strategy by simultaneous reduction of NOx/NMVOC and methane. *Atmospheric Environment* 122:588-595. DOI: 10.1016/j.atmosenv.2015.10.003.

Al-Abadleh, H. A., and V. H. Grassian. 2003. Oxide surfaces as environmental interfaces. *Surface Science Reports* 52(3-4):63-161. DOI: 10.1016/j.surfrep.2003.09.001.

Al-Saadi, J., A. Soja, R. B. Pierce, J. Szykman, C. Wiedinmyer, L. Emmons, S. Kondragunta, X. Zhang, C. Kittaka, T. Schaack, and K. Bowman. 2008. Intercomparison of near-real-time biomass burning emissions estimates constrained by satellite fire data. *Journal of Applied Remote Sensing* 2. DOI: 10.1117/1.2948785.

Albrecht, B. A. 1989. Aerosols, Cloud Microphysics, and Fractional Cloudiness. *Science* 245(4923):1227-1230. DOI: 10.1126/science.245.4923.1227.

Allen, R. J. 2015. A 21st century northward tropical precipitation shift caused by future anthropogenic aerosol reductions. *Journal Of Geophysical Research-Atmospheres* 120(18):9087-9102. DOI: 10.1002/2015JD023623.

Allen, R. J., J. R. Norris, and M. Kovilakam. 2014. Influence of anthropogenic aerosols and the Pacific Decadal Oscillation on tropical belt width. *Nature geoscience* 7(4):270-274. DOI: 10.1038/ngeo2091.

Almeida, J., S. Schobesberger, A. Kurten, I. K. Ortega, O. Kupiainen-Maatta, A. P. Praplan, A. Adamov, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, J. Dommen, N. M. Donahue, A. Downard, E. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, R. Guida, J. Hakala, A. Hansel, M. Heinritzi, H. Henschel, T. Jokinen, H. Junninen, M. Kajos, J. Kangasluoma, H. Keskinen, A. Kupc, T. Kurten, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, M. Leiminger, J. Leppa, V. Loukonen, V. Makhmutov, S. Mathot, M. J. McGrath, T. Nieminen, T. Olenius, A. Onnela, T. Petaja, F. Riccobono, I. Riipinen, M. Rissanen, L. Rondo, T. Ruuskanen, F. D. Santos, N. Sarnela, S. Schallhart, R. Schnitzhofer, J. H. Seinfeld, M. Simon, M. Sipila, Y. Stozhkov, F. Stratmann, A. Tome, J. Trostl, G. Tsagkogeorgas, P. Vaattovaara, Y. Viisanen, A. Virtanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, C. Williamson, D. Wimmer, P. L. Ye, T. Yli-Juuti, K. S. Carslaw, M. Kulmala, J. Curtius, U. Baltensperger, D. R. Worsnop, H. Vehkamaki, and J. Kirkby. 2013. Molecular understanding of sulphuric acid-amine particle nucleation in the atmosphere. *Nature* 502(7471):359-+. DOI: 10.1038/nature12663.

Altieri, K. E., M. G. Hastings, A. J. Peters, S. Oleynik, and D. M. Sigman. 2014. Isotopic evidence for a marine ammonium source in rainwater at Bermuda. *Global Biogeochemical Cycles* 28(10):1066-1080. DOI: 10.1002/2014GB004809.

Anastasio, C., and A. L. Jordan. 2004. Photoformation of hydroxyl radical and hydrogen peroxide in aerosol particles from Alert, Nunavut: implications for aerosol and snowpack chemistry in the Arctic. *Atmospheric Environment* 38(8):1153-1166. DOI: 10.1016/j.atmosenv.2003.11.016.

Anderson, H. R., R. W. Atkinson, S. A. Bremner, J. Carrington, and J. Peacock. 2007. *Quantitative systematic review of short term associations between ambient air pollution (particulate matter, ozone, nitrogen dioxide, sulphur dioxide and carbon monoxide), and mortality and morbidity.* . London: Division of Community Health Sciences, St George's, University of London.

Anderson, J. G., D. W. Toohey, and W. H. Brune. 1991. Free radicals within the Antarctic vortex: The role of CFCs in Antarctic ozone loss. *Science* 251(4989):39-46. DOI: 10.1126/science.251.4989.39.

Anderson, J. G., D. M. Wilmouth, J. B. Smith, and D. S. Sayres. 2012. UV Dosage Levels in Summer: Increased Risk of Ozone Loss from Convectively Injected Water Vapor. *Science* 337(6096):835-839. DOI: 10.1126/science.1222978.

Andreae, M. O., and P. J. Crutzen. 1997. Atmospheric aerosols: Biogeochemical sources and role in atmospheric chemistry. *Science* 276(5315):1052-1058. DOI: 10.1126/science.276.5315.1052.

Andreae, M. O., D. Rosenfeld, P. Artaxo, A. A. Costa, G. P. Frank, K. M. Longo, and M. A. F. Silva-Dias. 2004. Smoking rain clouds over the Amazon. *Science* 303(5662):1337-1342. DOI: 10.1126/science.1092779.

Appel, K. W., A. B. Gilliland, G. Sarwar, and R. C. Gilliam. 2007. Evaluation of the Community Multiscale Air Quality (CMAQ) model version 4.5: Sensitivities impacting model performance Part I - Ozone. *Atmospheric Environment* 41(40):9603-9615. DOI: 10.1016/j.atmosenv.2007.08.044.

Araujo, J. A. 2011a. Particulate air pollution, systemic oxidative stress, inflammation, and atherosclerosis. *Air Quality Atmosphere and Health* 4(1):79-93. DOI: 10.1007/s11869-010-0101-8.

Araujo, J. A. 2011b. Are Ultrafine Particles a Risk Factor for Cardiovascular Diseases? *Revista Espanola De Cardiologia* 64(8):642-645. DOI: 10.1016/j.rec.2011.05.006.

Ariya, P. A., M. Amyot, A. Dastoor, D. Deeds, A. Feinberg, G. Kos, A. Poulain, A. Ryjkov, K. Semeniuk, M. Subir, and K. Toyota. 2015. Mercury Physicochemical and Biogeochemical Transformation in the Atmosphere and at Atmospheric Interfaces: A Review and Future Directions. *Chemical Reviews* 115(10):3760-3802. DOI: 10.1021/cr500667e.

Arneth, A., U. Niinemets, S. Pressley, J. Back, P. Hari, T. Karl, S. Noe, I. C. Prentice, D. Serca, T. Hickler, A. Wolf, and B. Smith. 2007. Process-based estimates of terrestrial ecosystem isoprene emissions: incorporating the effects of a direct CO2-isoprene interaction. *Atmospheric Chemistry and Physics* 7:31-53.

Atkinson, J. D., B. J. Murray, M. T. Woodhouse, T. F. Whale, K. J. Baustian, K. S. Carslaw, S. Dobbie, D. O'Sullivan, and T. L. Malkin. 2013. The importance of feldspar for ice nucleation by mineral dust in mixed-phase clouds. *Nature* 498(7454):355-358. DOI: 10.1038/nature12278.

Atkinson, R., and J. Arey. 2003. Atmospheric degradation of volatile organic compounds. *Chemical Reviews* 103(12):4605-4638. DOI: 10.1021/cr0206420.

Ault, A. P., C. R. Williams, A. B. White, P. J. Neiman, J. M. Creamean, C. J. Gaston, F. M. Ralph, and K. A. Prather. 2011. Detection of Asian dust in California orographic precipitation. *Journal Of Geophysical Research-Atmospheres* 116. DOI: 10.1029/2010jd015351.

Auvray, M., I. Bey, E. Llull, M. G. Schultz, and S. Rast. 2007. A model investigation of tropospheric ozone chemical tendencies in long-range transported pollution plumes. *Journal Of Geophysical Research-Atmospheres* 112(D5). DOI: 10.1029/2006jd007137.

Avnery, S., D. L. Mauzerall, J. F. Liu, and L. W. Horowitz. 2011. Global crop yield reductions due to surface ozone exposure: 1. Year 2000 crop production losses and economic damage. *Atmos. Env.* 45(13):2284-2296. DOI: 10.1016/j.atmosenv.2010.11.045.

Baergen, A. M., and D. J. Donaldson. 2013. Photochemical Renoxification of Nitric Acid on Real Urban Grime. *Environmental Science & Technology* 47(2):815-820. DOI: 10.1021/es3037862.

Baker, A. R., and P. L. Croot. 2010. Atmospheric and marine controls on aerosol iron solubility in seawater. *Marine Chemistry* 120(1-4):4-13. DOI: 10.1016/j.marchem.2008.09.003.

Baker, A. R., M. French, and K. L. Linge. 2006. Trends in aerosol nutrient solubility along a west-east transect of the Saharan dust plume. *Geophysical Research Letters* 33(7). DOI: 10.1029/2005gl024764.

Baklanov, A., K. Schlunzen, P. Suppan, J. Baldasano, D. Brunner, S. Aksoyoglu, G. Carmichael, J. Douros, J. Flemming, R. Forkel, S. Galmarini, M. Gauss, G. Grell, M. Hirtl, S. Joffre, O. Jorba, E. Kaas, M. Kaasik, G. Kallos, X. Kong, U. Korsholm, A. Kurganskiy, J. Kushta, U. Lohmann, A. Mahura, A. Manders-Groot, A. Maurizi, N. Moussiopoulos, S. T. Rao, N. Savage, C. Seigneur, R. S. Sokhi, E. Solazzo, S. Solomos, B. Sorensen, G. Tsegas, E. Vignati, B. Vogel, and Y. Zhang. 2014. Online coupled regional meteorology chemistry models in Europe: current status and prospects. *Atmospheric Chemistry and Physics* 14(1):317-398. DOI: 10.5194/acp-14-317-2014.

Barahona, D. 2012. On the ice nucleation spectrum. *Atmospheric Chemistry and Physics* 12(8):3733-3752. DOI: 10.5194/acp-12-3733-2012.

Barberán, A., J. Ladau, J. W. Leff, K. S. Pollard, H. L. Menninger, R. R. Dunn, and N. Fierer. 2015. Continental-scale distributions of dust-associated bacteria and fungi. *Proceedings of the National Academy of Sciences of the United States of America* 112(18):5756-5761. DOI: 10.1073/pnas.1420815112.

Barnes, E. A., and L. Polvani. 2013. Response of the Midlatitude Jets, and of Their Variability, to Increased Greenhouse Gases in the CMIP5 Models. *Journal of Climate* 26(18):7117-7135. DOI: 10.1175/Jcli-D-12-00536.1.

Barnes, E. A., and A. M. Fiore. 2013. Surface ozone variability and the jet position: Implications for projecting future air quality. *Geophysical Research Letters* 40(11):2839-2844. DOI: 10.1002/grl.50411.

Barnosky, A. D., E. A. Hadly, J. Bascompte, E. L. Berlow, J. H. Brown, M. Fortelius, W. M. Getz, J. Harte, A. Hastings, P. A. Marquet, N. D. Martinez, A. Mooers, P. Roopnarine, G. Vermeij, J. W. Williams, R. Gillespie, J. Kitzes, C. Marshall, N. Matzke, D. P. Mindell,

E. Revilla, and A. B. Smith. 2012. Approaching a state shift in Earth's biosphere. *Nature* 486(7401):52-58. DOI: 10.1038/nature11018.

Barrett, S. R. H., R. L. Speth, S. D. Eastham, I. C. Dedoussi, A. Ashok, R. Malina, and D. W. Keith. 2015. Impact of the Volkswagen emissions control defeat device on US public health. *Environmental Research Letters* 10(11). DOI: 10.1088/1748-9326/10/11/114005.

Barrie, L. A., J. W. Bottenheim, R. C. Schnell, P. J. Crutzen, and R. A. Rasmussen. 1988. Ozone Destruction and Photochemical-Reactions at Polar Sunrise in the Lower Arctic Atmosphere. *Nature* 334(6178):138-141. DOI: 10.1038/334138a0.

Bateman, A. P., A. K. Bertram, and S. T. Martin. 2015. Hygroscopic Influence on the Semisolid-to-Liquid Transition of Secondary Organic Materials. *Journal of Physical Chemistry A* 119(19):4386-4395. DOI: 10.1021/jp508521c.

Bates, D. R., and P. B. Hays. 1967. Atmospheric Nitrous Oxide. *Planetary and Space Science* 15(1):189-&. DOI: 10.1016/0032-0633(67)90074-8.

Bauer, S. E., A. Ault, and K. A. Prather. 2013. Evaluation of aerosol mixing state classes in the GISS-modelE-MATRIX climate model using single particle mass spectrometry measurements. *Journal of Geophysical Research* 118:9834-9844. DOI: 10.1002/jgrd.50700.

Baustian, K. J., D. J. Cziczo, M. E. Wise, K. A. Pratt, G. Kulkarni, A. G. Hallar, and M. A. Tolbert. 2012. Importance of aerosol composition, mixing state, and morphology for heterogeneous ice nucleation: A combined field and laboratory approach. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2011jd016784.

Beamish, R. J., and H. H. Harvey. 1972. Acidification of the La Cloche Mountain Lakes, Ontario, and resulting fish mortalities. *Journal of the Fisheries Board of Canada* 29(8):1131-1143.

Beem, K. B., S. Raja, F. M. Schwandner, C. Taylor, T. Lee, A. P. Sullivan, C. M. Carrico, G. R. McMeeking, D. Day, E. Levin, J. Hand, S. M. Kreidenweis, B. Schichtel, W. C. Malm, and J. L. Collett. 2010. Deposition of reactive nitrogen during the Rocky Mountain Airborne Nitrogen and Sulfur (RoMANS) study. *Environmental Pollution* 158(3):862-872. DOI: 10.1016/j.envpol.2009.09.023.

Behrenfeld, M. J., R. T. O'Malley, D. A. Siegel, C. R. McClain, J. L. Sarmiento, G. C. Feldman, A. J. Milligan, P. G. Falkowski, R. M. Letelier, and E. S. Boss. 2006. Climate-driven trends in contemporary ocean productivity. *Nature* 444(7120):752-755. DOI: 10.1038/nature05317.

Behzad, H., T. Gojobori, and K. Mineta. 2015. Challenges and Opportunities of Airborne Metagenomics. *Genome Biology and Evolution* 7(5):1216-1226. DOI: 10.1093/gbe/evv064.

Bell, M. L., D. L. Davis, and T. Fletcher. 2004. A retrospective assessment of mortality from the London smog episode of 1952: The role of influenza and pollution. *Environmental Health Perspectives* 112(1):6-8. DOI: 10.1289/ehp.6539.

Bell, T. L., D. Rosenfeld, K. M. Kim, J. M. Yoo, M. I. Lee, and M. Hahnenberger. 2008. Midweek increase in US summer rain and storm heights suggests air pollution invigorates rainstorms. *Journal Of Geophysical Research-Atmospheres* 113(D2). DOI: 10.1029/2007jd008623.

Berezin, E. V., I. B. Konovalov, P. Ciais, A. Richter, S. Tao, G. Janssens-Maenhout, M. Beekmann, and E. D. Schulze. 2013. Multiannual changes of CO_2 emissions in China:

indirect estimates derived from satellite measurements of tropospheric NO_2 columns. *Atmospheric Chemistry and Physics* 13(18):9415-9438. DOI: 10.5194/acp-13-9415-2013.

Berhane, K., C.-C. Chang, R. McConnell, W. J. Gauderman, E. Avol, E. Rapapport, R. Urman, F. Lurmann, and F. Gilliland. 2016. Association of changes in air quality with bronchitic symptoms in children in California, 1993-2012. *Journal of the American Medical Association* 315(15):1491-1501. DOI: 10.1001/jama.2016.3444.

Berndt, T., T. Jokinen, M. Sipila, R. L. Mauldin, H. Herrmann, F. Stratmann, H. Junninen, and M. Kulmala. 2014. H_2SO_4 formation from the gas-phase reaction of stabilized Criegee Intermediates with SO_2: Influence of water vapour content and temperature. *Atmospheric Environment* 89:603-612. DOI: 10.1016/j.atmosenv.2014.02.062.

Bertram, T. H., J. A. Thornton, T. P. Riedel, A. M. Middlebrook, R. Bahreini, T. S. Bates, P. K. Quinn, and D. J. Coffman. 2009. Direct observations of N_2O_5 reactivity on ambient aerosol particles. *Geophysical Research Letters* 36. DOI: 10.1029/2009gl040248.

Bianchi, F., J. Trostl, H. Junninen, C. Frege, S. Henne, C. R. Hoyle, U. Molteni, E. Herrmann, A. Adamov, N. Bukowiecki, X. Chen, J. Duplissy, M. Gysel, M. Hutterli, J. Kangasluoma, J. Kontkanen, A. Kurten, H. E. Manninen, S. Munch, O. Perakyla, T. Petaja, L. Rondo, C. Williamson, E. Weingartner, J. Curtius, D. R. Worsnop, M. Kulmala, J. Dommen, and U. Baltensperger. 2016. New particle formation in the free troposphere: A question of chemistry and timing. *Science* 352(6289):1109-1112. DOI: 10.1126/science.aad5456.

Bingham, A., and E. Porter. 2015. Ozone effects on two ecosystem services at Great Smoky Mountains National Park, USA. *Park Science* 32(1):71-79. Available at http://www.nature.nps.gov/ParkScience/archive/PDF/Article_PDFs/ParkScience32(71)Summer2015_2071-2079_Bingham_Porter_3825.pdf, accessed April 2018, 2016.

Biswas, K. F., B. M. Ghauri, and L. Husain. 2008. Gaseous and aerosol pollutants during fog and clear episodes in South Asian urban atmosphere. *Atmospheric Environment* 42(33):7775-7785. DOI: 10.1016/j.atmosenv.2008.04.056.

Blanchard, D. C. 1964. Sea-to-Air Transport of Surface Active Material. *Science* 146(364):396-&. DOI: 10.1126/science.146.3642.396.

Bobbink, R., M. Hornung, and J. G. M. Roelofs. 1998. The effects of air-borne nitrogen pollutants on species diversity in natural and semi-natural European vegetation. *Journal of Ecology* 86(5):717-738. DOI: 10.1046/j.1365-2745.1998.8650717.x.

Bocquet, M., H. Elbern, H. Eskes, M. Hirtl, R. Zabkar, G. R. Carmichael, J. Flemming, A. Inness, M. Pagowski, J. L. P. Camano, P. E. Saide, R. San Jose, M. Sofiev, J. Vira, A. Baklanov, C. Carnevale, G. Grell, and C. Seigneur. 2015. Data assimilation in atmospheric chemistry models: current status and future prospects for coupled chemistry meteorology models. *Atmospheric Chemistry and Physics* 15(10):5325-5358. DOI: 10.5194/acp-15-5325-2015.

Bollasina, M. A., Y. Ming, and V. Ramaswamy. 2011. Anthropogenic Aerosols and the Weakening of the South Asian Summer Monsoon. *Science* 334(6055):502-505. DOI: 10.1126/science.1204994.

Bollasina, M. A., Y. Ming, and V. Ramaswamy. 2013. Earlier onset of the Indian monsoon in the late twentieth century: The role of anthropogenic aerosols. *Geophysical Research Letters* 40(14):3715-3720. DOI: 10.1002/grl.50719.

Bollasina, M. A., Y. Ming, V. Ramaswamy, M. D. Schwarzkopf, and V. Naik. 2014. Contribution of local and remote anthropogenic aerosols to the twentieth century weakening of the South Asian Monsoon. *Geophysical Research Letters* 41(2):680-687. DOI: 10.1002/2013GL058183.

Bonan, G. B. 2008. Forests and climate change: Forcings, feedbacks, and the climate benefits of forests. *Science* 320(5882):1444-1449. DOI: 10.1126/science.1155121.

Bond, T. C., S. J. Doherty, D. W. Fahey, P. M. Forster, T. Berntsen, B. J. DeAngelo, M. G. Flanner, S. Ghan, B. Karcher, D. Koch, S. Kinne, Y. Kondo, P. K. Quinn, M. C. Sarofim, M. G. Schultz, M. Schulz, C. Venkataraman, H. Zhang, S. Zhang, N. Bellouin, S. K. Guttikunda, P. K. Hopke, M. Z. Jacobson, J. W. Kaiser, Z. Klimont, U. Lohmann, J. P. Schwarz, D. Shindell, T. Storelvmo, S. G. Warren, and C. S. Zender. 2013. Bounding the role of black carbon in the climate system: A scientific assessment. *Journal Of Geophysical Research-Atmospheres* 118(11):5380-5552. DOI: 10.1002/jgrd.50171.

Booker, F., R. Muntifering, M. McGrath, K. Burkey, D. Decoteau, E. Fiscus, W. Manning, S. Krupa, A. Chappelka, and D. Grantz. 2009. The Ozone Component of Global Change: Potential Effects on Agricultural and Horticultural Plant Yield, Product Quality and Interactions with Invasive Species. *Journal of Integrative Plant Biology* 51(4):337-351. DOI: 10.1111/j.1744-7909.2008.00805.x.

Booth, B. B. B., N. J. Dunstone, P. R. Halloran, T. Andrews, and N. Bellouin. 2012. Aerosols implicated as a prime driver of twentieth-century North Atlantic climate variability. *Nature* 484(7393):228-U110. DOI: 10.1038/nature10946.

Boucher, O., D. Randall, P. Artaxo, C. Bretherton, G. Feingold, P. Forster, V.-M. Kerminen, Y. Kondo, H. Liao, U. Lohmann, P. Rasch, S. K. Satheesh, S. Sherwood, B. Stevens, and X. Y. Zhang. 2013. Clouds and Aerosols. In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P. M. Midgley, eds. Cambridge, United Kingdom and New York: Cambridge University Press.

Bowman, K., and D. K. Henze. 2012. Attribution of direct ozone radiative forcing to spatially resolved emissions. *Geophysical Research Letters* 39. DOI: 10.1029/2012gl053274.

Boyd, P. W., T. Jickells, C. S. Law, S. Blain, E. A. Boyle, K. O. Buesseler, K. H. Coale, J. J. Cullen, H. J. W. de Baar, M. Follows, M. Harvey, C. Lancelot, M. Levasseur, N. P. J. Owens, R. Pollard, R. B. Rivkin, J. Sarmiento, V. Schoemann, V. Smetacek, S. Takeda, A. Tsuda, S. Turner, and A. J. Watson. 2007. Mesoscale iron enrichment experiments 1993-2005: Synthesis and future directions. *Science* 315(5812):612-617. DOI: 10.1126/science.1131669.

Boyer, J. S., P. Byrne, K. G. Cassman, M. Cooper, D. Delmer, T. Greene, F. Gruis, J. Habben, N. Hausmann, N. Kenny, R. Lafitte, S. Paszkiewicz, D. Porter, A. Schlegel, J. Schussler, T. Setter, J. Shanahan, R. E. Sharp, T. J. Vyn, D. Warner, and J. Gaffney. 2013. The US drought of 2012 in perspective: A call to action. *Global Food Security-Agriculture Policy Economics and Environment* 2(3):139-143. DOI: 10.1016/j.gfs.2013.03.002.

Brauer, M. 2015. Air pollution, stroke and anxiety. *BMJ* 350:1-2. DOI: 10.1136/bmj.h1510.

Brewer, A. W. 1949. Evidence for a World Circulation Provided by the Measurements of Helium and Water Vapour Distribution in the Stratosphere. *Quarterly Journal of the Royal Meteorological Society* 75(326):351-363. DOI: 10.1002/qj.49707532603.

Brimblecombe, P. 1978. Interest in Air Pollution Among Early Fellows of the Royal Society. *Notes and Records of the Royal Society of London* 32(2):123-129.

Brioude, J., W. M. Angevine, R. Ahmadov, S. W. Kim, S. Evan, S. A. McKeen, E. Y. Hsie, G. J. Frost, J. A. Neuman, I. B. Pollack, J. Peischl, T. B. Ryerson, J. Holloway, S. S. Brown, J. B. Nowak, J. M. Roberts, S. C. Wofsy, G. W. Santoni, T. Oda, and M. Trainer. 2013. Top-down estimate of surface flux in the Los Angeles Basin using a mesoscale inverse modeling technique: assessing anthropogenic emissions of CO, NOx and CO2 and their impacts. *Atmospheric Chemistry and Physics* 13(7):3661-3677. DOI: 10.5194/acp-13-3661-2013.

Britigan, N., A. Alshawa, and S. A. Nizkorodov. 2006. Quantification of ozone levels in indoor environments generated by ionization and ozonolysis air purifiers. *Journal of the Air & Waste Management Association* 56(5):601-610.

Broadley, S. L., B. J. Murray, R. J. Herbert, J. D. Atkinson, S. Dobbie, T. L. Malkin, E. Condliffe, and L. Neve. 2012. Immersion mode heterogeneous ice nucleation by an illite rich powder representative of atmospheric mineral dust. *Atmospheric Chemistry and Physics* 12:287-3017. DOI: 10.5194/acp-12-287-2012.

Brown, N. 1989. Climate, ecology, and international security. *Survival: Global Politics and Strategy* 31(6):519-532. DOI: 10.1080/00396338908442497.

Brown, P. 1995. Race, Class, and Environmental-Health - a Review and Systematization of the Literature. *Environmental Research* 69(1):15-30. DOI: 10.1006/enrs.1995.1021.

Brown, S. S., and J. Stutz. 2012. Nighttime radical observations and chemistry. *Chemical Society Reviews* 41(19):6405-6447. DOI: 10.1039/c2cs35181a.

Brune, W. H., B. C. Baier, J. Thomas, X. Ren, R. C. Cohen, S. E. Pusede, E. C. Browne, A. H. Goldstein, D. R. Gentner, F. N. Keutsch, J. A. Thornton, S. Harrold, F. D. Lopez-Hilfiker, and P. O. Wennberg. 2016. Ozone production chemistry in the presence of urban plumes. *Faraday Discussions*. DOI: 10.1039/C5FD00204D.

Burkholder, J. B., R. A. Cox, and A. R. Ravishankara. 2015. Atmospheric Degradation of Ozone Depleting Substances, Their Substitutes, and Related Species. *Chemical Reviews* 115(10):3704-3759. DOI: 10.1021/cr5006759.

Burling, I. R., R. J. Yokelson, S. K. Akagi, S. P. Urbanski, C. E. Wold, D. W. T. Griffith, T. J. Johnson, J. Reardon, and D. R. Weise. 2012. Airborne and ground-based measurements of the trace gases and particles emitted by prescribed fires in the United States (vol 11, pg 12197, 2011). *Atmospheric Chemistry and Physics* 12(1):103-103. DOI: 10.5194/acp-12-103-2012.

Burrows, S. M., W. Elbert, M. G. Lawrence, and U. Poschl. 2009a. Bacteria in the global atmosphere - Part 1: Review and synthesis of literature data for different ecosystems. *Atmospheric Chemistry and Physics* 9(23):9263-9280. DOI: 10.5194/acp-9-9263-2009.

Burrows, S. M., T. Butler, P. Jockel, H. Tost, A. Kerkweg, U. Poschl, and M. G. Lawrence. 2009b. Bacteria in the global atmosphere - Part 2: Modeling of emissions and transport between different ecosystems. *Atmospheric Chemistry and Physics* 9(23):9281-9297. DOI: 10.5194/acp-9-9281-2009.

Burrows, S. M., C. Hoose, U. Poschl, and M. G. Lawrence. 2013. Ice nuclei in marine air: biogenic particles or dust? *Atmospheric Chemistry and Physics* 13(1):245-267. DOI: 10.5194/acp-13-245-2013.

Busby, J. W. 2007. *Climate Change and National Security: An Agenda for Action. Council Special Report No. 32*. Washington DC: Council on Foreign Relations Press.

Busby, J. W. 2008. Who Cares about the Weather?: Climate Change and U.S. National Security. *Security Studies* 17(3):468-504. DOI: 10.1080/09636410802319529.

Bzdek, B. R., M. R. Pennington, and M. V. Johnston. 2012. Single particle chemical analysis of ambient ultrafine aerosol: A review. *Journal of Aerosol Science* 52:109-120. DOI: 10.1016/j.jaerosci.2012.05.001.

Bzdek, B. R., A. J. Horan, M. R. Pennington, J. W. DePalma, J. Zhao, C. N. Jen, D. R. Hanson, J. N. Smith, P. H. McMurry, and M. V. Johnston. 2013. Quantitative and time-resolved nanoparticle composition measurements during new particle formation. *Faraday Discussions* 165:25-43. DOI: 10.1039/c3fd00039g.

Calderon-Garciduenas, L., and R. Torres-Jardon. 2015. The Impact of Air Pollutants on the Brain. *Jama Psychiatry* 72(6):529-530. DOI: 10.1001/jamapsychiatry.2015.0192.

Calderon-Garciduenas, L., R. Torres-Jardon, R. J. Kulesza, S. B. Park, and A. D'Angiulli. 2014. Air pollution and detrimental effects on children's brain. The need for a multidisciplinary approach to the issue complexity and challenges. *Frontiers in Human Neuroscience* 8. DOI: 10.3389/fnhum.2014.00613.

Calvert, J., J. Orlando, W. Stockwell, and T. Wallington. 2015. *The Mechanisms of Reactions Influencing Atmospheric Ozone*. New York: Oxford University Press.

Camredon, M., B. Aumont, J. Lee-Taylor, and S. Madronich. 2007. The SOA/VOC/NOx system: an explicit model of secondary organic aerosol formation. *Atmospheric Chemistry and Physics* 7(21):5599-5610.

Canagaratna, M. R., J. T. Jayne, J. L. Jimenez, J. D. Allan, M. R. Alfarra, Q. Zhang, T. B. Onasch, F. Drewnick, H. Coe, A. Middlebrook, A. Delia, L. R. Williams, A. M. Trimborn, M. J. Northway, P. F. DeCarlo, C. E. Kolb, P. Davidovits, and D. R. Worsnop. 2007. Chemical and microphysical characterization of ambient aerosols with the aerodyne aerosol mass spectrometer. *Mass Spectrom. Rev.* 26(2):185-222. DOI: 10.1002/mas.20115

Cantrell, C. A., G. D. Edwards, S. Stephens, L. Mauldin, E. Kosciuch, M. Zondlo, and F. Eisele. 2003. Peroxy radical observations using chemical ionization mass spectrometry during TOPSE. *Journal Of Geophysical Research-Atmospheres* 108(D6). DOI: 10.1029/2002jd002715.

CAPCOA (California Air Pollution Control Officers' Association). 2015. California's Progress Towards Clean Air. A report by the California Air Pollution Control Officers' Association. Available at http://www.capcoa.org/wp-content/uploads/2015/04/2015%20PTCA%20CAPCOA%20Report%20-%20FINAL.pdf. Available accessed.

Cappa, C. D., T. B. Onasch, P. Massoli, D. R. Worsnop, T. S. Bates, E. S. Cross, P. Davidovits, J. Hakala, K. L. Hayden, B. T. Jobson, K. R. Kolesar, D. A. Lack, B. M. Lerner, S. M. Li, D. Mellon, I. Nuaaman, J. S. Olfert, T. Petaja, P. K. Quinn, C. Song, R. Subramanian, E. J. Williams, and R. A. Zaveri. 2012. Radiative Absorption Enhancements Due to the Mixing State of Atmospheric Black Carbon. *Science* 337(6098):1078-1081. DOI: 10.1126/science.1223447.

Carpenter, L. J., and P. D. Nightingale. 2015. Chemistry and Release of Gases from the Surface Ocean. *Chemical Reviews* 115(10):4015-4034. DOI: 10.1021/cr5007123.

Carpenter, L. J., K. Hebestreit, U. Platt, and P. S. Liss. 2001. Coastal zone production of IO precursors: a 2-dimensional study. *Atmospheric Chemistry and Physics* 1:9-18.

Carpenter, L. J., S. D. Archer, and R. Beale. 2012. Ocean-atmosphere trace gas exchange. *Chemical Society Reviews* 41(19):6473-6506. DOI: 10.1039/c2cs35121h.

Carrió, G. G., W. R. Cotton, and W. Y. Y. Cheng. 2010. Urban growth and aerosol effects on convection over Houston. *Atmospheric Research* 96(4):560-574. DOI: 10.1016/j.atmosres.2010.01.005.

Carslaw, N., D. J. Creasey, D. Harrison, D. E. Heard, M. C. Hunter, P. J. Jacobs, M. E. Jenkin, J. D. Lee, A. C. Lewis, M. J. Pilling, S. M. Saunders, and P. W. Seakins. 2001. OH and HO_2 radical chemistry in a forested region of north-western Greece. *Atmospheric Environment* 35(27):4725-4737. DOI: 10.1016/S1352-2310(01)00089-9.

Carslaw, K. S., L. A. Lee, C. L. Reddington, K. J. Pringle, A. Rap, P. M. Forster, G. W. Mann, D. V. Spracklen, M. T. Woodhouse, L. A. Regayre, and J. R. Pierce. 2013. Large contribution of natural aerosols to uncertainty in indirect forcing. *Nature* 503(7474):67-71. DOI: 10.1038/nature12674.

Cash, D. W., W. C. Clark, F. Alcock, N. M. Dickson, N. Eckley, D. H. Guston, J. Jager, and R. B. Mitchell. 2003. Knowledge systems for sustainable development. *Proceedings of the National Academy of Sciences of the United States of America* 100(14):8086-8091. DOI: 10.1073/pnas.1231332100.

Cazorla, M., W. H. Brune, X. Ren, and B. Lefer. 2012. Direct measurement of ozone production rates in Houston in 2009 and comparison with two estimation methods. *Atmospheric Chemistry and Physics* 12(2):1203-1212. DOI: 10.5194/acp-12-1203-2012.

Chai, T. F., G. R. Carmichael, A. Sandu, Y. H. Tang, and D. N. Daescu. 2006. Chemical data assimilation of transport and chemical evolution over the Pacific (TRACE-P) aircraft measurements. *Journal Of Geophysical Research-Atmospheres* 111(D2). DOI: 10.1029/2005jd005883.

Changnon, Jr. S. A. 1981. METROMEX review and summary. *Meteor. Monogr.* 40:181.

Chappelka, A. H., and L. J. Samuelson. 1998. Ambient ozone effects on forest trees of the eastern United States: a review. *New Phytologist* 139(1):91-108. DOI: 10.1046/j.1469-8137.1998.00166.x.

Charlson, R. J., J. E. Lovelock, M. O. Andreae, and S. G. Warren. 1987. Oceanic phytoplankton, atmospheric sulfur, cloud albedo and climate. *Nature* 326(6114):655-661. DOI: 10.1038/326655a0.

Charrier, J. G., and C. Anastasio. 2011. Impacts of antioxidants on hydroxyl radical production from individual and mixed transition metals in a surrogate lung fluid. *Atmospheric Environment* 45(40):7555-7562. DOI: 10.1016/j.atmosenv.2010.12.021.

Chatani, S., N. Shimo, S. Matsunaga, Y. Kajii, S. Kato, Y. Nakashima, K. Miyazaki, K. Ishii, and H. Ueno. 2009. Sensitivity analyses of OH missing sinks over Tokyo metropolitan area in the summer of 2007. *Atmospheric Chemistry and Physics* 9(22):8975-8986. DOI: 10.5194/acp-9-8975-2009.

Chen, J.-C., and J. Schwartz. 2009. Neurobehavioral effects of ambient air pollution on cognitive performance in US adults. *Neurotoxicology* 30(2):231-239. DOI: 10.1016/j.neuro.2008.12.011.

Chen, Q., Y. J. Liu, N. M. Donahue, J. E. Shilling, and S. T. Martin. 2011. Particle-Phase Chemistry of Secondary Organic Material: Modeled Compared to Measured O:C and H:C Elemental Ratios Provide Constraints. *Environmental Science & Technology* 45(11):4763-4770. DOI: 10.1021/es104398s.

Chen, Z. H., M. T. Salam, S. P. Eckel, C. V. Breton, and F. D. Gilliland. 2015. Chronic effects of air pollution on respiratory health in Southern California children: findings from the

Southern California Children's Health Study. *Journal of Thoracic Disease* 7(1):46-+. DOI: 10.3978/j.issn.2072-1439.2014.12.20.

Cheng, C.-T., W.-C. Wang, and J.-P. Chen. 2010. Simulation of the effects of increasing cloud condensation nuclei on mixed-phase clouds and precipitation of a front system. *Atmospheric Research* 96(461-476). DOI: 10.1016/j.atmosres.2010.02.005.

Cheng, W. Y. Y., G. G. Carrio, W. R. Cotton, and S. M. Saleeby. 2009. Influence of cloud condensation and giant cloud condensation nuclei on the development of precipitating trade wind cumuli in a large eddy simulation. *Journal Of Geophysical Research-Atmospheres* 114. DOI: 10.1029/2008jd011011.

Chien, C.-T., K. R. M. Mackey, S. Dutkiewicz, N. M. Mahowald, J. M. Prospero, and A. Paytan. 2016. Effects of African dust deposition on phytoplankton in the western tropical Atlantic Ocean off Barbados. *Global Biogeochemical Cycles* 30(5):716-734. DOI: 10.1002/2015GB005334.

Christian, T. J., R. J. Yokelson, J. A. Carvalho, D. W. T. Griffith, E. C. Alvarado, J. C. Santos, T. G. S. Neto, C. A. G. Veras, and W. M. Hao. 2007. The tropical forest and fire emissions experiment: Trace gases emitted by smoldering logs and dung from deforestation and pasture fires in Brazil. *Journal Of Geophysical Research-Atmospheres* 112(D18). DOI: 10.1029/2006JD008147.

Chuang, P. Y., R. M. Duvall, M. M. Shafer, and J. J. Schauer. 2005. The origin of water soluble particulate iron in the Asian atmospheric outflow. *Geophysical Research Letters* 32(7). DOI: 10.1029/2004gl021946.

Chubachi, S. 1985. A Special Ozone Observation at Syowa Station, Antarctica from February 1982 to January 1983. In *Atmospheric Ozone: Proceedings of the Quadrennial Ozone Symposium held in Halkidiki, Greece 3–7 September 1984*. C. S. Zerefos and A. Ghazi, eds. Dordrecht: Springer Netherlands.

Ciais, P., C. Sabine, G. Bala, L. Bopp, V. Brovkin, J. Canadell, A. Chhabra, R. DeFries, J. Galloway, M. Heimann, C. Jones, C. L. Quéré, R. B. Myneni, S. Piao, and P. Thornton. 2013. Carbon and Other Biogeochemical Cycles. In *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change*. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P. M. Midgley, eds. Cambridge, United Kingdom and New York, NY: Cambridge University Press.

Cicerone, R. J., S. Walters, and S. C. Liu. 1983. Non-Linear Response of Stratospheric Ozone Column to Chlorine Injections. *Journal of Geophysical Research-Oceans and Atmospheres* 88(Nc6):3647-3661. DOI: 10.1029/Jc088ic06p03647.

Clarisse, L., C. Clerbaux, F. Dentener, D. Hurtmans, and P. F. Coheur. 2009. Global ammonia distribution derived from infrared satellite observations. *Nature geoscience* 2(7):479-483. DOI: 10.1038/ngeo551.

Clarke, J. F., E. S. Edgerton, and B. E. Martin. 1997. Dry Deposition Calculations for the Clean Air Status and Trends Network. *Atmospheric Environment* 31(21):3367-3678.

Cleveland, W. S., T. E. Graedel, B. Kleiner, and J. L. Warner. 1974. Sunday and Workday Variations in Photochemical Air-Pollutants in New-Jersey and New-York. *Science* 186(4168):1037-1038. DOI: 10.1126/science.186.4168.1037.

CNA MAB (Center for Naval Analaysis Military Advisory Board. 2014. *National Security and the Accelerating Risks of Climate Change*. Alexandria, VA: CNA Corporation.

Cofala, J., M. Amann, Z. Klimont, K. Kupiainen, and L. Hoglund-Isaksson. 2007. Scenarios of global anthropogenic emissions of air pollutants and methane until 2030. *Atmospheric Environment* 41(38):8486-8499. DOI: 10.1016/j.atmosenv.2007.07.010.

Coleman, B. K., H. Destaillats, A. T. Hodgson, and W. W. Nazaroff. 2008. Ozone consumption and volatile byproduct formation from surface reactions with aircraft cabin materials and clothing fabrics. *Atmospheric Environment* 42(4):642-654. DOI: 10.1016/j.atmosenv.2007.10.001.

Conley, S., G. Franco, I. Faloona, D. R. Blake, J. Peischl, and T. B. Ryerson. 2016. Methane emissions from the 2015 Aliso Canyon blowout in Los Angeles, CA. *Science* 351(6279):1317-1320. DOI: 10.1126/science.aaf2348.

Connolly, P. J., O. Mohler, P. R. Field, H. Saathoff, R. Burgess, T. Choularton, and M. Gallagher. 2009. Studies of heterogeneous freezing by three different desert dust samples. *Atmospheric Chemistry and Physics* 9(8):2805-2824. DOI: 10.5194/acp-9-2805-2009.

Cox, P. M., P. P. Harris, C. Huntingford, R. A. Betts, M. Collins, C. D. Jones, T. E. Jupp, J. A. Marengo, and C. A. Nobre. 2008. Increasing risk of Amazonian drought due to decreasing aerosol pollution. *Nature* 453(7192):212-U217. DOI: 10.1038/nature06960.

Cramer, W., A. Bondeau, F. I. Woodward, I. C. Prentice, R. A. Betts, V. Brovkin, P. M. Cox, V. Fisher, J. A. Foley, A. D. Friend, C. Kucharik, M. R. Lomas, N. Ramankutty, S. Sitch, B. Smith, A. White, and C. Young-Molling. 2001. Global response of terrestrial ecosystem structure and function to CO_2 and climate change: results from six dynamic global vegetation models. *Global Change Biology* 7(4):357-373. DOI: 10.1046/j.1365-2486.2001.00383.x.

Creamean, J. M., K. J. Suski, D. Rosenfeld, A. Cazorla, P. J. DeMott, R. C. Sullivan, A. B. White, F. M. Ralph, P. Minnis, J. M. Comstock, J. M. Tomlinson, and K. A. Prather. 2013. Dust and Biological Aerosols from the Sahara and Asia Influence Precipitation in the Western U.S. *Science* 339(6127):1572-1578. DOI: 10.1126/science.1227279.

Criegee, R. 1948. Die Umlagerung Der Dekalin-Peroxydester Als Folge Von Kationischem Sauerstoff. *Annalen Der Chemie-Justus Liebig* 560(1-2):127-135. DOI: 10.1002/jlac.19485600106.

Crounse, J. D., F. Paulot, H. G. Kjaergaard, and P. O. Wennberg. 2011. Peroxy radical isomerization in the oxidation of isoprene. *Physical Chemistry Chemical Physics* 13(30):13607-13613. DOI: 10.1039/c1cp21330j.

Crounse, J. D., H. C. Knap, K. B. Ornso, S. Jorgensen, F. Paulot, H. G. Kjaergaard, and P. O. Wennberg. 2012. Atmospheric Fate of Methacrolein. 1. Peroxy Radical Isomerization Following Addition of OH and O_2. *Journal of Physical Chemistry A* 116(24):5756-5762. DOI: 10.1021/jp211560u.

Crounse, J. D., L. B. Nielsen, S. Jorgensen, H. G. Kjaergaard, and P. O. Wennberg. 2013. Autoxidation of Organic Compounds in the Atmosphere. *Journal of Physical Chemistry Letters* 4(20):3513-3520. DOI: 10.1021/jz4019207.

Crutzen, P. J. 1970. The influence of nitrogen oxides on the atmospheric ozone content. *Quarterly Journal of the Royal Meteorological Society* 96(408):320-325. DOI: 10.1002/qj.49709640815.

Crutzen, P. J. 1971. Ozone Production Rates in an Oxygen-Hydrogen-Nitrogen Oxide Atmosphere. *Journal of Geophysical Research* 76(30):7311-7327. DOI: 10.1029/Jc076i030p07311.

Crutzen, P. J., and F. Arnold. 1986. Nitric-Acid Cloud Formation in the Cold Antarctic Stratosphere - a Major Cause for the Springtime Ozone Hole. *Nature* 324(6098):651-655. DOI: 10.1038/324651a0.

Crutzen, P. J., and J. Lelieveld. 2001. Human Impacts on Atmospheric Chemistry. *Annual Review of Earth and Planetary Sciences* 29(17-45). DOI: 10.1146/annurev.earth.29.1.17.

Crutzen, P. J., and E. F. Stoermer. 2000. The "Anthropocene." *Global Change Newsletter* 41:17-18.

Cziczo, D. J., K. D. Froyd, C. Hoose, E. J. Jensen, M. H. Diao, M. A. Zondlo, J. B. Smith, C. H. Twohy, and D. M. Murphy. 2013. Clarifying the Dominant Sources and Mechanisms of Cirrus Cloud Formation. *Science* 340(6138):1320-1324. DOI: 10.1126/science.1234145.

Davidovits, P., C. E. Kolb, L. R. Williams, J. T. Jayne, and D. R. Worsnop. 2011. Update 1 of: Mass Accommodation and Chemical Reactions at Gas-Liquid Interfaces. *Chemical Reviews* 111:Pr76-Pr109. DOI: 10.1021/cr100360b.

Davidson, P., K. Schere, R. Draxler, S. Kondragunta, R. A. Wayland, J. F. Meagher, and R. Mathur. 2008. Toward a US National Air Quality Forecast Capability: Current and Planned Capabilities. *Air Pollution Modeling and Its Application XIX*:226-234. DOI: 10.1007/978-1-4020-8453-9_25.

de Gouw, J. A., A. M. Middlebrook, C. Warneke, P. D. Goldan, W. C. Kuster, J. M. Roberts, F. C. Fehsenfeld, D. R. Worsnop, M. R. Canagaratna, A. A. P. Pszenny, W. C. Keene, M. Marchewka, S. B. Bertman, and T. S. Bates. 2005. Budget of organic carbon in a polluted atmosphere: Results from the New England Air Quality Study in 2002. *Journal Of Geophysical Research-Atmospheres* 110(D16). DOI: 10.1029/2004jd005623.

de Groot, R., L. Brander, S. van der Ploeg, R. Costanza, F. Bernard, L. Braat, M. Christie, N. Crossman, A. Ghermandi, L. Hein, S. Hussain, P. Kumar, A. McVittie, R. Portela, L. C. Rodriguez, P. ten Brink, and P. van Beukering. 2012. Global estimates of the value of ecosystems and their services in monetary units. *Ecosystem Services* 1(1):50-61.

DeMott, P. J., K. Sassen, M. R. Poellot, D. Baumgardner, D. C. Rogers, S. D. Brooks, A. J. Prenni, and S. M. Kreidenweis. 2003a. African dust aerosols as atmospheric ice nuclei. *Geophysical Research Letters* 30(14). DOI: 10.1029/2003gl017410.

DeMott, P. J., D. J. Cziczo, A. J. Prenni, D. M. Murphy, S. M. Kreidenweis, D. S. Thomson, R. Borys, and D. C. Rogers. 2003b. Measurements of the concentration and composition of nuclei for cirrus formation. *Proceedings of the National Academy of Sciences of the United States of America* 100(25):14655-14660. DOI: 10.1073/pnas.2532677100.

DeMott, P. J., T. C. J. Hill, C. S. McCluskey, K. A. Prather, D. B. Collins, R. C. Sullivan, M. J. Ruppel, R. H. Mason, V. E. Irish, T. Lee, C. Y. Hwang, T. S. Rhee, J. R. Snider, G. R. McMeeking, S. Dhaniyala, E. R. Lewis, J. J. B. Wentzell, J. Abbatt, C. Lee, C. M. Sultana, A. P. Ault, J. L. Axson, M. D. Martinez, I. Venero, G. Santos-Figueroa, M. D. Stokes, G. B. Deane, O. L. Mayol-Bracero, V. H. Grassian, T. H. Bertram, A. K. Bertram, B. F. Moffett, and G. D. Franc. 2016. Sea spray aerosol as a unique source of ice nucleating particles. *Proceedings of the National Academy of Sciences of the United States of America* 113(21):5797-5803. DOI: 10.1073/pnas.1514034112.

DePalma, J. W., D. J. Doren, and M. V. Johnston. 2014. Formation and Growth of Molecular Clusters Containing Sulfuric Acid, Water, Ammonia, and Dimethylamine. *Journal of Physical Chemistry A* 118(29):5464-5473. DOI: 10.1021/jp503348b.

Deroubaix, A., N. Martiny, I. Chiapello, and B. Marticorena. 2013. Suitability of OMI aerosol index to reflect mineral dust surface conditions: Preliminary application for studying the link with meningitis epidemics in the Sahel. *Remote Sensing of Environment* 133:116-127. DOI: 10.1016/j.rse.2013.02.009.

Despres, V. R., J. A. Huffman, S. M. Burrows, C. Hoose, A. S. Safatov, G. Buryak, J. Frohlich-Nowoisky, W. Elbert, M. O. Andreae, U. Poschl, and R. Jaenicke. 2012. Primary biological aerosol particles in the atmosphere: a review. *Tellus Series B-Chemical and Physical Meteorology* 64. DOI: 10.3402/Tellusb.V64i0.15598.

Di Carlo, P., W. H. Brune, M. Martinez, H. Harder, R. Lesher, X. R. Ren, T. Thornberry, M. A. Carroll, V. Young, P. B. Shepson, D. Riemer, E. Apel, and C. Campbell. 2004. Missing OH reactivity in a forest: Evidence for unknown reactive biogenic VOCs. *Science* 304(5671):722-725. DOI: 10.1126/science.1094392.

Dobson, G. M. B. 1956. Origin and Distribution of the Polyatomic Molecules in the Atmosphere. *Proceedings of the Royal Society of London Series a-Mathematical and Physical Sciences* 236(1205):187-193. DOI: 10.1098/rspa.1956.0127.

DOD (Department of Defense). 2014. *Climate Change Adaptation Roadmap*. Alexandria, VA: Office of the Deputy Under Secretary of Defense for Installations and Environment.

Doherty, R. M., O. Wild, D. T. Shindell, G. Zeng, I. A. MacKenzie, W. J. Collins, A. M. Fiore, D. S. Stevenson, F. J. Dentener, M. G. Schultz, P. Hess, R. G. Derwent, and T. J. Keating. 2013. Impacts of climate change on surface ozone and intercontinental ozone pollution: A multi-model study. *Journal Of Geophysical Research-Atmospheres* 118(9):3744-3763. DOI: 10.1002/jgrd.50266.

Donahue, N. M., A. L. Robinson, C. O. Stanier, and S. N. Pandis. 2006. Coupled partitioning, dilution, and chemical aging of semivolatile organics. *Environmental Science & Technology* 40(8):2635-2643. DOI: 10.1021/es052297c.

Donahue, N. M., E. R. Trump, J. R. Pierce, and I. Riipinen. 2011. Theoretical constraints on pure vapor-pressure driven condensation of organics to ultrafine particles. *Geophysical Research Letters* 38. DOI: 10.1029/2011gl048115.

Donahue, N. M., J. H. Kroll, S. N. Pandis, and A. L. Robinson. 2012. A two-dimensional volatility basis set – Part 2: Diagnostics of organic-aerosol evolution *Atmospheric Chemistry and Physics* 12:615-634. DOI: 10.5194/acp-12-615-2012.

Donaldson, D. J., and K. T. Valsaraj. 2010. Adsorption and Reaction of Trace Gas-Phase Organic Compounds on Atmospheric Water Film Surfaces: A Critical Review. *Environmental Science & Technology* 44(3):865-873.

Douwes, J., P. Thorne, N. Pearce, and D. Heederik. 2003. Bioaerosol health effects and exposure assessment: Progress and prospects. *Annals of Occupational Hygiene* 47(3):187-200. DOI: 10.1093/annhyg/meg032.

Driscoll, C. T., G. B. Lawrence, A. J. Bulger, T. J. Butler, C. S. Cronan, C. Eagar, K. F. Lambert, G. E. Likens, J. L. Stoddard, and K. C. Weathers. 2001. Acidic Deposition in the Northeastern United States: Sources and Inputs, Ecosystem Effects, and Management Strategies *BioScience* 51(3). DOI: 10.1641/0006-3568(2001)051[0180:ADITNU]2.0.CO;2.

Du, J., and L. Zhu. 2011. Quantification of the absorption cross sections of surface-adsorbed nitric acid in the 335-365 nm region by Brewster angle cavity ring-down spectroscopy. *Chemical Physics Letters* 511(4-6):213-218. DOI: 10.1016/j.cplett.2011.06.062.

Du, J., R. G. Keesee, and L. Zhu. 2014. Experimental Study of the Competitive Adsorption of HNO3 and H2O on Surfaces by Using Brewster Angle Cavity Ring-Down Spectroscopy in the 295-345 nm Region. *Journal of Physical Chemistry A* 118(37):8177-8181. DOI: 10.1021/jp500913e.

Duce, R. A., J. LaRoche, K. Altieri, K. R. Arrigo, A. R. Baker, D. G. Capone, S. Cornell, F. Dentener, J. Galloway, R. S. Ganeshram, R. J. Geider, T. Jickells, M. M. Kuypers, R. Langlois, P. S. Liss, S. M. Liu, J. J. Middelburg, C. M. Moore, S. Nickovic, A. Oschlies, T. Pedersen, J. Prospero, R. Schlitzer, S. Seitzinger, L. L. Sorensen, M. Uematsu, O. Ulloa, M. Voss, B. Ward, and L. Zamora. 2008. Impacts of atmospheric anthropogenic nitrogen on the open ocean. *Science* 320(5878):893-897. DOI: 10.1126/science.1150369.

Dukic, V., M. Hayden, A. A. Forgor, T. Hopson, P. Akweongo, A. Hodgson, A. Monaghan, C. Wiedinmyer, T. Yoksas, M. C. Thomson, S. Trzaska, and R. Pandya. 2012. The Role of Weather in Meningitis Outbreaks in Navrongo, Ghana: A Generalized Additive Modeling Approach. *Journal of Agricultural Biological and Environmental Statistics* 17(3):442-460. DOI: 10.1007/s13253-012-0095-9.

Dunstone, N. J., D. M. Smith, B. B. B. Booth, L. Hermanson, and R. Eade. 2013. Anthropogenic aerosol forcing of Atlantic tropical storms. *Nature geoscience* 6(7):534-539. DOI: 10.1038/NGEO1854.

Dutkiewicz, V. A., L. Husain, U. K. Roychowdhury, and K. L. Demerjian. 2011. Impact of Canadian wildfire smoke on air quality at two rural sites in NY State. *Atmospheric Environment* 45(12):2028-2033. DOI: 10.1016/j.atmosenv.2011.01.072.

Ehn, M., J. A. Thornton, E. Kleist, M. Sipila, H. Junninen, I. Pullinen, M. Springer, F. Rubach, R. Tillmann, B. Lee, F. Lopez-Hilfiker, S. Andres, I. H. Acir, M. Rissanen, T. Jokinen, S. Schobesberger, J. Kangasluoma, J. Kontkanen, T. Nieminen, T. Kurten, L. B. Nielsen, S. Jorgensen, H. G. Kjaergaard, M. Canagaratna, M. Dal Maso, T. Berndt, T. Petaja, A. Wahner, V. M. Kerminen, M. Kulmala, D. R. Worsnop, J. Wildt, and T. F. Mentel. 2014. A large source of low-volatility secondary organic aerosol. *Nature* 506(7489):476-. DOI: 10.1038/nature13032.

EIA (Energy Information Administration). 2015. *Annual Energy Outlook 2015. DOE/EIA-0383(2015)*. Washington, DC: US Energy Information Administration.

Ellis, R. A., D. J. Jacob, M. P. Sulprizio, L. Zhang, C. D. Holmes, B. A. Schichtel, T. Blett, E. Porter, L. H. Pardo, and J. A. Lynch. 2013. Present and future nitrogen deposition to national parks in the United States: critical load exceedances. *Atmos. Chem. Phys.* 13(17):9083-9095. DOI: 10.5194/acp-13-9083-2013.

Emmerson, K. M., N. Carslaw, L. J. Carpenter, D. E. Heard, J. D. Lee, and M. J. Pilling. 2005. Urban atmospheric chemistry during the PUMA campaign 1: Comparison of modelled OH and HO2 concentrations with measurements. *Journal of Atmospheric Chemistry* 52(2):143-164. DOI: 10.1007/s10874-005-1322-3.

Emmerson, K. M., N. Carslaw, D. C. Carslaw, J. D. Lee, G. McFiggans, W. J. Bloss, T. Gravestock, D. E. Heard, J. Hopkins, T. Ingham, M. J. Pilling, S. C. Smith, M. Jacob, and P. S. Monks. 2007. Free radical modelling studies during the UK TORCH Campaign in Summer 2003. *Atmospheric Chemistry and Physics* 7:167-181.

Enami, S., M. R. Hoffmann, and A. J. Colussi. 2015. OH-Radical Specific Addition to Glutathione S-Atom at the Air-Water Interface: Relevance to the Redox Balance of the Lung Epithelial Lining Fluid. *Journal of Physical Chemistry Letters* 6(19):3935-3943. DOI: 10.1021/acs.jpclett.5b01819.

EPA (Environmental Protection Agency). 2009. *Final Report: Integrated Science Assessment for Particulate Matter. EPA/600/R-08/139F*. Washington, DC: U.S. Environmental Protection Agency.

EPA. 2012. *Regulatory Impact Analysis for the Final Revisions to the National Ambient Air Quality Standards for Particulate Matter, EPA-452/R-12-005*. Washington, DC: U. S. Environmental Protection Agency.

EPA. 2013. *Final Report: Integrated Science Assessment of Ozone and Related Photochemical Oxidants. EPA/600/R-10/076F*. Washington, DC: U.S. Environmental Protection Agency.

EPA. 2014a. *Health Risk and Exposure Assessment for Ozone*. Washington, DC: EPA.

EPA. 2014b. *Welfare Risk and Exposure Assessment for Ozone*. Washington, DC: EPA.

Erisman, J. W., A. Bleeker, J. Galloway, and M. S. Sutton. 2007. Reduced nitrogen in ecology and the environment. *Environmental Pollution* 150(1):140-149. DOI: 10.1016/j.envpol.2007.06.033.

Erisman, J. W., M. A. Sutton, J. Galloway, Z. Klimont, and W. Winiwater. 2008. How a century of ammonia synthesis changed the world. *Nature geoscience* 1(10):636-639. DOI: 10.1038/ngeo325.

Ervens, B. 2015. Modeling the Processing of Aerosol and Trace Gases in Clouds and Fogs. *Chemical Reviews* 115(10):4157-4198. DOI: 10.1021/cr5005887.

Eskes, H., V. Huijnen, A. Arola, A. Benedictow, A. M. Blechschmidt, E. Botek, O. Boucher, I. Bouarar, S. Chabrillat, E. Cuevas, R. Engelen, H. Flentje, A. Gaudel, J. Griesfeller, L. Jones, J. Kapsomenakis, E. Katragkou, S. Kinne, B. Langerock, M. Razinger, A. Richter, M. Schultz, M. Schulz, N. Sudarchikova, V. Thouret, M. Vrekoussis, A. Wagner, and C. Zerefos. 2015. Validation of reactive gases and aerosols in the MACC global analysis and forecast system. *Geoscientific Model Development* 8(11):3523-3543. DOI: 10.5194/gmd-8-3523-2015.

Evelyn, J. 1661. *Fumifugum, or, The inconveniencie of the aer and smoak of London dissipated together with some remedies humbly proposed by J.E. esq. to His Sacred Majestie, and to the Parliament now assembled.* Available at *http://www.gyford.com/archive/2009/04/28/www.geocities.com/Paris/LeftBank/1914/fumifug.html, accessed October 14, 2015*.

Everts, S., and M. Davenport. 2016. Drones swarm to science: Flying robots are doing experiments too hazardous, too expensive, or simply impossible for humans. *Chemical & Engineering News* 94(9):32-33.

Exley, C., J. S. Chappell, and J. D. Birchall. 1991. A mechanism for acute aluminum toxicity in fish. *Journal of Theoretical Biology* 151(3):417-428. DOI: 10.1016/s0022-5193(05)80389-3.

Fan, J., L. R. Leung, D. Rosenfeld, Q. Chen, Z. Li, J. Zhang, and H. Yan. 2013. Microphysical effects determine macrophysical response for aerosol impacts on deep convective clouds. *Proceedings of the National Academy of Sciences of the United States of America* 110(48):E4581-4590. DOI: 10.1073/pnas.1316830110.

Fares, S., M. McKay, R. Holzinger, and A. H. Goldstein. 2010. Ozone fluxes in a Pinus ponderosa ecosystem are dominated by non-stomatal processes: Evidence from long-term continuous measurements. *Agricultural and Forest Meteorology* 150(3):420-431. DOI: 10.1016/j.agrformet.2010.01.007.

Farman, J. C., B. G. Gardiner, and J. D. Shanklin. 1985. Large Losses of Total Ozone in Antarctica Reveal Seasonal ClOx/NOx Interaction. *Nature* 315(6016):207-210. DOI: 10.1038/315207a0.

Farmer, D. K., C. D. Cappa, and S. M. Kreidenweis. 2015. Atmospheric processes and their controlling influence on cloud condensation nuclei activity. *Chemical Reviews* 115(10):4199-4217. DOI: 10.1021/cr5006292.

Feiner, P. A., W. H. Brune, D. O. Miller, L. Zhang, R. C. Cohen, P. Romer, A. H. Goldstein, F. N. Keutsch, K. M. Skog, P. O. Wennberg, T. B. Nguyen, T. A.P, J. A. d. Gouw, A. Koss, R. J. Wild, S. S. Brown, A. Guenther, E. Edgerton, K. Baumann, and J. L. Fry. 2016. Testing atmospheric oxidation in an Alabama Forest. *Journal of the Atmospheric Sciences*(submitted).

Felzer, B. S., T. Cronin, J. M. Reilly, J. M. Melilloa, and X. D. Wang. 2007. Impacts of ozone on trees and crops. *Comptes Rendus Geoscience* 339(11-12):784-798. DOI: 10.1016/j.crte.2007.08.008.

Feng, Y., V. Ramanathan, and V. R. Kotamarthi. 2013. Brown carbon: a significant atmospheric absorber of solar radiation? *Atmospheric Chemistry and Physics* 13(17):8607-8621. DOI: 10.5194/acp-13-8607-2013.

Fernstrom, A., and M. Goldblatt. 2013. Aerobiology and Its Role in the Transmission of Infectious Diseases. *Journal of Pathogens* 2013(Article ID 493960). DOI: 10.1155/2013/493960.

Field, B. C., and M. K. Field. 2009. *Environmental Economics: An Introduction*. New York: McGraw-Hill.

Fierce, L., N. Riemer, and T. C. Bond. 2013. When is cloud condensation nuclei activity sensitive to particle characteristics at emission? *Journal Of Geophysical Research-Atmospheres* 118(24):13476-13488. DOI: 10.1002/2013JD020608.

Fingar, T. 2008. National Intelligence Assessment on the National Implications of Global Climate Change to 2030. Presented to the US House of Representatives, 25 June 2008, Washington DC. Available at https://fas.org/irp/congress/2008_hr/062508fingar.pdf, accessed October 19, 2015.

Finlayson-Pitts, B. J., and J. N. Pitts. 2000. *Chemistry of the upper and lower atmosphere: Theory, experiments, and applications*. San Diego: Academic Press.

Fiore, A. M., F. J. Dentener, O. Wild, C. Cuvelier, M. G. Schultz, P. Hess, C. Textor, M. Schulz, R. M. Doherty, L. W. Horowitz, I. A. MacKenzie, M. G. Sanderson, D. T. Shindell, D. S. Stevenson, S. Szopa, R. Van Dingenen, G. Zeng, C. Atherton, D. Bergmann, I. Bey, G. Carmichael, W. J. Collins, B. N. Duncan, G. Faluvegi, G. Folberth, M. Gauss, S. Gong, D. Hauglustaine, T. Holloway, I. S. A. Isaksen, D. J. Jacob, J. E. Jonson, J. W. Kaminski, T. J. Keating, A. Lupu, E. Marmer, V. Montanaro, R. J. Park, G. Pitari, K. J. Pringle, J. A. Pyle, S. Schroeder, M. G. Vivanco, P. Wind, G. Wojcik, S. Wu, and A. Zuber. 2009. Multimodel estimates of intercontinental source-receptor relationships for ozone pollution. *Journal Of Geophysical Research-Atmospheres* 114. DOI: 10.1029/2008jd010816.

Fiscus, E. L., F. L. Booker, and K. O. Burkey. 2005. Crop responses to ozone: uptake, modes of action, carbon assimilation and partitioning. *Plant Cell and Environment* 28(8):997-1011. DOI: 10.1111/j.1365-3040.2005.01349.x.

Forrister, H., J. Liu, E. Scheuer, J. Dibb, L. Ziemba, K. L. Thornhill, B. Anderson, G. Diskin, A. E. Perring, J. P. Schwarz, P. Campuzano-Jost, D. A. Day, B. B. Palm, J. L. Jimenez, A.

Nenes, and R. J. Weber. 2015. Evolution of brown carbon in wildfire plumes. *Geophysical Research Letters* 42(11):4623-4630. DOI: 10.1002/2015GL063897.

Franco, B., E. Mahieu, L. Emmons, Z. Tzompa-Sosa, E. Fischer, K. Sudo, B. Bovy, S. Conway, D. Griffin, J. Hannigan, K. Strong, and K. Walker. 2016. Evaluating ethane and methane emissions associated with the development of oil and natural gas extraction in North America *Environmental Research Letters* 11. DOI: 10.1088/1748-9326/11/4/044010.

Franco, V., F. P. Sánchez, J. German, and P. Mock. 2014. *Real-world exhaust emissions from modern diesel cars: A meta-analysis of PEMS emissions data from EU (euro 6) and US (tier 2 bin 5/ULEV II) diesel passenger cars. Part 1: Aggregated results*. Available at http://www.theicct.org/sites/default/files/publications/ICCT_PEMS-study_diesel-cars_20141013.pdf, accessed April 15, 2016. Berlin: International Council on Clean Transportation.

Friedl, M. A., D. K. McIver, J. C. F. Hodges, X. Y. Zhang, D. Muchoney, A. H. Strahler, C. E. Woodcock, S. Gopal, A. Schneider, A. Cooper, A. Baccini, F. Gao, and C. Schaaf. 2002. Global land cover mapping from MODIS: algorithms and early results. *Remote Sensing of Environment* 83(1-2):287-302.

Froyd, K. D., S. M. Murphy, D. M. Murphy, J. A. d. Gouw, N. C. Eddingsass, and P. O. Wennberg. 2010. Contribution of isoprene-derived organosulfates to free tropospheric aerosol mass. *Proceedings of the National Academy of Sciences of the United States of America* 107(50):21360-21365.

Fuchs, H., B. Bohn, A. Hofzumahaus, F. Holland, K. D. Lu, S. Nehr, F. Rohrer, and A. Wahner. 2011. Detection of HO_2 by laser-induced fluorescence: calibration and interferences from RO_2 radicals. *Atmospheric Measurement Techniques* 4(6):1209-1225. DOI: 10.5194/amt-4-1209-2011.

Fuchs, H., Z. F. Tan, A. Hofzumahaus, S. Broch, H. P. Dorn, F. Holland, C. Kunstler, S. Gomm, F. Rohrer, S. Schrade, R. Tillmann, and A. Wahner. 2016. Investigation of potential interferences in the detection of atmospheric RO_x radicals by laser-induced fluorescence under dark conditions. *Atmospheric Measurement Techniques* 9(4):1431-1447. DOI: 10.5194/amt-9-1431-2016.

Gabey, A. M., M. W. Gallagher, J. Whitehead, J. R. Dorsey, P. H. Kaye, and W. R. Stanley. 2010. Measurements and comparison of primary biological aerosol above and below a tropical forest canopy using a dual channel fluorescence spectrometer. *Atmospheric Chemistry and Physics* 10(10):4453-4466. DOI: 10.5194/acp-10-4453-2010.

Gaffney, J. S., and N. A. Marley. 2003. Atmospheric chemistry and air pollution. *The Scientific World Journal* 3:199-234.

Garcia, R. R., D. E. Kinnison, and D. R. Marsh. 2012. "World avoided" simulations with the Whole Atmosphere Community Climate Model. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2012jd018430.

Garner, G. G., and A. M. Thompson. 2012. The Value of Air Quality Forecasting in the Mid-Atlantic Region. *Weather, Climate, and Society* 4:69-79. DOI: 10.1175/WCAS-D-10-05010.1.

Gasso, S., A. Stein, F. Marino, E. Castellano, R. Udisti, and J. Ceratto. 2010. A combined observational and modeling approach to study modern dust transport from the Patagonia desert to East Antarctica. *Atmospheric Chemistry and Physics* 10(17):8287-8303. DOI: 10.5194/acp-10-8287-2010.

Gauderman, W. J. 2006. Air pollution and children - An unhealthy mix. *N. Engl. J. Med.* 355(1):78-79.

Gautam, R., N. C. Hsu, S. C. Tsay, K. M. Lau, B. Holben, S. Bell, A. Smirnov, C. Li, R. Hansell, Q. Ji, S. Payra, D. Aryal, R. Kayastha, and K. M. Kim. 2011. Accumulation of aerosols over the Indo-Gangetic plains and southern slopes of the Himalayas: distribution, properties and radiative effects during the 2009 pre-monsoon season. *Atmospheric Chemistry and Physics* 11(24):12841-12863. DOI: 10.5194/acp-11-12841-2011.

Ge, X. L., A. S. Wexler, and S. L. Clegg. 2011. Atmospheric amines - Part I. A review. *Atmospheric Environment* 45(3):524-546. DOI: 10.1016/j.atmosenv.2010.10.012.

Gentner, D. R., G. Isaacman, D. R. Worton, A. W. H. Chan, T. R. Dallmann, L. Davis, S. Liu, D. A. Day, L. M. Russell, K. R. Wilson, R. Weber, A. Guha, R. A. Harley, and A. H. Goldstein. 2012. Elucidating secondary organic aerosol from diesel and gasoline vehicles through detailed characterization of organic carbon emissions. *Proceedings of the National Academy of Sciences of the United States of America* 109(45):18318-18323. DOI: 10.1073/pnas.1212272109.

George, C., M. Ammann, B. D'Anna, D. J. Donaldson, and S. A. Nizkorodov. 2015. Heterogeneous Photochemistry in the Atmosphere. *Chemical Reviews* 115(10):4218-4258. DOI: 10.1021/cr500648z.

Gettelman, A., X. Liu, D. Barahona, U. Lohmann, and C. Chen. 2012. Climate impacts of ice nucleation. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2012jd017950.

Ghan, S. J., G. Guzman, and H. Abdul-Razzak. 1998. Competition between sea salt and sulfate particles as cloud condensation nuclei. *Journal of the Atmospheric Sciences* 55(22):3340-3347. DOI: 10.1175/1520-0469(1998)055<3340:Cbssas>2.0.Co;2.

Giglio, L., I. Csiszar, and C. O. Justice. 2006. Global distribution and seasonality of active fires as observed with the Terra and Aqua Moderate Resolution Imaging Spectroradiometer (MODIS) sensors. *Journal of Geophysical Research-Biogeosciences* 111(G2). DOI: 10.1029/2005jg000142.

Gil, Y., M. Chan, B. Gomez, and B. Caron. 2014. *EarthCube: Past, Present, and Future. EarthCube Project Report EC-2014-3*. Arlington, VA: National Science Foundation.

Gilbert, N. 2016. Drought devastates US crops: Impact on food prices likely to be felt around the world. *Nature*. DOI: 10.1038/nature.2012.11065.

Gilliland, A. B., C. Hogrefe, R. W. Pinder, J. M. Godowitch, K. L. Foley, and S. T. Rao. 2008. Dynamic evaluation of regional air quality models: Assessing changes in O_3 stemming from changes in emissions and meteorology. *Atmospheric Environment* 42(20):5110-5123. DOI: 10.1016/j.atmosenv.2008.02.018.

Ginoux, P., J. M. Prospero, T. E. Gill, N. C. Hsu, and M. Zhao. 2012. Global-scale attribution of anthropogenic and natural dust sources and their emission rates based on MODIS Deep Blue aerosol products. *Reviews of Geophysics* 50(3):36. DOI: 10.1029/2012rg000388.

Glasoe, W. A., K. Volz, B. Panta, N. Freshour, R. Bachman, D. R. Hanson, P. H. McMurry, and C. Jen. 2015. Sulfuric acid nucleation: An experimental study of the effect of seven bases. *Journal Of Geophysical Research-Atmospheres* 120(5):1933-1950. DOI: 10.1002/2014JD022730.

Gligorovski, S., and C. J. Weschler. 2013. The Oxidative Capacity of Indoor Atmospheres. *Environmental Science & Technology* 47(24):13905-13906. DOI: 10.1021/es404928t.

Gligorovski, S., H. Wortham, and J. Kleffmann. 2014. The hydroxyl radical (OH) in indoor air: Sources and implications. *Atmospheric Environment* 99:568-570. DOI: 10.1016/j.atmosenv.2014.10.031.

Goldstein, A. H., and I. E. Galbally. 2007. Known and unexplored organic constituents in the earth's atmosphere. *Environmental Science & Technology* 41(5):1514-1521. DOI: Doi 10.1021/Es072476p.

Gonzalez, P. A., M. Minkler, A. P. Garcia, M. Gordon, C. Garzon, M. Palaniappan, S. Prakash, and B. Beveridge. 2011. Community-Based Participatory Research and Policy Advocacy to Reduce Diesel Exposure in West Oakland, California. *American Journal of Public Health* 101:S166-S175. DOI: 10.2105/Ajph.2010.196204.

Goodhill, V. 1971. Maimonides--Modern Medical Relevance. *Transactions--American Academy of Ophthalmology and Otolaryngology*:463.

Grannas, A. M., P. B. Shepson, C. Guimbaud, A. L. Sumner, M. Albert, W. Simpson, F. Domine, H. Boudries, J. Bottenheim, H. J. Beine, R. Honrath, and X. L. Zhou. 2002. A study of photochemical and physical processes affecting carbonyl compounds in the Arctic atmospheric boundary layer. *Atmospheric Environment* 36(15-16):2733-2742. DOI: 10.1016/S1352-2310(02)00134-6.

Grenier, L. 1998. *Working with Indigenous Knowledge: A Guide for Researchers*. Ottowa, Ont., Canada: International Development Research Centre.

Grgic, I., Nieto-Gligorovski, L.I., Net, S., Temime-Roussel, B., Gligorovski, S., Wortham, H. 2010. Light induced multiphase chemistry of gas-phase ozone on aqueous pyruvic and oxalic acids. *Physical Chemistry Chemical Physics* 12:698-707.

Griffith, E. C., and V. Vaida. 2012. In situ observation of peptide bond formation at the water-air interface. *Proceedings of the National Academy of Sciences of the United States of America* 109(39):15697-15701. DOI: 10.1073/pnas.1210029109.

Griffith, S. M., R. F. Hansen, S. Dusanter, V. Michoud, J. B. Gilman, W. C. Kuster, P. R. Veres, M. Graus, J. A. de Gouw, J. Roberts, C. Young, R. Washenfelder, S. S. Brown, R. Thalman, E. Waxman, R. Volkamer, C. Tsai, J. Stutz, J. H. Flynn, N. Grossberg, B. Lefer, S. L. Alvarez, B. Rappenglueck, L. H. Mielke, H. D. Osthoff, and P. S. Stevens. 2016. Measurements of hydroxyl and hydroperoxy radicals during CalNex-LA: Model comparisons and radical budgets. *Journal of Geophysical Research - Atmospheres* 121(8):4211-4232. DOI: 10.1002/2015JD024358.

Gruber, N. 2011. Warming up, turning sour, losing breath: ocean biogeochemistry under global change. *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences* 369(1943):1980-1996. DOI: 10.1098/rsta.2011.0003.

Guenther, A., C. N. Hewitt, D. Erickson, R. Fall, C. Geron, T. Graedel, P. Harley, L. Klinger, M. Lerdau, W. A. Mckay, T. Pierce, B. Scholes, R. Steinbrecher, R. Tallamraju, J. Taylor, and P. Zimmerman. 1995. A Global-Model of Natural Volatile Organic-Compound Emissions. *Journal Of Geophysical Research-Atmospheres* 100(D5):8873-8892. DOI: 10.1029/94jd02950.

Guenther, A., T. Karl, P. Harley, C. Wiedinmyer, P. I. Palmer, and C. Geron. 2006. Estimates of global terrestrial isoprene emissions using MEGAN (Model of Emissions of Gases and Aerosols from Nature). *Atmospheric Chemistry and Physics* 6:3181-3210.

Guenther, A. B., X. Jiang, C. L. Heald, T. Sakulyanontvittaya, T. Duhl, L. K. Emmons, and X. Wang. 2012. The Model of Emissions of Gases and Aerosols from Nature version 2.1

(MEGAN 2.1): an extended and updated framework for modeling biogenic emissions. *Geoscientific Model Development* 5:1471-1492. DOI: doi:10.5194/gmd-5-1471-2012.

Guieu, C., S. Bonnet, T. Wagener, and M. D. Loye-Pilot. 2005. Biomass burning as a source of dissolved iron to the open ocean? *Geophysical Research Letters* 32(19). DOI: 10.1029/2005gl022962.

Guieu, C., O. Aumont, A. Paytan, L. Bopp, C. S. Law, N. Mahowald, E. P. Achterberg, E. Maranon, B. Salihoglu, A. Crise, T. Wagener, B. Herut, K. Desboeufs, M. Kanakidou, N. Olgun, F. Peters, E. Pulido-Villena, A. Tovar-Sanchez, and C. Volker. 2014. The significance of the episodic nature of atmospheric deposition to Low Nutrient Low Chlorophyll regions. *Global Biogeochemical Cycles* 28(11):1179-1198. DOI: 10.1002/2014GB004852.

Haagen-Smit, A. J. 1950. The air pollution problem in Los Angeles. *Engineering and Science* 14(3):7-13.

Haagen-Smit, A. J. 1952. Chemistry and Physiology of Los-Angeles Smog. *Industrial and Engineering Chemistry* 44(6):1342-1346. DOI: 10.1021/Ie50510a045.

Haagen-Smit, A. J., and M. M. Fox. 1954. Photochemical Ozone Formation with Hydrocarbons and Automobile Exhaust. *Air Repair* 4(3):105-136. DOI: 10.1080/00966665.1954.10467649.

Hader, D. P., H. D. Kumar, R. C. Smith, and R. C. Worrest. 2007. Effects of solar UV radiation on aquatic ecosystems and interactions with climate change. *Photochemical & Photobiological Sciences* 6(3):267-285. DOI: 10.1039/b700020k.

Hahn, K. 1967. *A factual record of correspondence between Kenneth Hahn, Los Angeles County supervisor, and the presidents of General Motors, Ford, and Chrysler, regarding the automobile industry's obligation to meet its rightful responsibility in controlling air pollution from automobiles, February 1953-January 1967*. Los Angeles: County of Los Angeles, California.

Hallquist, M., J. C. Wenger, U. Baltensperger, Y. Rudich, D. Simpson, M. Claeys, J. Dommen, N. M. Donahue, C. George, A. H. Goldstein, J. F. Hamilton, H. Herrmann, T. Hoffmann, Y. Iinuma, M. Jang, M. E. Jenkin, J. L. Jimenez, A. Kiendler-Scharr, W. Maenhaut, G. McFiggans, T. F. Mentel, A. Monod, A. S. H. Prevot, J. H. Seinfeld, J. D. Surratt, R. Szmigielski, and J. Wildt. 2009. The formation, properties and impact of secondary organic aerosol: current and emerging issues. *Atmospheric Chemistry and Physics* 9(14):5155-5236.

Hamer, P. D., D. E. Shallcross, A. Yabushita, M. Kawasaki, V. Marecal, and C. S. Boxe. 2014. Investigating the photo-oxidative and heterogeneous chemical production of HCHO in the snowpack at the South Pole, Antarctica. *Environmental Chemistry* 11(4):459-471. DOI: 10.1071/EN13227.

Hamra, G. B., N. Guha, A. Cohen, F. Laden, O. Raaschou-Nielsen, J. M. Samet, P. Vineis, F. Forastiere, P. Saldiva, T. Yorifuji, and D. Loomis. 2014. Outdoor Particulate Matter Exposure and Lung Cancer: A Systematic Review and Meta-Analysis. *Environmental Health Perspectives* 122(9):906-911. DOI: 10.1289/ehp.1408092.

Hansen, G., M. Auffhammer, and A. R. Solow. 2014. On the Attribution of a Single Event to Climate Change. *Journal of Climate* 27(22):8297-8301. DOI: 10.1175/jcli-d-14-00399.1.

Hansen, J., A. Lacis, and M. Prather. 1989. Greenhouse-Effect of Chlorofluorocarbons and Other Trace Gases. *Journal Of Geophysical Research-Atmospheres* 94(D13):16417-16421. DOI: 10.1029/Jd094id13p16417.

Hayes, P. L., A. G. Carlton, K. R. Baker, R. Ahmadov, R. A. Washenfelder, S. Alvarez, B. Rappengluck, J. B. Gilman, W. C. Kuster, J. A. de Gouw, P. Zotter, A. S. H. Prevot, S. Szidat, T. E. Kleindienst, J. H. Offenberg, P. K. Ma, and J. L. Jimenez. 2015. Modeling the formation and aging of secondary organic aerosols in Los Angeles during CalNex 2010. *Atmospheric Chemistry and Physics* 15(10):5773-5801. DOI: 10.5194/acp-15-5773-2015.

Haywood, J., and O. Boucher. 2000. Estimates of the direct and indirect radiative forcing due to tropospheric aerosols: A review. *Reviews of Geophysics* 38(4):513-543. DOI: 10.1029/1999rg000078.

Heald, C. L., and D. V. Spracklen. 2009. Atmospheric budget of primary biological aerosol particles from fungal spores. *Geophys. Res. Lett.* 36. DOI: 10.1029/2009gl037493.

Heald, C. L., and D. V. Spracklen. 2015. Land Use Change Impacts on Air Quality and Climate. *Chemical Reviews* 115(10):4476-4496. DOI: 10.1021/cr500446g.

Heald, C. L., D. J. Jacob, R. J. Park, L. M. Russell, B. J. Huebert, J. H. Seinfeld, H. Liao, and R. J. Weber. 2005. A large organic aerosol source in the free troposphere missing from current models. *Geophysical Research Letters* 32(18). DOI: 10.1029/2005gl023831.

Heald, C. L., H. Coe, J. L. Jimenez, R. J. Weber, R. Bahreini, A. M. Middlebrook, L. M. Russell, M. Jolleys, T. M. Fu, J. D. Allan, K. N. Bower, G. Capes, J. Crosier, W. T. Morgan, N. H. Robinson, P. I. Williams, M. J. Cubison, P. F. DeCarlo, and E. J. Dunlea. 2011. Exploring the vertical profile of atmospheric organic aerosol: Comparing 17 aircraft field campaigns with a global model. *Atmospheric Chemistry and Physics* 11(24):12673-12696. DOI: 10.5194/acp-11-12673-2011.

Heard, D. E., and M. J. Pilling. 2003. Measurement of OH and HO_2 in the troposphere. *Chemical Reviews* 103(12):5163-5198. DOI: 10.1021/cr020522s.

Heath, A. A., and K. T. Valsaraj. 2015. Effects of Temperature, Oxygen Level, Ionic Strength, and pH on the Reaction of Benzene with Hydroxyl Radicals at the Air-Water Interface in Comparison to the Bulk Aqueous Phase. *Journal of Physical Chemistry A* 119(31):8527-8536. DOI: 10.1021/acs.jpca.5b05152.

Hegglin, M. I., and T. G. Shepherd. 2009. Large climate-induced changes in ultraviolet index and stratosphere-to-troposphere ozone flux. *Nature geoscience* 2(10):687-691. DOI: 10.1038/NGEO604.

Hegglin, M. I., D. A. Plummer, T. G. Shepherd, J. F. Scinocca, J. Anderson, L. Froidevaux, B. Funke, D. Hurst, A. Rozanov, J. Urban, T. von Clarmann, K. A. Walker, H. J. Wang, S. Tegtmeier, and K. Weigel. 2014. Vertical structure of stratospheric water vapour trends derived from merged satellite data. *Nature geoscience* 7(10):768-776. DOI: 10.1038/NGEO2236.

Hegglin, M. I., D. W. Fahey, M. McFarland, S. A. Montzka, and E. R. Nash. 2015. Twenty Questions and Answers About the Ozone Layer: 2014 Update. In *Scientific Assessment of Ozone Depletion: 2014*, eds. Geneva, Switzerland: World Meteorological Organization.

Held, I. M., T. L. Delworth, J. Lu, K. L. Findell, and T. R. Knutson. 2005. Simulation of Sahel drought in the 20th and 21st centuries. *Proceedings of the National Academy of Sciences of the United States of America* 102(50):17891-17896. DOI: 10.1073/pnas.0509057102.

Helmig, D., S. Rossabi, J. Hueber, P. Tans, S. A. Montzka, K. Masarie, K. Thoning, C. Plass-Duelmer, A. Claude, L. J. Carpenter, A. C. Lewis, S. Punjabi, S. Reimann, M. K. Vollmer, R. Steinbrecher, J. W. Hannigan, L. K. Emmons, E. Mahieu, B. Franco, D. Smale, and A. Pozzer. 2016. Reversal of global atmospheric ethane and propane trends largely due to US oil and natural gas production. *Nature geoscience* 9:490-495. DOI: 10.1038/ngeo2721.

Hens, K., A. Novelli, M. Martinez, J. Auld, R. Axinte, B. Bohn, H. Fischer, P. Keronen, D. Kubistin, A. C. Nolscher, R. Oswald, P. Paasonen, T. Petaja, E. Regelin, R. Sander, V. Sinha, M. Sipila, D. Taraborrelli, C. T. Ernest, J. Williams, J. Lelieveld, and H. Harder. 2014. Observation and modelling of HOx radicals in a boreal forest. *Atmospheric Chemistry and Physics* 14(16):8723-8747. DOI: 10.5194/acp-14-8723-2014.

Henson, R. 2005. Reflective research: New UAVs may help solve the albedo puzzle. *UCAR Quarterly*, Summer 2005. Available at http://www.ucar.edu/communications/quarterly/summer05/reflective.html, accessed April 27, 2016.

Herbert, R. J., B. J. Murray, T. F. Whale, S. J. Dobbie, and J. D. Atkinson. 2014. Representing time-dependent freezing behaviour in immersion mode ice nucleation. *Atmospheric Chemistry and Physics* 14(16):8501-8520. DOI: 10.5194/acp-14-8501-2014.

Herrmann, H., T. Schaefer, A. Tilgner, S. A. Styler, C. Weller, M. Teich, and T. Otto. 2015. Tropospheric Aqueous-Phase Chemistry: Kinetics, Mechanisms, and Its Coupling to a Changing Gas Phase. *Chemical Reviews* 115(10):4259-4334. DOI: 10.1021/cr500447k.

Hidy, G. M., and C. L. Blanchard. 2015. Precursor reductions and ground-level ozone in the Continental United States. *Journal of the Air & Waste Management Association* 65(10):1261-1282. DOI: 10.1080/10962247.2015.1079564.

Hilboll, A., R. A., and J. P. Burrows. 2013. Long-term changes of trophspheric NO_2 over megacities derived from multiple satellite instruments. *Atmospheric Chemistry and Physics* 13(4145-4169).

Hodgson, A., T. Smith, S. Gagneux, M. Adjuik, G. Pluschke, N. K. Mensah, F. Binka, and B. Genton. 2001. Risk factors for meningococcal meningitis in northern Ghana. *Transactions of the Royal Society of Tropical Medicine and Hygiene* 95(5):477-480. DOI: 10.1016/s0035-9203(01)90007-0.

Hoegh-Guldberg, O., and J. F. Bruno. 2010. The impact of climate change on the world's marine ecosystems. *Science* 328(5985):1523-1528. DOI: 10.1126/science.1189930.

Hoffman, M. R. 1984. Reply on Comment on Acid Fog. *Environmental Science and Technology* 18(1):61-64. DOI: 10.1021/es00119a601.

Hofzumahaus, A., F. Rohrer, K. D. Lu, B. Bohn, T. Brauers, C. C. Chang, H. Fuchs, F. Holland, K. Kita, Y. Kondo, X. Li, S. R. Lou, M. Shao, L. M. Zeng, A. Wahner, and Y. H. Zhang. 2009. Amplified Trace Gas Removal in the Troposphere. *Science* 324(5935):1702-1704. DOI: 10.1126/science.1164566.

Holland, E. A., F. J. Dentener, B. H. Braswell, and J. M. Sulzman. 1999. Contemporary and pre-industrial global reactive nitrogen budgets. *Biogeochemistry* 46(1-3):7-43. DOI: 10.1023/A:1006148011944.

Holmes, C. D., D. J. Jacob, E. S. Corbitt, J. Mao, X. Yang, R. Talbot, and F. Slemr. 2010. Global atmospheric model for mercury including oxidation by bromine atoms. *Atmospheric Chemistry and Physics* 10(24):12037-12057. DOI: 10.5194/acp-10-12037-2010.

Hoose, C., J. E. Kristjansson, J. P. Chen, and A. Hazra. 2010. A Classical-Theory-Based Parameterization of Heterogeneous Ice Nucleation by Mineral Dust, Soot, and Biological Particles in a Global Climate Model. *Journal of the Atmospheric Sciences* 67(8):2483-2503. DOI: 10.1175/2010JAS3425.1.Hoose, C., and O. Möhler. 2012. Heterogeneous ice nucleation on atmospheric aerosols: a review of results from laboratory experiments. *Atmospheric Chemistry and Physics* 12(20):9817-9854. DOI: 10.5194/acp-12-9817-2012.

Huang, R. J., Y. L. Zhang, C. Bozzetti, K. F. Ho, J. J. Cao, Y. M. Han, K. R. Daellenbach, J. G. Slowik, S. M. Platt, F. Canonaco, P. Zotter, R. Wolf, S. M. Pieber, E. A. Bruns, M. Crippa, G. Ciarelli, A. Piazzalunga, M. Schwikowski, G. Abbaszade, J. Schnelle-Kreis, R. Zimmermann, Z. S. An, S. Szidat, U. Baltensperger, I. El Haddad, and A. S. H. Prevot. 2014. High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* 514(7521):218-222. DOI: 10.1038/nature13774.

Huang, S. K., Q. L. Zhang, Z. M. Qiu, and K. F. Chung. 2015. Mechanistic impact of outdoor air pollution on asthma and allergic diseases. *Journal of Thoracic Disease* 7(1):23-33. DOI: 10.3978/j.issn.2072-1439.2014.12.13.

Huffman, J. A., B. Treutlein, and U. Poschl. 2010. Fluorescent biological aerosol particle concentrations and size distributions measured with an Ultraviolet Aerodynamic Particle Sizer (UV-APS) in Central Europe. *Atmospheric Chemistry and Physics* 10(7):3215-3233.

Hwang, Y. T., D. M. W. Frierson, and S. M. Kang. 2013. Anthropogenic sulfate aerosol and the southward shift of tropical precipitation in the late 20th century. *Geophysical Research Letters* 40(11):2845-2850. DOI: 10.1002/grl.50502.

IEA (International Energy Agency). 2014. *World Energy Outlook 2014. Executive Summary*. Paris, france: International Energy Agency.

Ifa, D. R., C. P. Wu, Z. Ouyang, and R. G. Cooks. 2010. Desorption electrospray ionization and other ambient ionization methods: current progress and preview. *Analyst* 135(4):669-681. DOI: 10.1039/b925257f.

Im, U., R. Bianconi, E. Solazzo, I. Kioutsioukis, A. Badia, A. Balzarini, R. Baró, R. Bellasio, D. Brunner, C. Chemel, G. Curci, J. Flemming, R. Forkel, L. Giordano, P. Jiménez-Guerrero, M. Hirtl, A. Hodzic, L. Honzak, O. Jorba, C. Knote, J. J. P. Kuenen, P. A. Makar, A. Manders-Groot, L. Neal, J. L. Pérez, G. Pirovano, G. Pouliot, R. S. Jose, N. Savage, W. Schroder, R. S. Sokhi, D. Syrakov, A. Torian, P. Tuccella, J. Werhahn, R. Wolke, K. Yahya, R. Zabkar, Y. Zhang, J. Zhang, C. Hogrefe, and S. Galmarini. 2015. Evaluation of operational on-line-coupled regional air quality models over Europe and North America in the context of AQMEII phase 2. Part I: Ozone. *Atmospheric Environment* 115:404-420. DOI: 10.1016/j.atmosenv.2014.09.042.

IPCC (Intergovernmental Panel on Climate Change). 2013. *Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. T. F. Stocker, D. Qin, G.-K. Plattner, M. Tignor, S. K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P. M. Midgley, eds*. Cambridge, United Kingdom and New York: Cambridge University Press.

IPCC. 2014. *Climate Change 2014: Synthesis Report. Contribution of Working Groups I, II and III to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change. Core Writing Team, R. K. Pachauri and L. A. Meyer, eds.* Geneva, Switzerland: IPCC.

Isaacman, G., N. M. Kreisberg, L. D. Yee, D. R. Worton, A. W. H. Chan, J. A. Moss, S. V. Hering, and A. H. Goldstein. 2014. Online derivatization for hourly measurements of gas- and particle-phase semi-volatile oxygenated organic compounds by thermal desorption aerosol gas chromatography (SV-TAG). *Atmospheric Measurement Techniques* 7(12):4417-4429. DOI: 10.5194/amt-7-4417-2014.

Isaksen, I. S. A., T. K. Berntsen, S. B. Dalsoren, K. Eleftheratos, Y. Orsolini, B. Rognerud, F. Stordal, O. A. Sovde, C. Zerefos, and C. D. Holmes. 2014. Atmospheric Ozone and Methane in a Changing Climate. *Atmosphere* 5(3):518-535. DOI: 10.3390/atmos5030518.

Ito, T., A. Nenes, M. S. Johnson, N. Meskhidze, and C. Deutsch. 2016. Acceleration of oxygen decline in the tropical Pacific over the past decades by aerosol pollutants. *Nature geoscience* 9(6):443-447. DOI: 10.1038/NGEO2717.

IUPAC (International Union of Pure and Applied Chemistry). 2015. Evaluated Kinetic Data. http://iupac.pole-ether.fr/. Available at http://iupac.pole-ether.fr/, accessed.

Jacob, D. J. 2000. *Introduction to Atmospheric Chemistry*. Princeton, NJ: Princeton University Press.

Jacob, D. J., J. M. Waldman, J. W. Munger, and M. R. Hoffmann. 1985. Chemical-Composition of Fogwater Collected Along the California Coast. *Environmental Science & Technology* 19(8):730-736. DOI: 10.1021/Es00138a013.

Jacob, D. J., M. J. Prather, S. C. Wofsy, and M. B. McElroy. 1987. Atmospheric distribution of ^{85}Kr simulated with a general circulation model. *Journal of Geophysical Research - Atmospheres* 92(D6):6614–6626. DOI: 10.1029/JD092iD06p06614.

Jacob, D. J., M. J. Prather, P. J. Rasch, R. L. Shia, Y. J. Balkanski, S. R. Beagley, D. J. Bergmann, W. T. Blackshear, M. Brown, M. Chiba, M. P. Chipperfield, J. deGrandpre, J. E. Dignon, J. Feichter, C. Genthon, W. L. Grose, P. S. Kasibhatla, I. Kohler, M. A. Kritz, K. Law, J. E. Penner, M. Ramonet, C. E. Reeves, D. A. Rotman, D. Z. Stockwell, P. F. J. VanVelthoven, G. Verver, O. Wild, H. Yang, and P. Zimmermann. 1997. Evaluation and intercomparison of global atmospheric transport models using Rn-222 and other short-lived tracers. *Journal Of Geophysical Research-Atmospheres* 102(D5):5953-5970. DOI: 10.1029/96jd02955.

Jacobson, M. Z., and D. G. Streets. 2009. Influence of future anthropogenic emissions on climate, natural emissions, and air quality. *Journal Of Geophysical Research-Atmospheres* 114:21. DOI: doi:10.1029/2008jd011476.

Jaegle, L., R. V. Martin, K. Chance, L. Steinberger, T. P. Kurosu, D. J. Jacob, A. I. Modi, V. Yoboue, L. Sigha-Nkamdjou, and C. Galy-Lacaux. 2004. Satellite mapping of rain-induced nitric oxide emissions from soils. *Journal Of Geophysical Research-Atmospheres* 109(D21):14. DOI: D21310, 10.1029/2004jd004787.

Jayne, J. T., D. C. Leard, X. F. Zhang, P. Davidovits, K. A. Smith, C. E. Kolb, and D. R. Worsnop. 2000. Development of an aerosol mass spectrometer for size and composition analysis of submicron particles. *Aerosol Science and Technology* 33(1-2):49-70. DOI: 10.1080/027868200410840.

Jensen, E. J., L. Pfister, T. P. Bui, P. Lawson, and D. Baumgardner. 2010. Ice nucleation and cloud microphysical properties in tropical tropopause layer cirrus. *Atmospheric Chemistry and Physics* 10(3):1369-1384. DOI: 10.5194/acp-10-1369-2010.

Jerrett, M., R. T. Burnett, P. Kanaroglou, J. Eyles, N. Finkelstein, C. Giovis, and J. R. Brook. 2001. A GIS - environmental justice analysis of particulate air pollution in Hamilton, Canada. *Environment and Planning A* 33(6):955-973. DOI: 10.1068/A33137.

Jerrett, M., R. T. Burnett, C. A. Pope, K. Ito, G. Thurston, D. Krewski, Y. L. Shi, E. Calle, and M. Thun. 2009a. Long-Term Ozone Exposure and Mortality. *New England Journal of Medicine* 360(11):1085-1095. DOI: 10.1056/NEJMoa0803894.

Jerrett, M., R. T. Burnett, C. A. Pope, II, K. Ito, G. Thurston, D. Krewski, Y. Shi, E. Calle, and M. Thun. 2009b. Long-Term Ozone Exposure and Mortality. *New England Journal of Medicine* 360(11):1085-1095. DOI: 10.1056/NEJMoa0803894.

Jickells, T. D., Z. S. An, K. K. Andersen, A. R. Baker, G. Bergametti, N. Brooks, J. J. Cao, P. W. Boyd, R. A. Duce, K. A. Hunter, H. Kawahata, N. Kubilay, J. laRoche, P. S. Liss, N. Mahowald, J. M. Prospero, A. J. Ridgwell, I. Tegen, and R. Torres. 2005. Global iron connections between desert dust, ocean biogeochemistry, and climate. *Science* 308(5718):67-71. DOI: 10.1126/science.1105959.

Jimenez, J. L., M. R. Canagaratna, N. M. Donahue, A. S. H. Prevot, Q. Zhang, J. H. Kroll, P. F. DeCarlo, J. D. Allan, H. Coe, N. L. Ng, A. C. Aiken, K. S. Docherty, I. M. Ulbrich, A. P. Grieshop, A. L. Robinson, J. Duplissy, J. D. Smith, K. R. Wilson, V. A. Lanz, C. Hueglin, Y. L. Sun, J. Tian, A. Laaksonen, T. Raatikainen, J. Rautiainen, P. Vaattovaara, M. Ehn, M. Kulmala, J. M. Tomlinson, D. R. Collins, M. J. Cubison, E. J. Dunlea, J. A. Huffman, T. B. Onasch, M. R. Alfarra, P. I. Williams, K. Bower, Y. Kondo, J. Schneider, F. Drewnick, S. Borrmann, S. Weimer, K. Demerjian, D. Salcedo, L. Cottrell, R. Griffin, A. Takami, T. Miyoshi, S. Hatakeyama, A. Shimono, J. Y. Sun, Y. M. Zhang, K. Dzepina, J. R. Kimmel, D. Sueper, J. T. Jayne, S. C. Herndon, A. M. Trimborn, L. R. Williams, E. C. Wood, A. M. Middlebrook, C. E. Kolb, U. Baltensperger, and D. R. Worsnop. 2009. Evolution of organic aerosols in the atmosphere. *Science* 326(5959):1525-1529. DOI: 10.1126/science.1180353.

Johanson, C. M., and Q. Fu. 2009. Hadley Cell Widening: Model Simulations versus Observations. *Journal of Climate* 22(10):2713-2725. DOI: 10.1175/2008JCLI2620.1.

Johnson, D., S. R. Utembe, M. E. Jenkin, R. G. Derwent, G. D. Hayman, M. R. Alfarra, H. Coe, and G. McFiggans. 2006. Simulating regional scale secondary organic aerosol formation during the TORCH 2003 campaign in the southern UK. *Atmospheric Chemistry and Physics* 6:403-418.

Johnston, H. 1971. Reduction of Stratospheric Ozone by Nitrogen Oxide Catalysts from Supersonic Transport Exhaust. *Science* 173(3996):517-&. DOI: 10.1126/science.173.3996.517.

Jones, L., A. Provins, M. Holland, G. Mills, F. Hayes, J. H. B. Emmetta, L. Sheppard, R. Smith, M. Sutton, K. Hicks, M. Ashmore, R. Haines-Young, and L. Harper-Simmonds. 2014. A review and application of the evidence for nitrogen impacts on ecosystem services. *Ecosystem Services* 7:76-88. DOI: 10.1016/j.ecoser.2013.09.001.

Journet, E., K. V. Desboeufs, S. Caquineau, and J. L. Colin. 2008. Mineralogy as a critical factor of dust iron solubility. *Geophysical Research Letters* 35(7). DOI: 10.1029/2007gl031589.

Kameel, F. R., F. Riboni, M. R. Hoffmann, S. Enami, and A. J. Colussi. 2014. Fenton Oxidation of Gaseous Isoprene on Aqueous Surfaces. *Journal of Physical Chemistry C* 118(50):29151-29158. DOI: 10.1021/jp505010e.

Kamphus, M., M. Ettner-Mahl, T. Klimach, F. Drewnick, L. Keller, D. J. Cziczo, S. Mertes, S. Borrmann, and J. Curtius. 2010. Chemical composition of ambient aerosol, ice residues and cloud droplet residues in mixed-phase clouds: single particle analysis during the Cloud and Aerosol Characterization Experiment (CLACE 6). *Atmospheric Chemistry and Physics* 10(16):8077-8095. DOI: 10.5194/acp-10-8077-2010.

Kanakidou, M., R. A. Duce, J. M. Prospero, A. R. Baker, C. Benitez-Nelson, F. J. Dentener, K. A. Hunter, P. S. Liss, N. Mahowald, G. S. Okin, M. Sarin, K. Tsigaridis, M. Uematsu, L.

M. Zamora, and T. Zhu. 2012. Atmospheric fluxes of organic N and P to the global ocean. *Global Biogeochemical Cycles* 26. DOI: 10.1029/2011gb004277.

Kanaya, Y., R. Q. Cao, H. Akimoto, M. Fukuda, Y. Komazaki, Y. Yokouchi, M. Koike, H. Tanimoto, N. Takegawa, and Y. Kondo. 2007. Urban photochemistry in central Tokyo: 1. Observed and modeled OH and HO_2 radical concentrations during the winter and summer of 2004. *Journal Of Geophysical Research-Atmospheres* 112(D21). DOI: 10.1029/2007jd008670.

Kanaya, Y., M. Fukuda, H. Akimoto, N. Takegawa, Y. Komazaki, Y. Yokouchi, M. Koike, and Y. Kondo. 2008. Urban photochemistry in central Tokyo: 2. Rates and regimes of oxidant (O-3+NO2) production. *Journal Of Geophysical Research-Atmospheres* 113(D6). DOI: 10.1029/2007jd008671.

Karl, T., A. Guenther, C. Lindinger, A. Jordan, R. Fall, and W. Lindinger. 2001. Eddy covariance measurements of oxygenated volatile organic compound fluxes from crop harvesting using a redesigned proton-transfer-reaction mass spectrometer. *Journal Of Geophysical Research-Atmospheres* 106(D20):24157-24167. DOI: 10.1029/2000jd000112.

Karl, T., E. Apel, A. Hodzic, D. D. Riemer, D. R. Blake, and C. Wiedinmyer. 2009. Emissions of volatile organic compounds inferred from airborne flux measurements over a megacity. *Atmospheric Chemistry and Physics* 9(1):271-285.

Karl, T., P. Harley, L. Emmons, B. Thornton, A. Guenther, C. Basu, A. Turnipseed, and K. Jardine. 2010. Efficient Atmospheric Cleansing of Oxidized Organic Trace Gases by Vegetation. *Science* 330(6005):816-819. DOI: 10.1126/science.1192534.

Karl, T., P. K. Misztal, H. H. Jonsson, S. Shertz, A. H. Goldstein, and A. B. Guenther. 2013. Airborne Flux Measurements of BVOCs above Californian Oak Forests: Experimental Investigation of Surface and Entrainment Fluxes, OH Densities, and Damkohler Numbers. *Journal of the Atmospheric Sciences* 70(10):3277-3287. DOI: 10.1175/jas-d-13-054.1.

Kasibhatla, P., M. Heimann, P. Rayner, N. Mahowald, R. G. Prinn, and D. E. Hartley, eds. 2013. *Inverse Methods in Global Biogeochemical Cycles*. Washington, DC: American Geophysical Union.

Kaufman, Y. J., D. Tanre, L. A. Remer, E. F. Vermote, A. Chu, and B. N. Holben. 1997. Operational remote sensing of tropospheric aerosol over land from EOS moderate resolution imaging spectroradiometer. *Journal Of Geophysical Research-Atmospheres* 102(D14):17051-17067. DOI: 10.1029/96jd03988.

Kaufman, Y. J., I. Koren, L. A. Remer, D. Tanre, P. Ginoux, and S. Fan. 2005. Dust transport and deposition observed from the Terra-Moderate Resolution Imaging Spectroradiometer (MODIS) spacecraft over the Atlantic ocean. *Journal Of Geophysical Research-Atmospheres* 110(D10). DOI: 10.1029/2003JD004436.

Kellogg, C. A., and D. W. Griffin. 2006. Aerobiology and the global transport of desert dust. *Trends in Ecology & Evolution* 21(11):638-644. DOI: 10.1016/j.tree.2006.07.004.

Kerl, P. Y., W. X. Zhang, J. B. Moreno-Cruz, A. Nenes, M. J. Realff, A. G. Russell, J. Sokol, and V. M. Thomas. 2015. New approach for optimal electricity planning and dispatching with hourly time-scale air quality and health considerations. *Proceedings of the National Academy of Sciences of the United States of America* 112(35):10884-10889. DOI: 10.1073/pnas.1413143112.

Khvorostyanov, V. I., and J. A. Curry. 2005. The theory of ice nucleation by heterogeneous freezing of deliquescent mixed CCN. Part II: Parcel model simulation. *Journal of the Atmospheric Sciences* 62(2):261-285. DOI: 10.1175/Jas-3367.1.

Kidd, C., V. Perraud, L. Wingen, and B. J. Finlayson-Pitts. 2014. Integrating Phase and Composition of Secondary Organic Aerosol from the Ozonolysis of α-Pinene *PNAS* revised version under review.

Kim, M. J., D. K. Farmer, and T. H. Bertram. 2014. A controlling role for the air-sea interface in the chemical processing of reactive nitrogen in the coastal marine boundary layer. *Proceedings of the National Academy of Sciences of the United States of America* 111(11):3943-3948. DOI: 10.1073/pnas.1318694111.

Kirkby, J., J. Duplissy, K. Sengupta, C. Frege, H. Gordon, C. Williamson, M. Heinritzi, M. Simon, C. Yan, J. Almeida, J. Trostl, T. Nieminen, I. K. Ortega, R. Wagner, A. Adamov, A. Amorim, A. K. Bernhammer, F. Bianchi, M. Breitenlechner, S. Brilke, X. M. Chen, J. Craven, A. Dias, S. Ehrhart, R. C. Flagan, A. Franchin, C. Fuchs, R. Guida, J. Hakala, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kangasluoma, J. Kim, M. Krapf, A. Kurten, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, U. Molteni, A. Onnela, O. Perakyla, F. Piel, T. Petaja, A. P. Praplan, K. Pringle, A. Rap, N. A. D. Richards, I. Riipinen, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, C. E. Scott, J. H. Seinfeld, M. Sipila, G. Steiner, Y. Stozhkov, F. Stratmann, A. Tome, A. Virtanen, A. L. Vogel, A. C. Wagner, P. E. Wagner, E. Weingartner, D. Wimmer, P. M. Winkler, P. L. Ye, X. Zhang, A. Hansel, J. Dommen, N. M. Donahue, D. R. Worsnop, U. Baltensperger, M. Kulmala, K. S. Carslaw, and J. Curtius. 2016. Ion-induced nucleation of pure biogenic particles. *Nature* 533(7604):521-526. DOI: 10.1038/nature17953.

Kloster, S., N. M. Mahowald, J. T. Randerson, and P. J. Lawrence. 2012. The impacts of climate, land use, and demography on fires during the 21st century simulated by CLM-CN. *Biogeosciences* 9(1):509-525. DOI: 10.5194/bg-9-509-2012.

Knapton, S. 2015. Volkswagen scandal: nearly 12,000 deaths could be avoided if industry met emissions targets. *The Telegraph*, September 22, 2015. Available at http://www.telegraph.co.uk/news/health/news/11883416/vw-scandal-emission-target-death-rate.html, accessed April 15, 2016.

Knipping, E. M., M. J. Lakin, K. L. Foster, P. Jungwirth, D. J. Tobias, R. B. Gerber, D. Dabdub, and B. J. Finlayson-Pitts. 2000. Experiments and simulations of ion-enhanced interfacial chemistry on aqueous NaCl aerosols. *Science* 288:301-306. DOI: 10.1126/science.288.5464.301.

Knorr, W., L. Jiang, and A. Arneth. 2016. Climate, CO_2, and human population impacts on global wildfire emissions. *Biogeosciences* 13(1):267-282. DOI: 10.5194/bg-13-267-2016.

Kolb, C. E., R. A. Cox, J. P. D. Abbatt, M. Ammann, E. J. Davis, D. J. Donaldson, B. C. Garrett, C. George, P. T. Griffiths, D. R. Hanson, M. Kulmala, G. McFiggans, U. Poschl, I. Riipinen, M. J. Rossi, Y. Rudich, P. E. Wagner, P. M. Winkler, D. R. Worsnop, and C. D. O' Dowd. 2010. An overview of current issues in the uptake of atmospheric trace gases by aerosols and clouds. *Atmospheric Chemistry and Physics* 10(21):10561-10605. DOI: 10.5194/acp-10-10561-2010.

Kolusu, S. R., J. H. Marsham, J. Mulcahy, B. Johnson, C. Dunning, M. Bush, and D. V. Spracklen. 2015. Impacts of Amazonia biomass burning aerosols assessed from short-range weather forecasts. *Atmospheric Chemistry and Physics* 15(21):12251-12266. DOI: 10.5194/acp-15-12251-2015.

Kong, X., R. Forkel, R. S. Sokhi, P. Suppan, A. Baklanov, M. Gauss, D. Brunner, R. Baro, A. Balzarini, C. Chemel, G. Curci, P. Jimenez-Guerrero, M. Hirtl, L. Honzak, U. Im, J. L. Perez, G. Pirovano, R. San Jose, K. H. Schlunzen, G. Tsegas, P. Tuccella, J. Werhahn, R. Zabkar, and S. Galmarini. 2015. Analysis of meteorology-chemistry interactions during air pollution episodes using online coupled models within AQMEII phase-2. *Atmospheric Environment* 115:527-540. DOI: 10.1016/j.atmosenv.2014.09.020.

Konovalov, I. B., M. Beekmann, J. P. Burrows, and A. Richter. 2008. Satellite measurement based estimates of decadal changes in European nitrogen oxides emissions. *Atmospheric Chemistry and Physics* 8(10):2623-2641.

Koren, I., O. Altaratz, L. A. Remer, G. Feingold, J. V. Martins, and R. H. Heiblum. 2012. Aerosol-induced intensification of rain from the tropics to the mid-latitudes. *Nature geoscience* 5(2):118-122. DOI: 10.1038/NGEO1364.

Kovacs, T. A., and W. H. Brune. 2001. Total OH loss rate measurement. *Journal of Atmospheric Chemistry* 39(2):105-122. DOI: 10.1023/A:1010614113786.

Krechmer, J. E., M. M. Coggon, P. Massoli, T. B. Nguyen, J. D. Crounse, W. W. Hu, D. A. Day, G. S. Tyndall, D. K. Henze, J. C. Rivera-Rios, J. B. Nowak, J. R. Kimmel, R. L. Mauldin, H. Stark, J. T. Jayne, M. Sipila, H. Junninen, J. M. S. Clair, X. Zhang, P. A. Feiner, L. Zhang, D. O. Miller, W. H. Brune, F. N. Keutsch, P. O. Wennberg, J. H. Seinfeld, D. R. Worsnop, J. L. Jimenez, and M. R. Canagaratna. 2015. Formation of Low Volatility Organic Compounds and Secondary Organic Aerosol from Isoprene Hydroxyhydroperoxide Low-NO Oxidation. *Environmental Science & Technology* 49(17):10330-10339. DOI: 10.1021/acs.est.5b02031.

Krey, V., G. Luderer, L. Clarke, and E. Kriegler. 2014. Getting from here to there: Energy technology transformation pathways in the EMF27 scenarios. *Climatic Change* 123(3-4):369-382. DOI: 10.1007/s10584-013-0947-5.

Krishnamurthy, A., J. K. Moore, C. S. Zender, and C. Luo. 2007. Effects of atmospheric inorganic nitrogen deposition on ocean biogeochemistry. *Journal of Geophysical Research-Biogeosciences* 112(G2). DOI: 10.1029/2006jg000334.

Krueger, A. J., M. R. Schoeberl, and R. S. Stolarski. 1987. TOMS Observations of Total Ozone in the 1986 Antarctic Spring. *Geophysical Research Letters* 14(5):527-530. DOI: 10.1029/Gl014i005p00527.

Kuang, C., P. H. McMurry, and A. V. McCormick. 2009. Determination of cloud condensation nuclei production from measured new particle formation events. *Geophysical Research Letters* 36. DOI: 10.1029/2009gl037584.

Kuhlmann, J., and J. Quaas. 2010. How can aerosols affect the Asian summer monsoon? Assessment during three consecutive pre-monsoon seasons from CALIPSO satellite data. *Atmospheric Chemistry and Physics* 10(10):4673-4688. DOI: 10.5194/acp-10-4673-2010.

Kulmala, M., J. Kontkanen, H. Junninen, K. Lehtipalo, H. E. Manninen, T. Nieminen, T. Petaja, M. Sipila, S. Schobesberger, P. Rantala, A. Franchin, T. Jokinen, E. Jarvinen, M. Aijala, J. Kangasluoma, J. Hakala, P. P. Aalto, P. Paasonen, J. Mikkila, J. Vanhanen, J. Aalto, H. Hakola, U. Makkonen, T. Ruuskanen, R. L. Mauldin, J. Duplissy, H. Vehkamaki, J. Back, A. Kortelainen, I. Riipinen, T. Kurten, M. V. Johnston, J. N. Smith, M. Ehn, T. F. Mentel, K. E. J. Lehtinen, A. Laaksonen, V. M. Kerminen, and D. R. Worsnop. 2013. Direct Observations of Atmospheric Aerosol Nucleation. *Science* 339(6122):943-946. DOI: 10.1126/science.1227385.

Kumar, A., M. M. Sarin, and B. Srinivas. 2010. Aerosol iron solubility over Bay of Bengal: Role of anthropogenic sources and chemical processing. *Marine Chemistry* 121(1-4):167-175. DOI: 10.1016/j.marchem.2010.04.005.

Kupiainen-Maatta, O., T. Olenius, H. Korhonen, J. Malila, M. Dal Maso, K. Lehtinen, and H. Vehkamaki. 2014. Critical cluster size cannot in practice be determined by slope analysis in atmospherically relevant applications. *Journal of Aerosol Science* 77:127-144. DOI: 10.1016/j.jaerosci.2014.07.005.

Lamarque, J. F., F. Dentener, J. McConnell, C. U. Ro, M. Shaw, R. Vet, D. Bergmann, P. Cameron-Smith, S. Dalsoren, R. Doherty, G. Faluvegi, S. J. Ghan, B. Josse, Y. H. Lee, I. A. MacKenzie, D. Plummer, D. T. Shindell, R. B. Skeie, D. S. Stevenson, S. Strode, G. Zeng, M. Curran, D. Dahl-Jensen, S. Das, D. Fritzsche, and M. Nolan. 2013. Multi-model mean nitrogen and sulfur deposition from the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): evaluation of historical and projected future changes. *Atmospheric Chemistry and Physics* 13(16):7997-8018. DOI: 10.5194/acp-13-7997-2013.

Lammel, G., A. M. Sehili, T. C. Bond, J. Feichter, and H. Grassl. 2009. Gas/particle partitioning and global distribution of polycyclic aromatic hydrocarbons - A modelling approach. *Chemosphere* 76(1):98-106. DOI: 10.1016/j.chemosphere.2009.02.017.

Lance, S., M. Shupe, G. Feingold, C. Brock, J. Cozic, J. Holloway, R. H. Moore, A. Nenes, J. Schwarz, R. Spackman, K. D. Froyd, D. M. Murphy, J. Brioude, O. Cooper, A. Stohl, and J. F. Burkhart. 2011. Cloud condensation nuclei as a modulator for ice processes in Arctic mixed-phase clouds. *Atmospheric Chemistry and Physics* 11:8003-8015. DOI: 10.5194/acp-11-8003-2011.

Landis, M. S., and G. J. Keeler. 2002. Atmospheric mercury deposition to Lake Michigan during the Lake Michigan Mass Balance Study. *Environmental Science & Technology* 36(21):4518-4524. DOI: 10.1021/es011217b.

Landis, M. S., R. K. Stevens, F. Schaedlich, and E. M. Prestbo. 2002. Development and characterization of an annular denuder methodology for the measurement of divalent inorganic reactive gaseous mercury in ambient air. *Environmental Science & Technology* 36(13):3000-3009. DOI: 10.1021/es015887t.

Landreman, A. P., M. M. Shafer, J. C. Hemming, M. P. Hannigan, and J. J. Schauer. 2008. A macrophage-based method for the assessment of the reactive oxygen species (ROS) activity of atmospheric particulate matter (PM) and application to routine (daily-24 h) aerosol monitoring studies. *Aerosol Science and Technology* 42(11):946-957. DOI: 10.1080/02786820802363819.

Laskin, A., J. Laskin, and S. A. Nizkorodov. 2015. Chemistry of Atmospheric Brown Carbon. *Chemical Reviews* 115(10):4335-4382. DOI: 10.1021/cr5006167.

Lau, K. M., and K. M. Kim. 2006. Observational relationships between aerosol and Asian monsoon rainfall, and circulation. *Geophysical Research Letters* 33(21). DOI: 10.1029/2006gl027546.

Lau, K. M., M. K. Kim, and K. M. Kim. 2006. Asian summer monsoon anomalies induced by aerosol direct forcing: the role of the Tibetan Plateau. *Climate Dynamics* 26(7-8):855-864. DOI: 10.1007/s00382-006-0114-z.

Lebo, Z. J., and H. Morrison. 2014. Dynamical Effects of Aerosol Perturbations on Simulated Idealized Squall Lines. *Monthly Weather Review* 142(3):991-1009. DOI: 10.1175/mwr-d-13-00156.1.

Lebo, Z. J., and J. H. Seinfeld. 2011. Theoretical basis for convective invigoration due to increased aerosol concentration. *Atmospheric Chemistry and Physics* 11:5407-5429. DOI: 10.5194/acp-11-5407-2011.

Lee, D., Y. C. Sud, L. Oreopoulos, K. M. Kim, W. K. Lau, and I. S. Kang. 2014. Modeling the influences of aerosols on pre-monsoon circulation and rainfall over Southeast Asia. *Atmospheric Chemistry and Physics* 14(13):6853-6866. DOI: 10.5194/acp-14-6853-2014.

Lee, J. J., G. E. Neely, S. C. Perrigan, and L. C. Grothaus. 1981. Effect of simulated sulfuric-acid rain on yield, growth and foliar injury of several crops. *Environmental and Experimental Botany* 21(2):171-185. DOI: 10.1016/0098-8472(81)90024-1.

Lee, W. S., and M. K. Kim. 2010. Effects of radiative forcing by black carbon aerosol on spring rainfall decrease over Southeast Asia. *Atmospheric Environment* 44(31):3739-3744. DOI: 10.1016/j.atmosenv.2010.06.058.

Leibensperger, E. M., L. J. Mickley, D. J. Jacob, W. T. Chen, J. H. Seinfeld, A. Nenes, P. J. Adams, D. G. Streets, N. Kumar, and D. Rind. 2012. Climatic effects of 1950-2050 changes in US anthropogenic aerosols - Part 2: Climate response. *Atmospheric Chemistry and Physics* 12(7):3349-3362. DOI: 10.5194/acp-12-3349-2012.

Leighton, P. A. 1961. *Photochemistry of Air Pollution*. New York: Academic Press.

Lelieveld, J., T. M. Butler, J. N. Crowley, T. J. Dillon, H. Fischer, L. Ganzeveld, H. Harder, M. G. Lawrence, M. Martinez, D. Taraborrelli, and J. Williams. 2008. Atmospheric oxidation capacity sustained by a tropical forest. *Nature* 452(7188):737-740. DOI: 10.1038/nature06870.

Lelieveld, J., J. S. Evans, M. Fnais, D. Giannadaki, and A. Pozzer. 2015. The contribution of outdoor air pollution sources to premature mortality on a global scale. *Nature* 525(7569):367-+. DOI: 10.1038/nature15371.

Lenton, T. M. 2011. Early warning of climate tipping points. *Nature Climate Change* 1(4):201-209. DOI: 10.1038/Nclimate1143.

Levin, Z., and W. R. Cotton. 2009. *Aerosol pollution impact on precipitation : a scientific review*. Dordrecht ; London: Springer.

Liang, Q., L. Jaegle, D. A. Jaffe, P. Weiss-Penzias, A. Heckman, and J. A. Snow. 2004. Long-range transport of Asian pollution to the northeast Pacific: Seasonal variations and transport pathways of carbon monoxide. *Journal Of Geophysical Research-Atmospheres* 109(D23). DOI: 10.1029/2003jd004402.

Lin, M. Y., A. M. Fiore, O. R. Cooper, L. W. Horowitz, A. O. Langford, H. Levy, B. J. Johnson, V. Naik, S. J. Oltmans, and C. J. Senff. 2012. Springtime high surface ozone events over the western United States: Quantifying the role of stratospheric intrusions. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2012jd018151.

Lipfert, F. W., R. E. Wyzga, J. D. Baty, and J. P. Miller. 2006. Traffic density as a surrogate measure of environmental exposures in studies of air pollution health effects: Long-term mortality in a cohort of US veterans. *Atmospheric Environment* 40(1):154-169. DOI: 10.1016/j.atmosenv.2005.09.027.

Liu, X., X. Shi, K. Zhang, E. J. Jensen, A. Gettelman, D. Barahona, A. Nenes, and P. Lawson. 2012. Sensitivity studies of dust ice nuclei effect on cirrus clouds with the Community Atmosphere Model CAM5. *Atmospheric Chemistry and Physics* 12(24):12061-12079. DOI: 10.5194/acp-12-12061-2012.

Lohmann, U., and K. Diehl. 2006. Sensitivity studies of the importance of dust ice nuclei for the indirect aerosol effect on stratiform mixed-phase clouds. *Journal of the Atmospheric Sciences* 63(3):968-982. DOI: 10.1175/Jas3662.1.

Lohmann, U., and J. Feichter. 2005. Global indirect aerosol effects: a review. *Atmospheric Chemistry and Physics* 5:715-737.

Longo, A. F., Y. Feng, B. Lai, W. M. Landing, R. U. Shelley, A. Nenes, N. Mihalopoulos, K. Violaki, and E. D. Ingall. 2016. Influence of Atmospheric Processes on the Solubility and Composition of Iron in Saharan Dust. *Environmental Science and Technology* 50(13):6912-6920. DOI: 10.1021/acs.est.6b02605.

Lovelock, J. E. 1971. Atmospheric Fluorine Compounds as Indicators of Air Movements. *Nature* 230(5293):379. DOI: 10.1038/230379a0.

Lovelock, J. E., R. J. Maggs, and R. J. Wade. 1973. Halogenated hydrocarbons in and over the Atlantic. *Nature* 241:194-196. DOI: 10.1038/241194a0.

Lu, J., G. A. Vecchi, and T. Reichler. 2007. Expansion of the Hadley cell under global warming. *Geophysical Research Letters* 34(6). DOI: 10.1029/2006gl028443.

Maher, B. A., J. M. Prospero, D. Mackie, D. Gaiero, P. P. Hesse, and Y. Balkanski. 2010. Global connections between aeolian dust, climate and ocean biogeochemistry at the present day and at the last glacial maximum. *Earth-Science Reviews* 99(1-2):61-97. DOI: 10.1016/j.earscirev.2009.12.001.

Mahlman, J. D., H. Levy, and W. J. Moxim. 1980. 3-Dimensional Tracer Structure and Behavior as Simulated in 2 Ozone Precursor Experiments. *Journal of the Atmospheric Sciences* 37(3):655-685. DOI: 10.1175/1520-0469(1980)037<0655:Tdtsab>2.0.Co;2.

Mahowald, N. M. 2007. Anthropocene changes in desert area: Sensitivity to climate model predictions. *Geophysical Research Letters* 34(18):5. DOI: 10.1029/2007gl030472.

Mahowald, N. 2011. Aerosol Indirect Effect on Biogeochemical Cycles and Climate. *Science* 334(6057):794-796. DOI: 10.1126/science.1207374.

Mahowald, N. M., A. R. Baker, G. Bergametti, N. Brooks, R. A. Duce, T. D. Jickells, N. Kubilay, J. M. Prospero, and I. Tegen. 2005. Atmospheric global dust cycle and iron inputs to the ocean. *Global Biogeochemical Cycles* 19(4). DOI: 10.1029/2004gb002402.

Mahowald, N., T. D. Jickells, A. R. Baker, P. Artaxo, C. R. Benitez-Nelson, G. Bergametti, T. C. Bond, Y. Chen, D. D. Cohen, B. Herut, N. Kubilay, R. Losno, C. Luo, W. Maenhaut, K. A. McGee, G. S. Okin, R. L. Siefert, and S. Tsukuda. 2008a. Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Global Biogeochemical Cycles* 22(4):19. DOI: Gb4026, 10.1029/2008gb003240.

Mahowald, N., T. D. Jickells, A. R. Baker, P. Artaxo, C. R. Benitez-Nelson, G. Bergametti, T. C. Bond, Y. Chen, D. D. Cohen, B. Herut, N. Kubilay, R. Losno, C. Luo, W. Maenhaut, K. A. McGee, G. S. Okin, R. L. Siefert, and S. Tsukuda. 2008b. Global distribution of atmospheric phosphorus sources, concentrations and deposition rates, and anthropogenic impacts. *Global Biogeochemical Cycles* 22(4). DOI: 10.1029/2008gb003240.

Mahowald, N. M., S. Kloster, S. Engelstaedter, J. K. Moore, S. Mukhopadhyay, J. R. McConnell, S. Albani, S. C. Doney, A. Bhattacharya, M. A. J. Curran, M. G. Flanner, F. M. Hoffman, D. M. Lawrence, K. Lindsay, P. A. Mayewski, J. Neff, D. Rothenberg, E. Thomas, P. E. Thornton, and C. S. Zender. 2010. Observed 20th century desert dust variability: impact on climate and biogeochemistry. *Atmospheric Chemistry and Physics* 10(22):10875-10893. DOI: 10.5194/acp-10-10875-2010.

Mao, J. Q., X. R. Ren, S. A. Chen, W. H. Brune, Z. Chen, M. Martinez, H. Harder, B. Lefer, B. Rappengluck, J. Flynn, and M. Leuchner. 2010. Atmospheric oxidation capacity in the summer of Houston 2006: Comparison with summer measurements in other metropolitan studies. *Atmospheric Environment* 44(33):4107-4115. DOI: 10.1016/j.atmosenv.2009.01.013.

Mao, J., X. Ren, L. Zhang, D. M. Van Duin, R. C. Cohen, J. H. Park, A. H. Goldstein, F. Paulot, M. R. Beaver, J. D. Crounse, P. O. Wennberg, J. P. DiGangi, S. B. Henry, F. N. Keutsch, C. Park, G. W. Schade, G. M. Wolfe, J. A. Thornton, and W. H. Brune. 2012. Insights into hydroxyl measurements and atmospheric oxidation in a California forest. *Atmospheric Chemistry and Physics* 12(17):8009-8020. DOI: 10.5194/acp-12-8009-2012.

Marandino, C. A., W. J. De Bruyn, S. D. Miller, and E. S. Saltzman. 2007. Eddy correlation measurements of the air/sea flux of dimethylsulfide over the North Pacific Ocean. *Journal Of Geophysical Research-Atmospheres* 112(D3). DOI: 10.1029/2006jd007293.

Martin, J. H. 1990. Glacial-interglacial CO_2 change: The iron hypothesis. *Paleoceanography* 5(1):1-13. DOI: 10.1029/PA005i001p00001.

Martinez, M., H. Harder, T. A. Kovacs, J. B. Simpas, J. Bassis, R. Lesher, W. H. Brune, G. J. Frost, E. J. Williams, C. A. Stroud, B. T. Jobson, J. M. Roberts, S. R. Hall, R. E. Shetter, B. Wert, A. Fried, B. Alicke, J. Stutz, V. L. Young, A. B. White, and R. J. Zamora. 2003. OH and HO_2 concentrations, sources, and loss rates during the Southern Oxidants Study in Nashville, Tennessee, summer 1999. *Journal Of Geophysical Research-Atmospheres* 108(D19). DOI: 10.1029/2003jd003551.

Mathiesen, K., and A. Neslen. 2015. VW scandal caused nearly 1m tonnes of extra pollution, analysis shows. *The Guardian*, September 23, 2015. Available at http://www.theguardian.com/business/2015/sep/22/vw-scandal-caused-nearly-1m-tonnes-of-extra-pollution-analysis-shows, accessed April 15, 2016.

Mauldin, R. L., C. A. Cantrell, M. Zondlo, E. Kosciuch, F. L. Eisele, G. Chen, D. Davis, R. Weber, J. Crawford, D. Blake, A. Bandy, and D. Thornton. 2003. Highlights of OH, H_2SO_4, and methane sulfonic acid measurements made aboard the NASA P-3B during Transport and Chemical Evolution over the Pacific. *Journal Of Geophysical Research-Atmospheres* 108(D20). DOI: 10.1029/2003jd003410.

Mauldin, R. L., T. Berndt, M. Sipila, P. Paasonen, T. Petaja, S. Kim, T. Kurten, F. Stratmann, V. M. Kerminen, and M. Kulmala. 2012. A new atmospherically relevant oxidant of sulphur dioxide. *Nature* 488(7410):193-196. DOI: 10.1038/nature11278.

McCluskey, C. S., P. J. DeMott, A. J. Prenni, E. J. T. Levin, G. R. McMeeking, A. P. Sullivan, T. C. J. Hill, S. Nakao, C. M. Carrico, and S. M. Kreidenweis. 2014. Characteristics of atmospheric ice nucleating particles associated with biomass burning in the US: Prescribed burns and wildfires. *Journal Of Geophysical Research-Atmospheres* 119(17). DOI: 10.1002/2014JD021980.

McDonald, B. C., T. R. Dallmann, E. W. Martin, and R. A. Harley. 2012. Long-term trends in nitrogen oxide emissions from motor vehicles at national, state, and air basin scales. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2012jd018304.

McElroy, M. B., R. J. Salawitch, S. C. Wofsy, and J. A. Logan. 1986. Reductions of Antarctic Ozone Due to Synergistic Interactions of Chlorine and Bromine. *Nature* 321(6072):759-762. DOI: 10.1038/321759a0.

McFiggans, G., P. Artaxo, U. Baltensperger, H. Coe, M. C. Facchini, G. Feingold, S. Fuzzi, M. Gysel, A. Laaksonen, U. Lohmann, T. F. Mentel, D. M. Murphy, C. D. O'Dowd, J. R.

Snider, and E. Weingartner. 2006. The effect of physical and chemical aerosol properties on warm cloud droplet activation. *Atmospheric Chemistry and Physics* 6:2593-2649.

McGrath, J. M., A. M. Betzelberger, S. W. Wang, E. Shook, X. G. Zhu, S. P. Long, and E. A. Ainsworth. 2015. An analysis of ozone damage to historical maize and soybean yields in the United States. *Proceedings of the National Academy of Sciences of the United States of America* 112(46):14390-14395. DOI: 10.1073/pnas.1509777112.

McGrath, M. 2016. California methane leak 'largest in US history'. *BBC News*, February 20, 2016. Available at http://www.bbc.com/news/science-environment-35659947, accessed April 15, 2016.

McKeen, S. A., G. Mount, F. Eisele, E. Williams, J. Harder, P. Goldan, W. Kuster, S. C. Liu, K. Baumann, D. Tanner, A. Fried, S. Sewell, C. Cantrell, and R. Shetter. 1997. Photochemical modeling of hydroxyl and its relationship to other species during the Tropospheric OH Photochemistry Experiment. *Journal Of Geophysical Research-Atmospheres* 102(D5):6467-6493. DOI: 10.1029/96jd03322.

McNeill, V. F. 2015. Aqueous Organic Chemistry in the Atmosphere: Sources and Chemical Processing of Organic Aerosols. *Environmental Science & Technology* 49(3):1237-1244. DOI: 10.1021/es5043707.

McNeill, V. F., T. Loerting, F. M. Geiger, B. L. Trout, and M. J. Molina. 2006. Hydrogen chloride-induced surface disordering on ice. *Proceedings of the National Academy of Sciences of the United States of America* 103(25):9422-9427. DOI: 10.1073/pnas.0603494103.

Mellqvist, J., J. Samuelsson, J. Johansson, C. Rivera, B. Lefer, S. Alvarez, and J. Jolly. 2010. Measurements of industrial emissions of alkenes in Texas using the solar occultation flux method. *Journal of Geophysical Research - Atmospheres* 115(D7). DOI: 10.1029/2008JD011682.

Menon, S., J. Hansen, L. Nazarenko, and Y. F. Luo. 2002. Climate effects of black carbon aerosols in China and India. *Science* 297(5590):2250-2253. DOI: 10.1126/science.1075159.

Meskhidze, N., W. L. Chameides, and A. Nenes. 2005. Dust and pollution: A recipe for enhanced ocean fertilization? *Journal Of Geophysical Research-Atmospheres* 110(D3). DOI: 10.1029/2004jd005082.

Metzger, A., B. Verheggen, J. Dommen, J. Duplissy, A. S. H. Prevot, E. Weingartner, I. Riipinen, M. Kulmala, D. V. Spracklen, K. S. Carslaw, and U. Baltensperger. 2010. Evidence for the role of organics in aerosol particle formation under atmospheric conditions. *Proc. Natl. Acad. Sci. USA* 107(15):6646-6651. DOI: 10.1073/pnas.0911330107.

Mickley, L. J., E. M. Leibensperger, D. J. Jacob, and D. Rind. 2012. Regional warming from aerosol removal over the United States: Results from a transient 2010-2050 climate simulation. *Atmospheric Environment* 46:545-553. DOI: 10.1016/j.atmosenv.2011.07.030.

Middlebrook, A. M., D. M. Murphy, S. H. Lee, D. S. Thomson, K. A. Prather, R. J. Wenzel, D. Y. Liu, D. J. Phares, K. P. Rhoads, A. S. Wexler, M. V. Johnston, J. L. Jimenez, J. T. Jayne, D. R. Worsnop, I. Yourshaw, J. H. Seinfeld, and R. C. Flagan. 2003. A comparison of particle mass spectrometers during the 1999 Atlanta Supersite Project. *J. Geophys. Res.* 108(D7):Art. No. 8424. DOI: 10.1029/2001JD000660

Middleton, J. T. 1956. Response of Plants to Air Pollution. *Journal of the Air Pollution Control Association* 6(1):7-50. DOI: 10.1080/00966665.1956.10467730.

Middleton, J. T., J. B. Kendrick Jr., and H. W. Schwalm. 1950. Smog in the South Coastal Area: Injury to herbaceous plants in the affected area found to be result of air pollution by gases and aerosols *California Agriculture* 4(11):7-10.

Mielke, L. H., A. Furgeson, and H. D. Osthoff. 2011. Observation of $ClNO_2$ in a Mid-Continental Urban Environment. *Environmental Science & Technology* 45(20):8889-8896. DOI: 10.1021/es201955u.

Mihelcic, D., F. Holland, A. Hofzumahaus, L. Hoppe, S. Konrad, P. Musgen, H. W. Patz, H. J. Schafer, T. Schmitz, A. Volz-Thomas, K. Bachmann, S. Schlomski, U. Platt, A. Geyer, B. Alicke, and G. K. Moortgat. 2003. Peroxy radicals during BERLIOZ at Pabstthum: Measurements, radical budgets and ozone production. *Journal Of Geophysical Research-Atmospheres* 108(D4). DOI: 10.1029/2001jd001014.

Millennium Ecosystem Assessment. 2005. *Ecosystems and Human Well-being: General Synthesis*. Washington, DC: Island Press.

Milly, P. C. D., R. T. Wetherald, K. A. Dunne, and T. L. Delworth. 2002. Increasing risk of great floods in a changing climate. *Nature* 415(6871):514-517.

Ming, Y., and V. Ramaswamy. 2011. A Model Investigation of Aerosol-Induced Changes in Tropical Circulation. *Journal of Climate* 24(19):5125-5133. DOI: 10.1175/2011JCLI4108.1.

Minkler, M., A. P. Garcia, J. Williams, T. LoPresti, and J. Lilly. 2010. Si Se Puede: Using Participatory Research to Promote Environmental Justice in a Latino Community in San Diego, California. *Journal of Urban Health-Bulletin of the New York Academy of Medicine* 87(5):796-812. DOI: 10.1007/s11524-010-9490-0.

Moise, T., J. M. Flores, and Y. Rudich. 2015. Optical Properties of Secondary Organic Aerosols and Their Changes by Chemical Processes. *Chemical Reviews* 115(10):4400-4439. DOI: 10.1021/cr5005259.

Molina, L. T., and M. J. Molina. 1987. Production of chlorine oxide (Cl_2O_2) from the self-reaction of the chlorine oxide (ClO) radical. *Journal of Physical Chemistry* 91(2):433-436. DOI: 10.1021/j100286a035.

Monks, P. S. 2005. Gas-phase radical chemistry in the troposphere. *Chemical Society Reviews* 34(5):376-395. DOI: 10.1039/b307982c.

Montzka, S. A., E. J. Dlugokencky, and J. H. Butler. 2011. Non-CO2 greenhouse gases and climate change. *Nature* 476(7358):43-50. DOI: 10.1038/nature10322.

Morales Betancourt, R., and A. Nenes. 2014. Understanding the contributions of aerosol properties and parameterization discrepancies to droplet number variability in a global climate model. *Atmospheric Chemistry and Physics* 14(9):4809-4826. DOI: 10.5194/acp-14-4809-2014.

Morello-Frosch, R., and B. M. Jesdale. 2006. Separate and unequal: Residential segregation and estimated cancer risks associated with ambient air toxics in US metropolitan areas. *Environmental Health Perspectives* 114(3):386-393. DOI: 10.1289/ehp.8500.

Moreno, L. A. L., O. Stetzer, and U. Lohmann. 2013. Contact freezing: a review of experimental studies. *Atmospheric Chemistry and Physics* 13(19):9745-9769. DOI: 10.5194/acp-13-9745-2013.

Muhlbauer, A., and U. Lohmann. 2009. Sensitivity studies of aerosol-cloud interactions in mixed-phase orographic precipitation. *Journal of the Atmospheric Sciences* 66(9):2517-2538. DOI: 10.1175/2009JAS3001.1.

Muller, N. Z., and R. Mendelsohn. 2007. Measuring the damages of air pollution in the United States. *Journal of Environmental Economics and Management* 54(1):1-14. DOI: 10.1016/j.jeem.2006.12.002.

Munger, J. W., S. C. Wofsy, P. S. Bakwin, S. M. Fan, M. L. Goulden, B. C. Daube, A. H. Goldstein, K. E. Moore, and D. R. Fitzjarrald. 1996. Atmospheric deposition of reactive nitrogen oxides and ozone in a temperate deciduous forest and a subarctic woodland .1. Measurements and mechanisms. *Journal Of Geophysical Research-Atmospheres* 101(D7):12639-12657. DOI: 10.1029/96jd00230.

Murphy, D. M. 2007. The design of single particle laser mass spectrometers. *Mass Spectrom. Rev.* 26:150-165.

Murphy, D. M., D. J. Cziczo, K. D. Froyd, P. K. Hudson, B. M. Matthew, A. M. Middlebrook, R. E. Peltier, A. Sullivan, D. S. Thomson, and R. J. Weber. 2006. Single-particle mass spectrometry of tropospheric aerosol particles. *J. Geophys. Res.* 111:D23S32. DOI: 10.1029/2006JD007340.

Murray, B. J., T. W. Wilson, S. Dobbie, Z. Q. Cui, S. M. R. K. Al-Jumur, O. Mohler, M. Schnaiter, R. Wagner, S. Benz, M. Niemand, H. Saathoff, V. Ebert, S. Wagner, and B. Karcher. 2010. Heterogeneous nucleation of ice particles on glassy aerosols under cirrus conditions. *Nature geoscience* 3(4):233-237. DOI: 10.1038/NGEO817.

Murray, B. J., D. O'Sullivan, J. D. Atkinson, and M. E. Webb. 2012. Ice nucleation by particles immersed in supercooled cloud droplets. *Chemical Society Reviews* 41(19):6519-6554. DOI: 10.1039/c2cs35200a.

Myhre, G., B. H. Samset, M. Schulz, Y. Balkanski, S. Bauer, T. K. Berntsen, H. Bian, N. Bellouin, M. Chin, T. Diehl, R. C. Easter, J. Feichter, S. J. Ghan, D. Hauglustaine, T. Iversen, S. Kinne, A. Kirkevag, J. F. Lamarque, G. Lin, X. Liu, M. T. Lund, G. Luo, X. Ma, T. van Noije, J. E. Penner, P. J. Rasch, A. Ruiz, O. Seland, R. B. Skeie, P. Stier, T. Takemura, K. Tsigaridis, P. Wang, Z. Wang, L. Xu, H. Yu, F. Yu, J. H. Yoon, K. Zhang, H. Zhang, and C. Zhou. 2013. Radiative forcing of the direct aerosol effect from AeroCom Phase II simulations. *Atmospheric Chemistry and Physics* 13(4):1853-1877. DOI: 10.5194/acp-13-1853-2013.

Nadelhoffer, K. J., B. A. Emmett, P. Gundersen, O. J. Kjonaas, C. J. Koopmans, P. Schleppi, A. Tietema, and R. F. Wright. 1999. Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. *Nature* 398(6723):145-148. DOI: 10.1038/18205.

NADP (National Atospheric Deposition Program). 2014. NADP Map Highlight. *NADP: Keeping You Connected* 2014(1).

NASEM (The National Academies of Sciences, Engineering, and Medicine). 2015. *SBIR at the National Science Foundation*. Washington, DC: National Academies Press.

NASEM. 2016. *Attribution of Extreme Weather Events in the Context of Climate Change*. Washington, DC: National Academies Press.

Nasir, Z. A., I. Colbeck, Z. P. Bharucha, L. C. Campos, and Z. Ali. 2014. Ethno-Environmental Knowledge as A Tool to Combat Indoor Air Pollution in Low Income Countries: A Case Study from Rural Communities in Pakistan. *Journal of Environment and Human* 1(2):165-175.

Nazaroff, W. W., and A. H. Goldstein. 2015. Indoor chemistry: research opportunities and challenges. *Indoor Air* 25(4):357-361. DOI: 10.1111/ina.12219.

Nenes, A., M. D. Krom, N. Mihalopoulos, P. Van Cappellen, Z. Shi, A. Bougiatioti, P. Zarmpas, and B. Herut. 2011. Atmospheric acidification of mineral aerosols: a source of bioavailable phosphorus for the oceans. *Atmospheric Chemistry and Physics* 11(13):6265-6272. DOI: 10.5194/acp-11-6265-2011.

Neu, J. L., T. Flury, G. L. Manney, M. L. Santee, N. J. Livesey, and J. Worden. 2014. Tropospheric ozone variations governed by changes in stratospheric circulation. *Nature Geoscience* 7(5):340-344. DOI: 10.1038/ngeo2138.

Newman, P. A., L. D. Oman, A. R. Douglass, E. L. Fleming, S. M. Frith, M. M. Hurwitz, S. R. Kawa, C. H. Jackman, N. A. Krotkov, E. R. Nash, J. E. Nielsen, S. Pawson, R. S. Stolarski, and G. J. M. Velders. 2009. What would have happened to the ozone layer if chlorofluorocarbons (CFCs) had not been regulated? *Atmospheric Chemistry and Physics* 9(6):2113-2128. DOI: 10.5194/acp-9-2113-2009.

Nguyen, T. B., J. D. Crounse, A. P. Teng, J. M. S. Clair, F. Paulot, G. M. Wolfe, and P. O. Wennberg. 2015a. Rapid deposition of oxidized biogenic compounds to a temperate forest. *Proceedings of the National Academy of Sciences of the United States of America* 112(5):E392-E401. DOI: 10.1073/pnas.1418702112.

Nguyen, T. L., H. Lee, D. A. Matthews, M. C. McCarthy, and J. F. Stanton. 2015b. Stabilization of the Simplest Criegee Intermediate from the Reaction between Ozone and Ethylene: A High-Level Quantum Chemical and Kinetic Analysis of Ozonolysis. *Journal of Physical Chemistry A* 119(22):5524-5533. DOI: 10.1021/acs.jpca.5b02088.

NIC (National Intelligence Council). 2015. National Intelligence Council Publications. Available at http://www.dni.gov/index.php/about/organization/national-intelligence-council-nic-publications, accessed October 19, 2015. Available accessed.

Niedermeier, D., R. A. Shaw, S. Hartmann, H. Wex, T. Clauss, J. Voigtlander, and F. Stratmann. 2011. Heterogeneous ice nucleation: exploring the transition from stochastic to singular freezing behavior. *Atmospheric Chemistry and Physics* 11(16):8767-8775. DOI: 10.5194/acp-11-8767-2011.

Nøjgaard, J. K. 2010. Indoor measurements of the sum of the nitrate radical, NO_3, and nitrogen pentoxide, N_2O_5 in Denmark. *Chemosphere* 79(8):898-904. DOI: 10.1016/j.chemosphere.2010.02.025.

Nolscher, A. C., T. Butler, J. Auld, P. Veres, A. Munoz, D. Taraborrelli, L. Vereecken, J. Lelieveld, and J. Williams. 2014. Using total OH reactivity to assess isoprene photooxidation via measurement and model. *Atmospheric Environment* 89:453-463. DOI: 10.1016/j.atmosenv.2014.02.024.

Nordhaus, W. D. 1993. Rolling the Dice: An Optimal Transition Path for Controlling Greenhouse Gases. *Resource and Energy Economics* 15(1):27-50. DOI: 10.1016/0928-7655(93)90017-O.

Noziere, B., P. Dziedzic, and A. Cordova. 2009. Products and Kinetics of the Liquid-Phase Reaction of Glyoxal Catalyzed by Ammonium Ions (NH4+). *Journal of Physical Chemistry A* 113(1):231-237. DOI: 10.1021/jp8078293.

Nozière, B., M. Kalberer, M. Claeys, J. Allan, B. D’Anna, S. Decesari, E. Finessi, M. Glasius, I. Grgić, J. F. Hamilton, T. Hoffmann, Y. Iinuma, M. Jaoui, A. Kahnt, C. J. Kampf, I. Kourtchev, W. Maenhaut, N. Marsden, S. Saarikoski, J. Schnelle-Kreis, J. D. Surratt, S. Szidat, R. Szmigielski, and A. Wisthaler. 2015. The Molecular Identification of Organic

Compounds in the Atmosphere: State of the Art and Challenges. *Chemical Reviews* 115(10):3919-3983. DOI: 10.1021/cr5003485.

NRC (National Research Council). 1979. *Stratospheric Ozone Depletion by Halocarbons: Chemistry and Transport*. Washington, DC: National Academy Press.

NRC. 1982. *Causes and Effects of Stratospheric Ozone Reduction: an Update*. Washington, DC: National Academy Press.

NRC. 1983. *Acid Deposition: Atmospheric Processes in Eastern North America*. Washington, DC: National Academy Press.

NRC. 1984a. *Global Tropospheric Chemistry: A Plan for Action*. Washington, DC: National Academy Press.

NRC. 1984b. *Causes and Effects of Changes in Stratospheric Ozone: Update 1983*. Washington, DC: National Academy Press.

NRC. 1986. *Acid Deposition: Long-Term Trends*. Washington, DC: National Academy Press.

NRC. 1991. *Rethinking the Ozone Problem in Urban and Regional Air Pollution*. Washington, DC: National Academy Press.

NRC. 1993. *Protecting Visibility in National Parks and Wilderness Areas*. Washington, DC: National Academy Press.

NRC. 1996. *A Plan for a Research Program on Aerosol Radiative Forcing and Climate Change*. Washington, DC: The National Academies Press

NRC. 1998. *The Atmospheric Sciences: Entering the Twenty-First Century.*Washington, DC: National Academy Press.

NRC. 2001. *Global Air Quality: An Imperative for Long-Term Observational Strategies*. Washington, DC: National Academies Press.

NRC. 2003. *Critical Issues in Weather Modification Research*. Washington, DC: The National Academies Press.

NRC. 2005. *Radiative Forcing of Climate Change.* Washington, DC: National Academies Press.

NRC. 2005. *Strategic Guidance for the National Science Foundation's Support of the Atmospheric Sciences: An Interim Report*. Washington, DC: The National Academies Press.

NRC. 2007. *Strategic Guidance for the National Science Foundation's Support of the Atmospheric Sciences.* Washington, DC: National Academies Press.

NRC. 2010. *Global Sources of Local Pollution.* Washington, DC: National Academies Press.

NRC. 2011. *America's Climate Choices*. Washington, DC: National Academies Press.

NRC. 2013. *Abrupt Impacts of Climate Change: Anticipating Surprises*. Washington, DC: National Academies Press.

O'Brien, R. E., B. Wang, S. T. Kelly, N. Lindt, Y. You, A. K. Bertram, S. R. Leone, A. Laskin, and M. K. Gilles. 2015. Liquid–Liquid Phase Separation in Aerosol Particles: Imaging at the Nanometer Scale. *Environ. Sci. Technol.* 49(8):4995–5002.

Obrist, D., E. Tas, M. Peleg, V. Matveev, X. Fain, D. Asaf, and M. Luria. 2011. Bromine-induced oxidation of mercury in the mid-latitude atmosphere. *Nature geoscience* 4(1):22-26. DOI: 10.1038/NGEO1018.

O'Connor, F. M., O. Boucher, N. Gedney, C. D. Jones, G. A. Folberth, R. Coppell, P. Friedlingstein, W. J. Collins, J. Chappellaz, J. Ridley, and C. E. Johnson. 2010. Possible Role of Wetlands, Permafrost, and Methane Hydrates in the Methane Cycle under Future Climate Change: A Review. *Reviews of Geophysics* 48. DOI: 10.1029/2010rg000326.

Odén, S. 1968. *The acidification of air precipitation and its consequences in the natural environment. Bulletin of the Ecological Research Communications NFR*. Arlington, VA: Translation Consultants Ltd.

O'Dowd, C. D., M. C. Facchini, F. Cavalli, D. Ceburnis, M. Mircea, S. Decesari, S. Fuzzi, Y. J. Yoon, and J. P. Putaud. 2004. Biogenically driven organic contribution to marine aerosol. *Nature* 431(7009):676-680. DOI: 10.1038/nature02959.

O'Driscoll, P., N. Minogue, N. Takenaka, and J. Sodeau. 2008. Release of nitric oxide and iodine to the atmosphere from the freezing of sea-salt aerosol components. *Journal of Physical Chemistry A* 112(8):1677-1682. DOI: 10.1021/jp710464c.

Odum, J. R., T. P. W. Jungkamp, R. J. Griffin, R. C. Flagan, and J. H. Seinfeld. 1997. The atmospheric aerosol-forming potential of whole gasoline vapor. *Science* 276(5309):96-99. DOI: 10.1126/science.276.5309.96.

Office of Environmental Health Hazard Assessment. 2015. Air Toxicology and Epidemiology: Air Pollution and Children's Health. Available at http://www.oehha.ca.gov/public_info/facts/airkids.html, accessed September 18.

Olenius, T., I. Riipinen, K. Lehtipalo, and H. Vehkamaki. 2014. Growth rates of atmospheric molecular clusters based on appearance times and collision-evaporation fluxes: Growth by monomers. *Journal of Aerosol Science* 78:55-70. DOI: 10.1016/j.jaerosci.2014.08.008.

Olsson, D., I. Mogren, and B. Forsberg. 2013. Air pollution exposure in early pregnancy and adverse pregnancy outcomes: a register-based cohort study. *Bmj Open* 3(2). DOI: 10.1136/bmjopen-2012-001955.

Oppenheimer, M. 1983. The relationship of sulfur emissions to sulfate in precipitation. *Atmospheric Environment* 17(3):451-460. DOI: 10.1016/0004-6981(83)90118-x.

Ortega, I. K., O. Kupiainen, T. Kurten, T. Olenius, O. Wilkman, M. J. McGrath, V. Loukonen, and H. Vehkamaki. 2012. From quantum chemical formation free energies to evaporation rates. *Atmospheric Chemistry and Physics* 12(1):225-235. DOI: 10.5194/acp-12-225-2012.

Osborn, D. L., and C. A. Taatjes. 2015. The physical chemistry of Criegee intermediates in the gas phase. *International Reviews in Physical Chemistry* 34(3):309-360. DOI: 10.1080/0144235X.2015.1055676.

Owen, R. C., O. R. Cooper, A. Stohl, and R. E. Honrath. 2006. An analysis of the mechanisms of North American pollutant transport to the central North Atlantic lower free troposphere. *Journal Of Geophysical Research-Atmospheres* 111(D23). DOI: 10.1029/2006jd007062.

Palmer, P. I. 2008. Quantifying sources and sinks of trace gases using space-borne measurements: current and future science. *Philosophical Transactions of the Royal Society a-Mathematical Physical and Engineering Sciences* 366(1885):4509-4528. DOI: 10.1098/rsta.2008.0176.

Pandis, S. N., and J. H. Seinfeld. 2006. *Atmospheric Chemistry: From Air Pollution to Climate Change*. Hoboken, N. J. : Wiley.

Parekh, P., S. Dutkiewicz, M. J. Follows, and T. Ito. 2006. Atmospheric carbon dioxide in a less dusty world. *Geophysical Research Letters* 33(3):4. DOI: 10.1029/2005gl025098.

Park, J. H., A. H. Goldstein, J. Timkovsky, S. Fares, R. Weber, J. Karlik, and R. Holzinger. 2013. Active Atmosphere-Ecosystem Exchange of the Vast Majority of Detected Volatile Organic Compounds. *Science* 341(6146):643-647. DOI: 10.1126/science.1235053.

Park, R. J., S. K. Hong, H. A. Kwon, S. Kim, A. Guenther, J. H. Woo, and C. P. Loughner. 2014. An evaluation of ozone dry deposition simulations in East Asia. *Atmospheric Chemistry and Physics* 14(15):7929-7940. DOI: 10.5194/acp-14-7929-2014.

Patra, P. K., M. C. Krol, S. A. Montzka, T. Arnold, E. L. Atlas, B. R. Lintner, B. B. Stephens, B. Xiang, J. W. Elkins, P. J. Fraser, A. Ghosh, E. J. Hintsa, D. F. Hurst, K. Ishijima, P. B. Krummel, B. R. Miller, K. Miyazaki, F. L. Moore, J. Muhle, S. O'Doherty, R. G. Prinn, L. P. Steele, M. Takigawa, H. J. Wang, R. F. Weiss, S. C. Wofsy, and D. Young. 2014. Observational evidence for interhemispheric hydroxyl-radical parity. *Nature* 513(7517):219-223. DOI: 10.1038/nature13721.

Paulik, L. B., C. E. Donald, B. W. Smith, L. G. Tidwell, K. A. Hobbie, L. Kincl, E. N. Haynes, and K. A. Anderson. 2015. Impact of natural gas extraction on PAH levels in ambient air. *Environmental Science & Technology* 49(8):5203-5210. DOI: 10.1021/es506095e.

Paulot, F., J. D. Crounse, H. G. Kjaergaard, A. Kurten, J. M. St Clair, J. H. Seinfeld, and P. O. Wennberg. 2009. Unexpected Epoxide Formation in the Gas-Phase Photooxidation of Isoprene. *Science* 325(5941):730-733. DOI: 10.1126/science.1172910.

Peeters, J., and J. F. Muller. 2010. HOx radical regeneration in isoprene oxidation via peroxy radical isomerisations. II: experimental evidence and global impact. *Physical Chemistry Chemical Physics* 12(42):14227-14235. DOI: 10.1039/c0cp00811g.

Peeters, J., T. L. Nguyen, and L. Vereecken. 2009. HO_x radical regeneration in the oxidation of isoprene. *Physical Chemistry Chemical Physics* 11(28):5935-5939. DOI: 10.1039/b908511d.

Perraud, V., E. A. Bruns, M. J. Ezell, S. N. Johnson, Y. Yu, M. L. Alexander, A. Zelenyuk, D. Imre, W. L. Chang, D. Dabdub, J. F. Pankow, and B. J. Finlayson-Pitts. 2011. Non-equilibrium atmospheric secondary organic aerosol formation and growth. *PNAS* 109(8):2836-2841. DOI: 10.1073/pnas.1119909109

Perri, M. J., Y. B. Lim, S. P. Seitzinger, and B. J. Turpin. 2010. Organosulfates from glycolaldehyde in aqueous aerosols and clouds: Laboratory studies. *Atmospheric Environment* 44(21-22):2658-2664. DOI: 10.1016/j.atmosenv.2010.03.031.

Perring, A. E., J. P. Schwarz, D. Baumgardner, M. T. Hernandez, D. V. Spracklen, C. L. Heald, R. S. Gao, G. Kok, G. R. McMeeking, J. B. McQuaid, and D. W. Fahey. 2015. Airborne observations of regional variation in fluorescent aerosol across the United States. *Journal Of Geophysical Research-Atmospheres* 120(3):1153-1170. DOI: 10.1002/2014jd022495.

Peters, A., R. Ruckerl, and J. Cyrys. 2011. Lessons From Air Pollution Epidemiology for Studies of Engineered Nanomaterials. *Journal of Occupational and Environmental Medicine* 53(6):S8-S13. DOI: 10.1097/JOM.0b013e31821ad5c0.

Pétron, G., G. Frost, B. R. Miller, A. I. Hirsch, S. A. Montzka, A. Karion, M. Trainer, C. Sweeny, A. E. Andrews, L. Miller, J. Kofler, A. Bar-Ilan, E. J. Dlugokencky, L. Patrick, C. T. Moore Jr., T. B. Ryerson, C. Siso, W. Kolodzey, P. M. Lang, T. Conway, P. Novelli, K. Masarie, B. Hall, D. Guenther, D. Kitzis, J. Miller, D. Welsh, D. Wolfe, W. Neff, and P. Tans. 2012. Hydrocarbon emissions characterization in the Colorado Front Range: A pilot study. *Journal of Geophysical Research - Atmospheres* 117(D4).

Pfister, G., P. Hess, L. Emmons, J. Lamarque, C. Wiedinmyer, D. Edwards, G. Petron, J. Gille, and G. Sachse. 2005. Quantifying CO emissions from the 2004 Alaskan wildfires using MOPITT CO data. *Geophysical Research Letters* 32(11). DOI: 10.1029/2005GL022995.

Philipona, R., K. Behrens, and C. Ruckstuhl. 2009. How declining aerosols and rising greenhouse gases forced rapid warming in Europe since the 1980s. *Geophysical Research Letters* 36. DOI: 10.1029/2008gl036350.

Phillips, S. M., and G. D. Smith. 2014. Light Absorption by Charge Transfer Complexes in Brown Carbon Aerosols. *Environmental Science & Technology Letters* 1(10):382-386. DOI: 10.1021/ez500263j.

Phillips, S. M., and G. D. Smith. 2015. Further Evidence for Charge Transfer Complexes in Brown Carbon Aerosols from Excitation-Emission Matrix Fluorescence Spectroscopy. *Journal of Physical Chemistry A* 119(19):4545-4551. DOI: 10.1021/jp510709e.

Pilinis, C., S. N. Pandis, and J. H. Seinfeld. 1995. Sensitivity of Direct Climate Forcing by Atmospheric Aerosols to Aerosol-Size and Composition. *Journal Of Geophysical Research-Atmospheres* 100(D9):18739-18754. DOI: 10.1029/95jd02119.

Pinto, J. P., Y. L. Yung, D. Rind, G. L. Russell, J. A. Lerner, J. E. Hansen, and S. Hameed. 1983. A General-Circulation Model Study of Atmospheric Carbon-Monoxide. *Journal of Geophysical Research-Oceans and Atmospheres* 88(Nc6):3691-3702. DOI: 10.1029/Jc088ic06p03691.

Pirrone, N., S. Cinnirella, X. Feng, R. B. Finkelman, H. R. Friedli, J. Leaner, R. Mason, A. B. Mukherjee, G. B. Stracher, D. G. Streets, and K. Telmer. 2010. Global mercury emissions to the atmosphere from anthropogenic and natural sources. *Atmospheric Chemistry and Physics* 10(13):5951-5964. DOI: 10.5194/acp-10-5951-2010.

Plath, K. L., J. L. Axson, G. C. Nelson, K. Takahashi, R. T. Skodje, and V. Vaida. 2009. Gas-phase vibrational spectra of glyoxylic acid and its gem diol monohydrate: Implications for atmospheric chemistry. *Reaction Kinetics and Catalysis Letters* 96(2):209-224. DOI: 10.1007/s11144-009-5528-2.

Pleim, J., R. Mathur, S. T. Rao, J. Fast, and A. Baklanov. 2014. Integrated Meteorology and Chemistry Modeling Evaluation and Research Needs. *Bulletin of the American Meteorological Society* 95(4):Es81-Es84. DOI: 10.1175/Bams-D-13-00107.1.

Pollack, I. B., T. B. Ryerson, M. Trainer, D. D. Parrish, A. E. Andrews, E. L. Atlas, D. R. Blake, S. S. Brown, R. Commane, B. C. Daube, J. A. de Gouw, W. P. Dube, J. Flynn, G. J. Frost, J. B. Gilman, N. Grossberg, J. S. Holloway, J. Kofler, E. A. Kort, W. C. Kuster, P. M. Lang, B. Lefer, R. A. Lueb, J. A. Neuman, J. B. Nowak, P. C. Novelli, J. Peischl, A. E. Perring, J. M. Roberts, G. Santoni, J. P. Schwarz, J. R. Spackman, N. L. Wagner, C. Warneke, R. A. Washenfelder, S. C. Wofsy, and B. Xiang. 2012. Airborne and ground-based observations of a weekend effect in ozone, precursors, and oxidation products in the California South Coast Air Basin. *Journal Of Geophysical Research-Atmospheres* 117. DOI: 10.1029/2011jd016772.

Polson, D., D. Fowler, E. Nemitz, U. Skiba, A. McDonald, D. Famulari, C. Di Marco, I. Simmons, K. Weston, R. Purvis, H. Coe, A. J. Manning, H. Webster, M. Harrison, D. O'Sullivan, C. Reeves, and D. Oram. 2011. Estimation of spatial apportionment of greenhouse gas emissions for the UK using boundary layer measurements and inverse modelling technique. *Atmospheric Environment* 45(4):1042-1049. DOI: 10.1016/j.atmosenv.2010.10.011.

Pongracz, R., J. Bartholy, and A. Kis. 2014. Estimation of future precipitation conditions for Hungary with special focus on dry periods. *Idojaras* 118(4):305-321.

Pope III, C. A., and D. W. Dockery. 2006. Health effects of fine particulate air pollution: Lines that connect. *J. Air & Waste Manage. Assoc.* 56:709-742.

Pope, C. A., M. Ezzati, and D. W. Dockery. 2013. Fine particulate air pollution and life expectancies in the United States: The role of influential observations. *Journal of the Air & Waste Management Association* 63(2):129-132. DOI: 10.1080/10962247.2013.760353.

Pöschl, U. 2005. Atmospheric aerosols: composition, transformation, climate and health effects. *Angew. Chem. Int. Ed.* 44:7520-7540.

Pöschl, U., and M. Shiraiwa. 2015. Multiphase Chemistry at the Atmosphere–Biosphere Interface Influencing Climate and Public Health in the Anthropocene. *Chemical Reviews* 115(10):4440-4475. DOI: 10.1021/cr500487s.

Power, M. C., M.-A. Kioumourtzoglou, J. E. Hart, O. I. Okereke, F. Laden, and M. G. Weisskopf. 2015. The relation between past exposure to fine particulate air pollution and prevalent anxiety: observational cohort study. *BMJ* 350:1-9. DOI: 10.1136/bmj.h1111.

Prather, K. A., C. D. Hatch, and V. H. Grassian. 2008. Analysis of atmospheric aerosols. *Annu. Rev. Anal. Chem.* 1:485-514. DOI: 10.1146/annurev.anchem.1.031207.113030.

Prather, M. J., M. B. McElroy, and S. C. Wofsy. 1984. Reductions in Ozone at High-Concentrations of Stratospheric Halogens. *Nature* 312(5991):227-231. DOI: 10.1038/312227a0.

Prather, M., M. Mcelroy, S. Wofsy, G. Russell, and D. Rind. 1987. Chemistry of the Global Troposphere: Fluorocarbons as Tracers of Air Motion. *Journal Of Geophysical Research-Atmospheres* 92(D6):6579-6613. DOI: 10.1029/Jd092id06p06579.

Pratt, K. A., L. E. Hatch, and K. A. Prather. 2009a. Seasonal Volatility Dependence of Ambient Particle Phase Amines. *Environmental Science & Technology* 43(14):5276-5281. DOI: 10.1021/es803189n.

Pratt, K. A., P. J. DeMott, J. R. French, Z. Wang, D. L. Westphal, A. J. Heymsfield, C. H. Twohy, A. J. Prenni, and K. A. Prather. 2009b. In situ detection of biological particles in cloud ice-crystals. *Nature geoscience* 2(6):397-400. DOI: 10.1038/NGEO521.

Pratt, K. A., K. D. Custard, P. B. Shepson, T. A. Douglas, D. Pohler, S. General, J. Zielcke, W. R. Simpson, U. Platt, D. J. Tanner, L. G. Huey, M. Carlsen, and B. H. Stirm. 2013. Photochemical production of molecular bromine in Arctic surface snowpacks. *Nature geoscience* 6(5):351-356. DOI: 10.1038/NGEO1779.

Prospero, J. M., and P. J. Lamb. 2003. African droughts and dust transport to the Caribbean: Climate change implications. *Science* 302(5647):1024-1027. DOI: 10.1126/science.1089915.

Pusede, S. E., A. L. Steiner, and R. C. Cohen. 2015. Temperature and Recent Trends in the Chemistry of Continental Surface Ozone. *Chemical Reviews* 115(10):3898-3918. DOI: 10.1021/cr5006815.

Pyle, J., and M. Prather. 1996. *Global Tracer Transport Models: Report of a Scientific Symposium, Bermuda, 10-13 Dec 1990.* WCRP CAS/JSC Report No. 24 (WMO, TD-No.770). Geneva: WMO.

Quinn, P. K., D. B. Collins, V. H. Grassian, K. A. Prather, and T. S. Bates. 2015. Chemistry and Related Properties of Freshly Emitted Sea Spray Aerosol. *Chemical Reviews* 115(10):4383-4399. DOI: 10.1021/cr5007139.

Rao, S. T. 2015. Introduction to JA&WMA Special Issue on Air Quality and Human Health. *Journal of the Air & Waste Management Association* 65(5):515. DOI: 10.1080/10962247.2015.1032793.

Rappengluck, B., L. Ackermann, S. Alvarez, J. Golovko, M. Buhr, R. A. Field, J. Soltis, D. C. Montague, B. Hauze, S. Adamson, D. Risch, G. Wilkerson, D. Bush, T. Stoeckenius, and

C. Keslar. 2014. Strong wintertime ozone events in the Upper Green River basin, Wyoming. *Atmospheric Chemistry and Physics* 14(10):4909-4934. DOI: 10.5194/acp-14-4909-2014.

Reale, O., K. M. Lau, A. da Silva, and T. Matsui. 2014. Impact of assimilated and interactive aerosol on tropical cyclogenesis. *Geophysical Research Letters* 41(9):3282-3288. DOI: 10.1002/2014GL059918.

Ren, C., and A. R. Mackenzie. 2005. Cirrus parametrization and the role of ice nuclei. *Quarterly Journal of the Royal Meteorological Society* 131(608):1585-1605. DOI: 10.1256/qj.04.126.

Ren, X. R., H. Harder, M. Martinez, R. L. Lesher, A. Oliger, J. B. Simpas, W. H. Brune, J. J. Schwab, K. L. Demerjian, Y. He, X. L. Zhou, and H. G. Gao. 2003. OH and HO_2 chemistry in the urban atmosphere of New York City. *Atmospheric Environment* 37(26):3639-3651. DOI: 10.1016/S1352-2310(03)00459-X.

Ren, X. R., W. H. Brune, J. Q. Mao, M. J. Mitchell, R. L. Lesher, J. B. Simpas, A. R. Metcalf, J. J. Schwab, C. X. Cai, Y. Q. Li, K. L. Demerjian, H. D. Felton, G. Boynton, A. Adams, J. Perry, Y. He, X. L. Zhou, and J. Hou. 2006. Behavior of OH and HO_2 in the winter atmosphere in New York City. *Atmospheric Environment* 40:S252-S263. DOI: 10.1016/j.atmosenv.2005.11.073.

Ren, X. R., J. R. Olson, J. H. Crawford, W. H. Brune, J. Q. Mao, R. B. Long, Z. Chen, G. Chen, M. A. Avery, G. W. Sachse, J. D. Barrick, G. S. Diskin, L. G. Huey, A. Fried, R. C. Cohen, B. Heikes, P. O. Wennberg, H. B. Singh, D. R. Blake, and R. E. Shetter. 2008. HO_x chemistry during INTEX-A 2004: Observation, model calculation, and comparison with previous studies. *Journal Of Geophysical Research-Atmospheres* 113(D5). DOI: 10.1029/2007jd009166.

Ren, X., J. Mao, W. H. Brune, C. A. Cantrell, R. L. Mauldin, R. S. Hornbrook, E. Kosciuch, J. R. Olson, J. H. Crawford, G. Chen, and H. B. Singh. 2012. Airborne intercomparison of HO_x measurements using laser-induced fluorescence and chemical ionization mass spectrometry during ARCTAS. *Atmospheric Measurement Techniques* 5(8):2025-2037. DOI: 10.5194/amt-5-2025-2012.

Ren, X. R., D. van Duin, M. Cazorla, S. Chen, J. Q. Mao, L. Zhang, W. H. Brune, J. H. Flynn, N. Grossberg, B. L. Lefer, B. Rappengluck, K. W. Wong, C. Tsai, J. Stutz, J. E. Dibb, B. T. Jobson, W. T. Luke, and P. Kelley. 2013. Atmospheric oxidation chemistry and ozone production: Results from SHARP 2009 in Houston, Texas. *Journal Of Geophysical Research-Atmospheres* 118(11):5770-5780. DOI: 10.1002/jgrd.50342.

Renbaum-Wolff, L., J. W. Grayson, A. P. Bateman, M. Kuwata, M. Sallier, B. J. Murray, J. E. Shilling, S. T. Martin, and A. K. Bertram. 2013. Viscosity of α-pinene secondary organic aerosol material and implications for particle growth and reactivity. *PNAS* 110(20):8014-8019. DOI: 10.1073/pnas.12195488110.

Riccobono, F., S. Schobesberger, C. E. Scott, J. Dommen, I. K. Ortega, L. Rondo, J. Almeida, A. Amorim, F. Bianchi, M. Breitenlechner, A. David, A. Downard, E. M. Dunne, J. Duplissy, S. Ehrhart, R. C. Flagan, A. Franchin, A. Hansel, H. Junninen, M. Kajos, H. Keskinen, A. Kupc, A. Kurten, A. N. Kvashin, A. Laaksonen, K. Lehtipalo, V. Makhmutov, S. Mathot, T. Nieminen, A. Onnela, T. Petaja, A. P. Praplan, F. D. Santos, S. Schallhart, J. H. Seinfeld, M. Sipila, D. V. Spracklen, Y. Stozhkov, F. Stratmann, A. Tome, G. Tsagkogeorgas, P. Vaattovaara, Y. Viisanen, A. Vrtala, P. E. Wagner, E. Weingartner, H. Wex, D. Wimmer, K. S. Carslaw, J. Curtius, N. M. Donahue, J. Kirkby,

M. Kulmala, D. R. Worsnop, and U. Baltensperger. 2014. Oxidation Products of Biogenic Emissions Contribute to Nucleation of Atmospheric Particles. *Science* 344(6185):717-721. DOI: 10.1126/science.1243527.

Rich, N. 2016. The Invisible Catastrophe. *New York Times Magazine,* March 31, 2016. Available at http://www.nytimes.com/2016/04/03/magazine/the-invisible-catastrophe.html, accessed April 15, 2016.

Ridley, D. A., C. L. Heald, and B. J. Ford. 2012. North African Dust Export and Impacts: An Integrated Satellite and Model Perspective. *J. Geophys. Res.* 117(D02202). DOI: 10.1029/2011JD016794.

Ridley, D. A., C. L. Heald, and J. M. Prospero. 2014. What controls the recent changes in African mineral dust aerosol across the Atlantic? *Atmos. Chem. Phys.* 14:5735-5747. DOI: doi:10.5194/acp-14-5735-2014.

Riemer, D. D., E. C. Apel, J. J. Orlando, G. S. Tyndall, W. H. Brune, E. J. Williams, W. A. Lonneman, and J. D. Neece. 2008. Unique isoprene oxidation products demonstrate chlorine atom chemistry occurs in the Houston, Texas urban area. *Journal of Atmospheric Chemistry* 61(3):227-242. DOI: 10.1007/s10874-009-9134-5.

Riipinen, I., T. Yli-Juuti, J. R. Pierce, T. Petaja, D. R. Worsnop, M. Kulmala, and N. M. Donahue. 2012. The contribution of organics to atmospheric nanoparticle growth. *Nature geoscience* 5(7):453-458. DOI: 10.1038/ngeo1499.

Robine, J. M., S. L. K. Cheung, S. Le Roy, H. Van Oyen, C. Griffiths, J. P. Michel, and F. R. Herrmann. 2008. Death toll exceeded 70,000 in Europe during the summer of 2003. *Comptes Rendus Biologies* 331(2):171-U175. DOI: 10.1016/j.crvi.2007.12.001.

Robinson, A. L., N. M. Donahue, M. K. Shrivastava, E. A. Weitkamp, A. M. Sage, A. P. Grieshop, T. E. Lane, J. R. Pierce, and S. N. Pandis. 2007. Rethinking Organic Aerosols: Semivolatile Emissions and Photochemical Aging. *Science* 315(5816):1259-1262. DOI: 10.1126/science.1133061.

Rohrer, F., K. D. Lu, A. Hofzumahaus, B. Bohn, T. Brauers, C. C. Chang, H. Fuchs, R. Haseler, F. Holland, M. Hu, K. Kita, Y. Kondo, X. Li, S. R. Lou, A. Oebel, M. Shao, L. M. Zeng, T. Zhu, Y. H. Zhang, and A. Wahner. 2014. Maximum efficiency in the hydroxyl-radical-based self-cleansing of the troposphere. *Nature geoscience* 7(8):559-563. DOI: 10.1038/NGEO2199.

Roscioli, J. R., T. I. Yacovitch, C. Floerchinger, A. L. Mitchell, D. S. Tkacik, R. Subramanian, D. M. Martinez, T. L. Vaughn, L. Williams, D. Zimmerle, A. L. Robinson, S. C. Herndon, and A. J. Marchese. 2015. Measurements of methane emissions from natural gas gathering facilities and processing plants: measurement methods. *Atmospheric Measurement Techniques* 8(5):2017-2035. DOI: 10.5194/amt-8-2017-2015.

Rosenfeld, D. 2006. Atmosphere - Aerosols, clouds, and climate. *Science* 312(5778):1323-1324. DOI: 10.1126/science.1128972.

Rosenfeld, D., U. Lohmann, G. B. Raga, C. D. O'Dowd, M. Kulmala, S. Fuzzi, A. Reissell, and M. O. Andreae. 2008. Flood or drought: How do aerosols affect precipitation? *Science* 321(5894):1309-1313. DOI: 10.1126/science.1160606.

Rosenlof, K. H., and G. C. Reid. 2008. Trends in the temperature and water vapor content of the tropical lower stratosphere: Sea surface connection. *Journal Of Geophysical Research-Atmospheres* 113(D6). DOI: 10.1029/2007jd009109.

Rosenlof, K. H., S. J. Oltmans, D. Kley, J. M. Russell, E. W. Chiou, W. P. Chu, D. G. Johnson, K. K. Kelly, H. A. Michelsen, G. E. Nedoluha, E. E. Remsberg, G. C. Toon, and M. P.

McCormick. 2001. Stratospheric water vapor increases over the past half-century. *Geophysical Research Letters* 28(7):1195-1198. DOI: 10.1029/2000gl012502.

Rowland, F. S. 2001. Atmospheric changes caused by human activities: From science to regulation. *Ecology Law Quarterly* 27(4):1261-1293.

Rowland, F. S. 2006. Stratospheric ozone depletion. *Philosophical Transactions of the Royal Society B-Biological Sciences* 361(1469):769-790. DOI: 10.1098/rstb.2005.1783.

Rubasinghege, G., and V. H. Grassian. 2013. Role(s) of adsorbed water in the surface chemistry of environmental interfaces. *Chemical Communications* 49(30):3071-3094. DOI: 10.1039/c3cc38872g.

Ryder, O. S., N. R. Campbell, M. Shaloski, H. Al-Mashat, G. M. Nathanson, and T. H. Bertram. 2015. Role of Organics in Regulating $ClNO_2$ Production at the Air-Sea Interface. *Journal of Physical Chemistry A* 119(31):8519-8526. DOI: 10.1021/jp5129673.

Sage, A. M., E. A. Weitkamp, A. L. Robinson, and N. M. Donahue. 2008. Evolving mass spectra of the oxidized component of organic aerosol: results from aerosol mass spectrometer analyses of aged diesel emissions. *Atmospheric Chemistry and Physics* 8(5):1139-1152.

Saide, P. E., S. N. Spak, R. B. Pierce, J. A. Otkin, T. K. Schaack, A. K. Heidinger, A. M. da Silva, M. Kacenelenbogen, J. Redemann, and G. R. Carmichael. 2015. Central American biomass burning smoke can increase tornado severity in the US. *Geophysical Research Letters* 42(3):956-965. DOI: 10.1002/2014GL062826.

Sala, O. E., F. S. Chapin, J. J. Armesto, E. Berlow, J. Bloomfield, R. Dirzo, E. Huber-Sanwald, L. F. Huenneke, R. B. Jackson, A. Kinzig, R. Leemans, D. M. Lodge, H. A. Mooney, M. Oesterheld, N. L. Poff, M. T. Sykes, B. H. Walker, M. Walker, and D. H. Wall. 2000. Biodiversity - Global biodiversity scenarios for the year 2100. *Science* 287(5459):1770-1774. DOI: 10.1126/science.287.5459.1770.

Saleeby, S. M., W. R. Cotton, and J. D. Fuller. 2011. The Cumulative Impact of Cloud Droplet Nucleating Aerosols on Orographic Snowfall in Colorado. *Journal of Applied Meteorology and Climatology* 50(3):604-625. DOI: 10.1175/2010jamc2594.1.

Saleh, R., E. S. Robinson, D. S. Tkacik, A. T. Ahern, S. Liu, A. C. Aiken, R. C. Sullivan, A. A. Presto, M. K. Dubey, R. J. Yokelson, N. M. Donahue, and A. L. Robinson. 2014. Brownness of organics in aerosols from biomass burning linked to their black carbon content. *Nature geoscience* 7(9):647-650. DOI: 10.1038/NGEO2220.

Samet, J. M., F. Dominici, F. C. Curriero, I. Coursac, and S. L. Zeger. 2000. Fine particulate air pollution and mortality in 20 US Cities, 1987-1994. *New England Journal of Medicine* 343(24):1742-1749. DOI: 10.1056/nejm200012143432401.

San Martini, F. M., C. A. Hasenkopf, and D. C. Roberts. 2015. Statistical analysis of PM2.5 observations from diplomatic facilities in China. *Atmospheric Environment* 110:174-185. DOI: 10.1016/j.atmosenv.2015.03.060.

Sander, S. P., R. R. Friedl, J. R. Barker, D. M. Golden, M. J. Kurylo, P. H. Wine, J. P. D. Abbatt, J. B. Burkholder, C. E. Kolb, G. K. Moortgat, R. E. Huie, and V. L. Orkin. 2011. *Chemical Kinetics and Photochemical Data for Use in Atmospheric Studies. Evaluation Number 17. http://jpldataeval.jpl.nasa.gov/*. Jet Propulson Laboratory, Pasadena, California. JPL Publication 10-6.

Schiermeier, Q. 2015. The science behind the Volkswagen emissions scandal: Debacle has wide-ranging implications, but many already knew that diesel emissions tests were problematic. *Nature*. DOI: 10.1038/nature.2015.18426.

Schill, G. P., and M. A. Tolbert. 2013. Heterogeneous ice nucleation on phase-separated organic-sulfate particles: effect of liquid vs. glassy coatings. *Atmospheric Chemistry and Physics* 13(9):4681-4695. DOI: 10.5194/acp-13-4681-2013.

Schindler, D. W. 1988. Effects of acid-rain on fresh-water ecosystems. *Science* 239(4836):149-157. DOI: 10.1126/science.239.4836.149.

Schmid, P. E., and D. Niyogi. 2013. Impact of city size on precipitation-modifying potential. *Geophysical Research Letters* 40(19):5263-5267. DOI: 10.1002/grl.50656.

Schnell, R. C., S. J. Oltmans, R. R. Neely, M. S. Endres, J. V. Molenar, and A. B. White. 2009. Rapid photochemical production of ozone at high concentrations in a rural site during winter. *Nature geoscience* 2(2):120-122. DOI: 10.1038/NGEO415.

Schobesberger, S., H. Junninen, F. Bianchi, G. Lonn, M. Ehn, K. Lehtipalo, J. Dommen, S. Ehrhart, I. K. Ortega, A. Franchin, T. Nieminen, F. Riccobono, M. Hutterli, J. Duplissy, J. Almeida, A. Amorim, M. Breitenlechner, A. J. Downard, E. M. Dunne, R. C. Flagan, M. Kajos, H. Keskinen, J. Kirkby, A. Kupc, A. Kurten, T. Kurten, A. Laaksonen, S. Mathot, A. Onnela, A. P. Praplan, L. Rondo, F. D. Santos, S. Schallhart, R. Schnitzhofer, M. Sipila, A. Tome, G. Tsagkogeorgas, H. Vehkamaki, D. Wimmer, U. Baltensperger, K. S. Carslaw, J. Curtius, A. Hansel, T. Petaja, M. Kulmala, N. M. Donahue, and D. R. Worsnop. 2013. Molecular understanding of atmospheric particle formation from sulfuric acid and large oxidized organic molecules. *Proceedings of the National Academy of Sciences of the United States of America* 110(43):17223-17228. DOI: 10.1073/pnas.1306973110.

Schroth, A. W., J. Crusius, E. R. Sholkovitz, and B. C. Bostick. 2009. Iron solubility driven by speciation in dust sources to the ocean. *Nature geoscience* 2(5):337-340. DOI: 10.1038/NGEO501.

Schulz, M., J. M. Prospero, A. R. Baker, F. Dentener, L. Ickes, P. S. Liss, N. M. Mahowald, S. Nickovic, C. P. Garcia-Pando, S. Rodriguez, M. Sarin, I. Tegen, and R. A. Duce. 2012. Atmospheric Transport and Deposition of Mineral Dust to the Ocean: Implications for Research Needs. *Environmental Science & Technology* 46(19):10390-10404. DOI: 10.1021/es300073u.

Schumann, U., and H. Huntrieser. 2007. The global lightning-induced nitrogen oxides source. *Atmospheric Chemistry and Physics* 7(14):3823-3907.

Screen, J. A., and I. Simmonds. 2014. Amplified mid-latitude planetary waves favour particular regional weather extremes. *Nature Climate Change* 4(8):704-709.

Seinfeld, J. H. 2008. Atmospheric science: Black carbon and brown clouds*Nature geoscience* 1:15-16. DOI: 10.1038/ngeo.2007.62.

Seinfeld, J. H., and S. N. Pandis. 2006. *Atmospheric chemistry and physics: From air pollution to climate change*. Hoboken, N.J.: J. Wiley.

Seinfeld, J. H., C. Bretherton, K. S. Carslaw, H. Coe, P. J. DeMott, E. J. Dunlea, G. Feingold, S. Ghan, A. B. Guenther, R. Kahn, I. Kraucunas, S. M. Kreidenweis, M. J. Molina, A. Nenes, J. E. Penner, K. A. Prather, V. Ramanathan, V. Ramaswamy, P. J. Rasch, A. R. Ravishankara, D. Rosenfeld, G. Stephens, and R. Wood. 2016. Improving our fundamental understanding of the role of aerosol-cloud interactions in the climate system. *Proceedings of the National Academy of Sciences of the United States of America* 113(21): 5781–5790. DOI: 10.1073/pnas.1514043113.

Selin, N. E. 2009. Global Biogeochemical Cycling of Mercury: A Review. *Annual Review of Environment and Resources* 34:43-63. DOI: 10.1146/annurev.environ.051308.084314.

Sessions, W. R., J. S. Reid, A. Benedetti, P. R. Colarco, A. da Silva, S. Lu, T. Sekiyama, T. Y. Tanaka, J. M. Baldasano, S. Basart, M. E. Brooks, T. F. Eck, M. Iredell, J. A. Hansen, O. C. Jorba, H. M. H. Juang, P. Lynch, J. J. Morcrette, S. Moorthi, J. Mulcahy, Y. Pradhan, M. Razinger, C. B. Sampson, J. Wang, and D. L. Westphal. 2015. Development towards a global operational aerosol consensus: Basic climatological characteristics of the International Cooperative for Aerosol Prediction Multi-Model Ensemble (ICAP-MME). *Atmospheric Chemistry and Physics* 15(1):335-362. DOI: 10.5194/acp-15-335-2015.

Sevanian, A., J. F. Mead, and R. A. Stein. 1979. Epoxides as Products of Lipid Autoxidation in Rat Lungs. *Lipids* 14(7):634-643. DOI: 10.1007/Bf02533449.

Shaw, S. L., B. Gantt, and N. Meskhidze. 2010. Production and Emissions of Marine Isoprene and Monoterpenes: A Review. *Advances in Meteorology*. DOI: 10.1155/2010/408696.

Shindell, D., and G. Faluvegi. 2009. Climate response to regional radiative forcing during the twentieth century. *Nature geoscience* 2(4):294-300. DOI: 10.1038/NGEO473.

Shindell, D., G. Faluvegi, A. Lacis, J. Hansen, R. Ruedy, and E. Aguilar. 2006. Role of tropospheric ozone increases in 20th-century climate change. *Journal Of Geophysical Research-Atmospheres* 111(D8). DOI: 10.1029/2005jd006348.

Shindell, D., J. C. I. Kuylenstierna, E. Vignati, R. van Dingenen, M. Amann, Z. Klimont, S. C. Anenberg, N. Muller, G. Janssens-Maenhout, F. Raes, J. Schwartz, G. Faluvegi, L. Pozzoli, K. Kupiainen, L. Hoglund-Isaksson, L. Emberson, D. Streets, V. Ramanathan, K. Hicks, N. T. K. Oanh, G. Milly, M. Williams, V. Demkine, and D. Fowler. 2012. Simultaneously Mitigating Near-Term Climate Change and Improving Human Health and Food Security. *Science* 335(6065):183-189. DOI: 10.1126/science.1210026.

Shinn, E. A., G. W. Smith, J. M. Prospero, P. Betzer, M. L. Hayes, V. Garrison, and R. T. Barber. 2000. African dust and the demise of Caribbean coral reefs. *Geophysical Research Letters* 27(19):3029-3032. DOI: 10.1029/2000gl011599.

Shiogama, H., M. Watanabe, Y. Imada, M. Mori, Y. Kamae, M. Ishii, and M. Kimoto. 2014. Attribution of the June-July 2013 Heat Wave in the Southwestern United States. *Sola* 10:122-126. DOI: 10.2151/sola.2014-025.

Shiraiwa, M., M. Ammann, T. Koop, and U. Poschl. 2011. Gas uptake and chemical aging of semisolid organic aerosol particles. *Proceedings of the National Academy of Sciences of the United States of America* 108(27):11003-11008. DOI: 10.1073/pnas.1103045108

Shrivastava, M., J. Fast, R. Easter, W. I. Gustafson, R. A. Zaveri, J. L. Jimenez, P. Saide, and A. Hodzic. 2011. Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach. *Atmospheric Chemistry and Physics* 11(13):6639-6662. DOI: 10.5194/acp-11-6639-2011.

Simpson, W. R., S. S. Brown, A. Saiz-Lopez, J. A. Thornton, and R. von Glasow. 2015. Tropospheric Halogen Chemistry: Sources, Cycling, and Impacts. *Chemical Reviews* 115(10):4035-4062. DOI: 10.1021/cr5006638.

Singer, B. C., B. K. Coleman, H. Destaillats, A. T. Hodgson, M. M. Lunden, C. J. Weschler, and W. W. Nazaroff. 2006. Indoor secondary pollutants from cleaning product and air freshener use in the presence of ozone. *Atmospheric Environment* 40(35):6696-6710. DOI: 10.1016/j.atmosenv.2006.06.005.

Sinha, V., J. Williams, J. N. Crowley, and J. Lelieveld. 2008. The comparative reactivity method - a new tool to measure total OH reactivity in ambient air. *Atmospheric Chemistry and Physics* 8(8):2213-2227. DOI: 10.5194/acp-8-2213-2008.

Sitch, S., P. M. Cox, W. J. Collins, and C. Huntingford. 2007. Indirect radiative forcing of climate change through ozone effects on the land-carbon sink. *Nature* 448(7155):791-U794. DOI: 10.1038/nature06059.

Small, A. A., J. B. Stefik, J. Verlinde, and N. C. Johnson. 2011. The Cloud Hunter's Problem: An Automated Decision Algorithm to Improve the Productivity of Scientific Data Collection in Stochastic Environments. *Monthly Weather Review* 139(7):2276-2289. DOI: 10.1175/2010MWR3576.1.

Smith, A. B., and R. W. Katz. 2013. US billion-dollar weather and climate disasters: data sources, trends, accuracy and biases. *Natural Hazards* 67(2):387-410. DOI: 10.1007/s11069-013-0566-5.

Smith, J. N., K. C. Barsanti, H. R. Friedli, M. Ehn, M. Kulmala, D. R. Collins, J. H. Scheckman, B. J. Williams, and P. H. McMurry. 2010. Observations of aminium salts in atmospheric nanoparticles and possible climatic implications. *Proceedings of the National Academy of Sciences of the United States of America* 107(15):6634-6639. DOI: 10.1073/pnas.0912127107.

Smith, K. R., N. Bruce, K. Balakrishnan, H. Adair-Rohani, J. Balmes, Z. Chafe, M. Dherani, H. D. Hosgood, S. Mehta, D. Pope, E. Rehfuess, and H. C. R. E. Grp. 2014. Millions Dead: How Do We Know and What Does It Mean? Methods Used in the Comparative Risk Assessment of Household Air Pollution. *Annual Review of Public Health, Vol 35* 35:185-206. DOI: 10.1146/annurev-publhealth-032013-182356.

Smith, P. J. 2007. Climate Change, Mass Migration and the Military Response. *Orbis* 51(4):617-633.

Smith, R. A. 1852. On the air and rain of Manchester. *Memoirs of the Literary and Philosophical Society of Manchester* 2(10):207-217.

Snyder, L. P. 1994. “The Death-Dealing Smog Over Donora, Pennsylvania”: Industrial Air Pollution, Public Health Policy, and the Politics of Expertise, 1948–1949. *Environmental History Review* 18(1):117-139. DOI: 10.2307/3984747.

Soerensen, A. L., E. M. Sunderland, C. D. Holmes, D. J. Jacob, R. M. Yantosca, H. Skov, J. H. Christensen, S. A. Strode, and R. P. Mason. 2010. An Improved Global Model for Air-Sea Exchange of Mercury: High Concentrations over the North Atlantic. *Environmental Science & Technology* 44(22):8574-8580. DOI: 10.1021/es102032g.

Solomon, S., R. R. Garcia, F. S. Rowland, and D. J. Wuebbles. 1986. On the Depletion of Antarctic Ozone. *Nature* 321(6072):755-758. DOI: 10.1038/321755a0.

Solomon, S., D. J. Ivy, D. Kinnison, M. J. Mills, R. R. N. III, and A. Schmidt. 2016. Emergence of healing in the Antarctic ozone layer. *Science*. DOI: 10.1126/science.aae0061.

Son, S.-W., E. P. Gerber, J. Perlwitz, L. M. Polvani, N. P. Gillett, K.-H. Seo, V. Eyring, T. G. Shepherd, D. Waugh, H. Akiyoshi, J. Austin, A. Baumgaertner, S. Bekki, P. Braesicke, C. Brühl, N. Butchart, M. P. Chipperfield, D. Cugnet, M. Dameris, S. Dhomse, S. Frith, H. Garny, R. Garcia, S. C. Hardiman, P. Jöckel, J. F. Lamarque, E. Mancini, M. Marchand, M. Michou, T. Nakamura, O. Morgenstern, G. Pitari, D. A. Plummer, J. Pyle, E. Rozanov, J. F. Scinocca, K. Shibata, D. Smale, H. Teyssèdre, W. Tian, and Y. Yamashita. 2010. Impact of stratospheric ozone on Southern Hemisphere circulation change: A multimodel assessment. *Journal of Geophysical Research - Atmospheres* 115(D3). DOI: 10.1029/2010JD01427.

Sorooshian, A., V. Varutbangkul, F. J. Brechtel, B. Ervens, G. Feingold, R. Bahreini, S. M. Murphy, J. S. Holloway, E. L. Atlas, G. Buzorius, H. Jonsson, R. C. Flagan, and J. H.

Seinfeld. 2006. Oxalic acid in clear and cloudy atmospheres: Analysis of data from ICARTT 2004. *J. of Geophys. Res. - Atmosphere* 111(D23):D23S45.

Sorooshian, A., M. L. Lu, F. J. Brechtel, H. Jonsson, G. Feingold, R. C. Flagan, and J. H. Seinfeld. 2007. On the source of organic acid aerosol layers above clouds. *Environmental Science & Technology* 41(13):4647-4654.

Spracklen, D. V., and C. L. Heald. 2014. The contribution of fungal spores and bacteria to regional and global aerosol number and ice nucleation immersion freezing rates. *Atmospheric Chemistry and Physics* 14(17):9051-9059. DOI: 10.5194/acp-14-9051-2014.

Staudt, A. C., D. J. Jacob, J. A. Logan, D. Bachiochi, T. N. Krishnamurti, and G. W. Sachse. 2001. Continental sources, transoceanic transport, and interhemispheric exchange of carbon monoxide over the Pacific. *Journal Of Geophysical Research-Atmospheres* 106(D23):32571-32589. DOI: 10.1029/2001jd900078.

Staudt, A. C., D. J. Jacob, J. A. Logan, D. Bachiochi, T. N. Krishnamurti, and N. Poisson. 2002. Global chemical model analysis of biomass burning and lightning influences over the South Pacific in austral spring. *Journal Of Geophysical Research-Atmospheres* 107(D14). DOI: 10.1029/2000jd000296.

Steffen, A., T. Douglas, M. Amyot, P. Ariya, K. Aspmo, T. Berg, J. Bottenheim, S. Brooks, F. Cobbett, A. Dastoor, A. Dommergue, R. Ebinghaus, C. Ferrari, K. Gardfeldt, M. E. Goodsite, D. Lean, A. J. Poulain, C. Scherz, H. Skov, J. Sommar, and C. Temme. 2008. A synthesis of atmospheric mercury depletion event chemistry in the atmosphere and snow. *Atmospheric Chemistry and Physics* 8(6):1445-1482.

Steffen, W., P. J. Crutzen, and J. R. McNeill. 2007. The Anthropocene: Are humans now overwhelming the great forces of nature. *Ambio* 36(8):614-621. DOI: 10.1579/0044-7447(2007)36[614:Taahno]2.0.Co;2.

Steiner, A. L., S. D. Brooks, C. H. Deng, D. C. O. Thornton, M. W. Pendleton, and V. Bryant. 2015. Pollen as atmospheric cloud condensation nuclei. *Geophysical Research Letters* 42(9):3596-3602. DOI: 10.1002/2015gl064060.

Stevens, B., and G. Feingold. 2009. Untangling aerosol effects on clouds and precipitation in a buffered system. *Nature* 461(7264):607-613. DOI: 10.1038/nature08281.

Stockwell, C. E., P. R. Veres, J. Williams, and R. J. Yokelson. 2015. Characterization of biomass burning emissions from cooking fires, peat, crop residue, and other fuels with high-resolution proton-transfer-reaction time-of-flight mass spectrometry. *Atmospheric Chemistry and Physics* 15(2):845-865. DOI: 10.5194/acp-15-845-2015.

Stokes, D. E. 1997. *Pasteur's Quadrant: Basic Science and Technological Innovation*. Washington, DC: Brookings Institution Press.

Stolarski, R. S., and R. J. Cicerone. 1974. Stratospheric Chlorine: a Possible Sink for Ozone. *Canadian Journal of Chemistry* 52(8):1610-1615. DOI: 10.1139/v74-233.

Stolarski, R. S., A. J. Krueger, M. R. Schoeberl, R. D. Mcpeters, P. A. Newman, and J. C. Alpert. 1986. Nimbus-7 Satellite Measurements of the Springtime Antarctic Ozone Decrease. *Nature* 322(6082):808-811. DOI: 10.1038/322808a0.

Stone, D., L. K. Whalley, and D. E. Heard. 2012. Tropospheric OH and HO_2 radicals: field measurements and model comparisons. *Chemical Society Reviews* 41(19):6348-6404. DOI: 10.1039/c2cs35140d.

Storer, R. L., S. C. van den Heever, and G. L. Stephens. 2010. Modeling Aerosol Impacts on Convective Storms in Different Environments. *Journal of the Atmospheric Sciences* 67(12):3904-3915. DOI: 10.1175/2010jas3363.1.

Stott, P. 2015. Attribution: Weather risks in a warming world. *Nature Climate Change* 5(6):516-517. DOI: 10.1038/nclimate2640.

Stott, P. A., D. A. Stone, and M. R. Allen. 2004. Human contribution to the European heatwave of 2003. *Nature* 432(7017):610-614. DOI: 10.1038/nature03089.

Suntharalingam, P., E. Buitenhuis, C. Le Quere, F. Dentener, C. Nevison, J. H. Butler, H. W. Bange, and G. Forster. 2012. Quantifying the impact of anthropogenic nitrogen deposition on oceanic nitrous oxide. *Geophysical Research Letters* 39. DOI: 10.1029/2011gl050778.

Swap, R., M. Garstang, S. Greco, R. Talbot, and P. Kallberg. 1992. Saharan Dust in the Amazon Basin. *Tellus Series B-Chemical and Physical Meteorology* 44(2):133-149. DOI: 10.1034/j.1600-0889.1992.t01-1-00005.x.

Taatjes, C. A., O. Welz, A. J. Eskola, J. D. Savee, A. M. Scheer, D. E. Shallcross, B. Rotavera, E. P. F. Lee, J. M. Dyke, D. K. W. Mok, D. L. Osborn, and C. J. Percival. 2013. Direct Measurements of Conformer-Dependent Reactivity of the Criegee Intermediate CH_3CHOO. *Science* 340(6129):177-180. DOI: 10.1126/science.1234689.

Taatjes, C. A., D. E. Shallcross, and C. J. Percival. 2014. Research frontiers in the chemistry of Criegee intermediates and tropospheric ozonolysis. *Physical Chemistry Chemical Physics* 16(5):1704-1718. DOI: 10.1039/c3cp52842a.

Tai, P. K., M. Val Martin, and C. L. Heald. 2014. Threat to future global food security from climate change and ozone air pollution. *Nature Climate Change* 4:817-821. DOI: doi:10.1038/nclimate2317.

Tan, D., I. Faloona, J. B. Simpas, W. Brune, J. Olson, J. Crawford, M. Avery, G. Sachse, S. Vay, S. Sandholm, H.-W. Guan, T. Vaughn, J. Mastromarino, B. Heikes, J. Snow, J. Podolske, and H. Singh. 2001. OH and HO_2 in the tropical Pacific: Results from PEM-Tropics B. *Journal of Geophysical Research - Atmospheres* 106(D23). DOI: 10.1029/2001JD900002.

Taraborrelli, D., M. G. Lawrence, J. N. Crowley, T. J. Dillon, S. Gromov, C. B. M. Gross, L. Vereecken, and J. Lelieveld. 2012. Hydroxyl radical buffered by isoprene oxidation over tropical forests. *Nature geoscience* 5(3):190-193. DOI: 10.1038/NGEO1405.

Taylor, M. R., E. S. Rubin, and D. A. Hounshell. 2003. Effect of government actions on technological innovation for SO2 control. *Environmental Science & Technology* 37(20):4527-4534. DOI: 10.1021/es034223b.

Thomas, J. L., J. E. Dibb, L. G. Huey, J. Liao, D. Tanner, B. Lefer, R. von Glasow, and J. Stutz. 2012. Modeling chemistry in and above snow at Summit, Greenland - Part 2: Impact of snowpack chemistry on the oxidation capacity of the boundary layer. *Atmospheric Chemistry and Physics* 12(14):6537-6554. DOI: 10.5194/acp-12-6537-2012.

Thompson, D. W. J., and S. Solomon. 2002. Interpretation of recent Southern Hemisphere climate change. *Science* 296(5569):895-899. DOI: 10.1126/science.1069270.

Thornton, J. A., J. P. Kercher, T. P. Riedel, N. L. Wagner, J. Cozic, J. S. Holloway, W. P. Dube, G. M. Wolfe, P. K. Quinn, A. M. Middlebrook, B. Alexander, and S. S. Brown. 2010. A large atomic chlorine source inferred from mid-continental reactive nitrogen chemistry. *Nature* 464(7286):271-274. DOI: 10.1038/nature08905.

Tobias, D. J., and J. C. Hemminger. 2008. Chemistry - Getting specific about specific ion effects. *Science* 319(5867):1197-1198. DOI: 10.1126/science.1152799.

Tobias, H. J., and P. J. Ziemann. 2000. Thermal desorption mass spectrometric analysis of organic aerosol formed from reactions of 1-tetradecene and O_3 in the presence of alcohols and carboxylic acids. *Environmental Science and Technology* 34:2105-2115.

Tobo, Y., A. J. Prenni, P. J. DeMott, J. A. Huffman, C. S. McCluskey, G. X. Tian, C. Pohlker, U. Poschl, and S. M. Kreidenweis. 2013. Biological aerosol particles as a key determinant of ice nuclei populations in a forest ecosystem. *Journal Of Geophysical Research-Atmospheres* 118(17):10100-10110. DOI: 10.1002/jgrd.50801.

Tong, D. Q., P. Lee, and R. D. Saylor. 2012. New Directions: The need to develop process-based emission forecasting models. *Atmospheric Environment* 47:560-561. DOI: 10.1016/j.atmosenv.2011.10.070.

Toom-Sauntry, D., and L. A. Barrie. 2002. Chemical composition of snowfall in the high Arctic: 1990-1994. *Atmospheric Environment* 36(15-16):2683-2693. DOI: 10.1016/S1352-2310(02)00115-2.

Tröstl, J., W. K. Chuang, H. Gordon, M. Heinritzi, C. Yan, U. Molteni, L. Ahlm, C. Frege, F. Bianchi, R. Wagner, M. Simon, K. Lehtipalo, C. Williamson, J. S. Craven, J. Duplissy, A. Adamov, J. Almeida, A. K. Bernhammer, M. Breitenlechner, S. Brilke, A. Dias, S. Ehrhart, R. C. Flagan, A. Franchin, C. Fuchs, R. Guida, M. Gysel, A. Hansel, C. R. Hoyle, T. Jokinen, H. Junninen, J. Kangasluoma, H. Keskinen, J. Kim, M. Krapf, A. Kurten, A. Laaksonen, M. Lawler, M. Leiminger, S. Mathot, O. Mohler, T. Nieminen, A. Onnela, T. Petaja, F. M. Piel, P. Miettinen, M. P. Rissanen, L. Rondo, N. Sarnela, S. Schobesberger, K. Sengupta, M. Sipilaa, J. N. Smith, G. Steiner, A. Tome, A. Virtanen, A. C. Wagner, E. Weingartner, D. Wimmer, P. M. Winkler, P. L. Ye, K. S. Carslaw, J. Curtius, J. Dommen, J. Kirkby, M. Kulmala, I. Riipinen, D. R. Worsnop, N. M. Donahue, and U. Baltensperger. 2016. The role of low-volatility organic compounds in initial particle growth in the atmosphere. *Nature* 533(7604):527-531. DOI: 10.1038/nature18271.

Turner, J., T. J. Bracegirdle, T. Phillips, G. J. Marshall, and J. S. Hosking. 2013. An Initial Assessment of Antarctic Sea Ice Extent in the CMIP5 Models. *Journal of Climate* 26(5):1473-1484. DOI: 10.1175/Jcli-D-12-00068.1.

UCAR (University Corporation for Atmospheric Chemistry). 1959. *Preliminary plans for a National Institute for Atmsoperic Research. Second progress report of the University Committee on Atmospheric Research*. Boulder, Colorado: National Center for Atmospheric Research. Available at http://www.ncar.ucar.edu/documents/bluebook1959.pdf, accessed April 18, 2016.

UNEP (United Nations Environment Programme) and WMO (World Meteorological Organization. 2011. *Integrated Assessment of Black Carbon and Tropospheric Ozone*. Nairobi, Kenya: United Nations Environment Programme.

Unger, N. 2014. Human land-use-driven reduction of forest volatiles cools global climate. *Nature Climate Change* 4(10):907-910. DOI: 10.1038/nclimate2347.

United Nations. 2015a. Transforming our world: the 2030 Agenda for Sustainable Development. https://sustainabledevelopment.un.org/post2015/transformingourworld, accessed October 14, 2015.

United Nations. 2015b. *World Population Prospects: The 2015 Revision. Key Findings and Advance Tables*. Working Paper No. ESA/P/WP.241. New York: United Nations, Department of Economic and Social Affairs, Population Division.

Urbanski, S. P., W. M. Hao, and B. Nordgren. 2011. The wildland fire emission inventory: western United States emission estimates and an evaluation of uncertainty. *Atmospheric Chemistry and Physics* 11(24):12973-13000. DOI: 10.5194/acp-11-12973-2011.

USGCRP (United States Global Change Research Program). 2009. *Global Climate Change Impacts in the United States.* T. R. Karl, J. M. Melillo, and T. C. Peterson, eds. New York: Cambridge University Press.

Val Martin, M., C. L. Heald, and S. R. Arnold. 2014. Coupling dry deposition to vegetation phenology in the Community Earth System Model: Implications for the simulation of surface O3. *Geophys. Res. Lett.* 41. DOI: doi:10.1002/2014GL059651.

Vali, G. 1994. Freezing Rate Due to Heterogeneous Nucleation. *Journal of the Atmospheric Sciences* 51(13):1843-1856. DOI: 10.1175/1520-0469(1994)051<1843:Frdthn>2.0.Co;2.

Van den Heever, S. C., G. G. Carrió, W. R. Cotton, P. J. DeMott, and A. J. Prenni. 2006. Impacts of nucleating aerosol on Florida storms. Part I: Mesoscale simulations. *Journal of the Atmospheric Sciences* 63(7):1752-1775.

van der Werf, G. R., J. T. Randerson, L. Giglio, G. J. Collatz, P. S. Kasibhatla, and A. F. Arellano. 2006. Interannual variability in global biomass burning emissions from 1997 to 2004. *Atmospheric Chemistry and Physics* 6:3423-3441.

van Dijk, A., H. Slaper, P. N. den Outer, O. Morgenstern, P. Braesicke, J. A. Pyle, H. Garny, A. Stenke, M. Dameris, A. Kazantzidis, K. Tourpali, and A. F. Bais. 2013. Skin Cancer Risks Avoided by the Montreal Protocol-Worldwide Modeling Integrating Coupled Climate-Chemistry Models with a Risk Model for UV. *Photochemistry and Photobiology* 89(1):234-246. DOI: 10.1111/j.1751-1097.2012.01223.x.

Vautard, R., M. Beekmann, and L. Menut. 2000. Applications of adjoint modelling in atmospheric chemistry: sensitivity and inverse modelling. *Environmental Modelling & Software* 15(6-7):703-709. DOI: 10.1016/S1364-8152(00)00058-X.

Vehkamaki, H., and I. Riipinen. 2012. Thermodynamics and kinetics of atmospheric aerosol particle formation and growth. *Chemical Society Reviews* 41(15):5160-5173. DOI: 10.1039/c2cs00002d.

Vereecken, L., J. F. Muller, and J. Peeters. 2007. Low-volatility poly-oxygenates in the OH-initiated atmospheric oxidation of alpha-pinene: impact of non-traditional peroxyl radical chemistry. *Physical Chemistry Chemical Physics* 9(38):5241-5248. DOI: 10.1039/b708023a.

Vereecken, L., D. R. Glowacki, and M. J. Pilling. 2015. Theoretical Chemical Kinetics in Tropospheric Chemistry: Methodologies and Applications. *Chemical Reviews* 115(10):4063-4114. DOI: 10.1021/cr500488p.

Virtanen, A., J. Joutsensaari, T. Koop, J. Kannosto, P. Yli-Pirila, J. Leskinen, J. M. Makela, J. K. Holopainen, U. Poschl, M. Kulmala, D. R. Worsnop, and A. Laaksonen. 2010. An amorphous solid state of biogenic secondary organic aerosol particles. *Nature* 467(7317):824-827. DOI: Doi 10.1038/Nature09455.

Virtanen, A., J. Kannosto, H. Kuuluvainen, A. Arffman, J. Joutsensaari, E. Saukko, L. Hao, P. Yli-Pirila, P. Tiitta, J. K. Holopainen, J. Keskinen, D. R. Worsnop, J. N. Smith, and A. Laaksonen. 2011. Bounce behavior of freshly nucleated biogenic secondary organic aerosol particles. *Atmos. Chem. Phys.* 11:8759-8766. DOI: 10.5194/acp-11-8759-2011.

Vitousek, P., J. Aber, R. Howarth, G. Likens, P. Matson, D. Schindler, W. Schlesinger, and G. Tilman. 1997. Human Alteration of the Global Nitrogen Cycle: Causes and Consequences. *Issues in Ecology* 1.

Volkamer, R., J. L. Jimenez, F. San Martini, K. Dzepina, Q. Zhang, D. Salcedo, L. T. Molina, D. R. Worsnop, and M. J. Molina. 2006. Secondary organic aerosol formation from anthropogenic air pollution: Rapid and higher than expected. *Geophysical Research Letters* 33(17). DOI: 10.1029/2006gl026899.

von Bobrutzki, K., C. F. Braban, D. Famulari, S. K. Jones, T. Blackall, T. E. L. Smith, M. Blom, H. Coe, M. Gallagher, M. Ghalaieny, M. R. McGillen, C. J. Percival, J. D. Whitehead, R. Ellis, J. Murphy, A. Mohacsi, A. Pogany, H. Junninen, S. Rantanen, M. A. Sutton, and E. Nemitz. 2010. Field inter-comparison of eleven atmospheric ammonia measurement techniques. *Atmospheric Measurement Techniques* 3(1):91-112.

von Ehrenstein, O. S., H. Aralis, M. Cockburn, and B. Ritz. 2014. In Utero Exposure to Toxic Air Pollutants and Risk of Childhood Autism. *Epidemiology* 25(6):851-858. DOI: 10.1097/ede.0000000000000150.

von Hobe, M., S. Bekki, S. Borrmann, F. Cairo, F. D'Amato, G. Di Donfrancesco, A. Dornbrack, A. Ebersoldt, M. Ebert, C. Emde, I. Engel, M. Ern, W. Frey, S. Genco, S. Griessbach, J. U. Grooss, T. Gulde, G. Gunther, E. Hosen, L. Hoffmann, V. Homonnai, C. R. Hoyle, I. S. A. Isaksen, D. R. Jackson, I. M. Janosi, R. L. Jones, K. Kandler, C. Kalicinsky, A. Keil, S. M. Khaykin, F. Khosrawi, R. Kivi, J. Kuttippurath, J. C. Laube, F. Lefevre, R. Lehmann, S. Ludmann, B. P. Luo, M. Marchand, J. Meyer, V. Mitev, S. Molleker, R. Muller, H. Oelhaf, F. Olschewski, Y. Orsolini, T. Peter, K. Pfeilsticker, C. Piesch, M. C. Pitts, L. R. Poole, F. D. Pope, F. Ravegnani, M. Rex, M. Riese, T. Rockmann, B. Rognerud, A. Roiger, C. Rolf, M. L. Santee, M. Scheibe, C. Schiller, H. Schlager, M. S. de Cumis, N. Sitnikov, O. A. Sovde, R. Spang, N. Spelten, F. Stordal, O. Suminska-Ebersoldt, A. Ulanovski, J. Ungermann, S. Viciani, C. M. Volk, M. V. Scheidt, P. von der Gathen, K. Walker, T. Wegner, R. Weigel, S. Weinbruch, G. Wetzel, F. G. Wienhold, I. Wohltmann, W. Woiwode, I. A. K. Young, V. Yushkov, B. Zobrist, and F. Stroh. 2013. Reconciliation of essential process parameters for an enhanced predictability of Arctic stratospheric ozone loss and its climate interactions (RECONCILE): activities and results. *Atmospheric Chemistry and Physics* 13(18):9233-9268. DOI: 10.5194/acp-13-9233-2013.

Wagner, N. L., C. A. Brock, W. M. Angevine, A. Beyersdorf, P. Campuzano-Jost, D. Day, J. A. de Gouw, G. S. Diskin, T. D. Gordon, M. G. Graus, J. S. Holloway, G. Huey, J. L. Jimenez, D. A. Lack, J. Liao, X. Liu, M. Z. Markovic, A. M. Middlebrook, T. Mikoviny, J. Peischl, A. E. Perring, M. S. Richardson, T. B. Ryerson, J. P. Schwarz, C. Warneke, A. Welti, A. Wisthaler, L. D. Ziemba, and D. M. Murphy. 2015. In situ vertical profiles of aerosol extinction, mass, and composition over the southeast United States during SENEX and SEAC(4)RS: observations of a modest aerosol enhancement aloft. *Atmospheric Chemistry and Physics* 15(12):7085-7102. DOI: 10.5194/acp-15-7085-2015.

Wagner, T., C. Leue, M. Wenig, K. Pfeilsticker, and U. Platt. 2001. Spatial and temporal distribution of enhanced boundary layer BrO concentrations measured by the GOME instrument aboard ERS-2. *Journal Of Geophysical Research-Atmospheres* 106(D20):24225-24235. DOI: 10.1029/2000jd000201.

Wang, Q. Q., D. J. Jacob, J. R. Spackman, A. E. Perring, J. P. Schwarz, N. Moteki, E. A. Marais, C. Ge, J. Wang, and S. R. H. Barrett. 2014a. Global budget and radiative forcing of black carbon aerosol: Constraints from pole-to-pole (HIPPO) observations across the Pacific. *Journal Of Geophysical Research-Atmospheres* 119(1):195-206. DOI: 10.1002/2013JD020824.

Wang, T. J., B. L. Zhuang, S. Li, J. Liu, M. Xie, C. Q. Yin, Y. Zhang, C. Yuan, J. L. Zhu, L. Q. Ji, and Y. Han. 2015a. The interactions between anthropogenic aerosols and the East Asian summer monsoon using RegCCMS. *Journal Of Geophysical Research-Atmospheres* 120(11):5602-5621. DOI: 10.1002/2014JD022877.

Wang, X., C. L. Heald, D. A. Ridley, J. P. Schwarz, J. R. Spackman, A. E. Perring, H. Coe, D. Liu, and A. D. Clarke. 2014b. Exploiting simultaneous observational constraints on mass and absorption to estimate the global direct radiative forcing of black carbon and brown carbon. *Atmospheric Chemistry and Physics* 14(20):10989-11010. DOI: 10.5194/acp-14-10989-2014.

Wang, Y., J. H. Jiang, and H. Su. 2015b. Atmospheric responses to the redistribution of anthropogenic aerosols. *Journal Of Geophysical Research-Atmospheres* 120(18):9625-9641. DOI: 10.1002/2015JD023665.

Wang, Y., M. H. Wang, R. Y. Zhang, S. J. Ghan, Y. Lin, J. X. Hu, B. W. Pan, M. Levy, J. H. Jiang, and M. J. Molina. 2014c. Assessing the effects of anthropogenic aerosols on Pacific storm track using a multiscale global climate model. *Proceedings of the National Academy of Sciences of the United States of America* 111(19):6894-6899. DOI: 10.1073/pnas.1403364111.

Waring, M. S., and J. R. Wells. 2015. Volatile organic compound conversion by ozone, hydroxyl radicals, and nitrate radicals in residential indoor air: Magnitudes and impacts of oxidant sources. *Atmospheric Environment* 106:382-391. DOI: 10.1016/j.atmosenv.2014.06.062.

Warneke, C., J. A. de Gouw, L. Del Negro, J. Brioude, S. McKeen, H. Stark, W. C. Kuster, P. D. Goldan, M. Trainer, F. C. Fehsenfeld, C. Wiedinmyer, A. B. Guenther, A. Hansel, A. Wisthaler, E. Atlas, J. S. Holloway, T. B. Ryerson, J. Peischl, L. G. Huey, and A. T. C. Hanks. 2010. Biogenic emission measurement and inventories determination of biogenic emissions in the eastern United States and Texas and comparison with biogenic emission inventories. *Journal Of Geophysical Research-Atmospheres* 115. DOI: 10.1029/2009JD012445.

Warneke, C., F. Geiger, P. M. Edwards, W. Dube, G. Petron, J. Kofler, A. Zahn, S. S. Brown, M. Graus, J. B. Gilman, B. M. Lerner, J. Peischl, T. B. Ryerson, J. A. de Gouw, and J. M. Roberts. 2014. Volatile organic compound emissions from the oil and natural gas industry in the Uintah Basin, Utah: oil and gas well pad emissions compared to ambient air composition. *Atmospheric Chemistry and Physics* 14(20):10977-10988. DOI: 10.5194/acp-14-10977-2014.

Webster, P. J., G. J. Holland, J. A. Curry, and H.-R. Chang. 2005. Changes in Tropical Cyclone Number, Duration, and Intensity in a Warming Environment. *Science* 309(5742):1844-1846. DOI: 10.1126/science.1116448.

Welz, O., J. D. Savee, D. L. Osborn, S. S. Vasu, C. J. Percival, D. E. Shallcross, and C. A. Taatjes. 2012. Direct Kinetic Measurements of Criegee Intermediate (CH_2OO) Formed by Reaction of CH_2I with O_2. *Science* 335(6065):204-207. DOI: 10.1126/science.1213229.

Wennberg, P. O., R. C. Cohen, R. M. Stimpfle, J. P. Koplow, J. G. Anderson, R. J. Salawitch, D. W. Fahey, E. L. Woodbridge, E. R. Keim, R. S. Gao, C. R. Webster, R. D. May, D. W. Toohey, L. M. Avallone, M. H. Proffitt, M. Loewenstein, J. R. Podolske, K. R. Chan, and S. C. Wofsy. 1994. Removal of Stratospheric O_3 by Radicals: In Situ Measurements of OH, HO_2, NO, NO_2, ClO, and BrO. *Science* 266(5184):398-404.

Werrell, C. E., and F. Femia. 2013. *The Arab Spring and Climate Change: A Climate and Security Correlations Series*. Washington, DC: Center for Climate & Security.

Weschler, C. J. 2000. Ozone in indoor environments: Concentration and chemistry. *Indoor Air-International Journal of Indoor Air Quality and Climate* 10(4):269-288. DOI: 10.1034/j.1600-0668.2000.010004269.x.

Weschler, C. J., and H. C. Shields. 1997. Potential reactions among indoor pollutants. *Atmospheric Environment* 31(21):3487-3495. DOI: 10.1016/S1352-2310(97)00219-7.

West, J. J., A. M. Fiore, and L. W. Horowitz. 2012. Scenarios of methane emission reductions to 2030: abatement costs and co-benefits to ozone air quality and human mortality. *Climatic Change* 114(3-4):441-461. DOI: 10.1007/s10584-012-0426-4.

Whalley, L. K., P. M. Edwards, K. L. Furneaux, A. Goddard, T. Ingham, M. J. Evans, D. Stone, J. R. Hopkins, C. E. Jones, A. Karunaharan, J. D. Lee, A. C. Lewis, P. S. Monks, S. J. Moller, and D. E. Heard. 2011. Quantifying the magnitude of a missing hydroxyl radical source in a tropical rainforest. *Atmospheric Chemistry and Physics* 11(14):7223-7233. DOI: 10.5194/acp-11-7223-2011.

Whalley, L. K., M. A. Blitz, M. Desservettaz, P. W. Seakins, and D. E. Heard. 2013. Reporting the sensitivity of laser-induced fluorescence instruments used for HO_2 detection to an interference from RO_2 radicals and introducing a novel approach that enables HO_2 and certain RO_2 types to be selectively measured. *Atmospheric Measurement Techniques* 6(12):3425-3440. DOI: 10.5194/amt-6-3425-2013.

WHO (World Health Organization). 2005. Effects of air pollution on children's health and development-A review of the evidence. Bonn, Germany, E. C. f. E. a. H. Special Programme on Health and the Environment.

WHO. 2013 *Review of evidence on health aspects of air pollution – REVIHAAP Project*. Copenhagen, Denmark: World Health Organization Regional Office for Europe.

WHO. 2014. 7 million premature deaths annually linked to air pollution. Available at http://www.who.int/mediacentre/news/releases/2014/air-pollution/en/, accessed February.

Wiedinmyer, C., S. K. Akagi, R. J. Yokelson, L. K. Emmons, J. A. Al-Saadi, J. J. Orlando, and A. J. Soja. 2011. The Fire INventory from NCAR (FINN): a high resolution global model to estimate the emissions from open burning. *Geoscientific Model Development* 4(3):625-641. DOI: 10.5194/gmd-4-625-2011.

Wild, O., J. K. Sundet, M. J. Prather, I. S. A. Isaksen, H. Akimoto, E. V. Browell, and S. J. Oltmans. 2003. Chemical transport model ozone simulations for spring 2001 over the western Pacific: Comparisons with TRACE-P lidar, ozonesondes, and Total Ozone Mapping Spectrometer columns. *Journal Of Geophysical Research-Atmospheres* 108(D21). DOI: 10.1029/2002jd003283.

Wild, R. J., P. M. Edwards, T. S. Bates, R. C. Cohen, J. A. de Gouw, W. P. Dube, J. B. Gilman, J. Holloway, J. Kercher, A. R. Koss, L. Lee, B. M. Lerner, R. McLaren, P. K. Quinn, J. M. Roberts, J. Stutz, J. A. Thornton, P. R. Veres, C. Warneke, E. Williams, C. J. Young, B. Yuan, K. J. Zarzana, and S. S. Brown. 2016. Reactive nitrogen partitioning and its relationship to winter ozone events in Utah. *Atmospheric Chemistry and Physics* 16(2):573-583. DOI: 10.5194/acp-16-573-2016.

Wilson, D. W., H. H. Aung, M. W. Lame, L. Plummer, K. E. Pinkerton, W. Ham, M. Kleeman, J. W. Norris, and F. Tablin. 2010. Exposure of mice to concentrated ambient particulate matter results in platelet and systemic cytokine activation. *Inhalation Toxicology* 22(4):267-276. DOI: 10.3109/08958370903278069.

Wilson, T. W., L. A. Ladino, P. A. Alpert, M. N. Breckels, I. M. Brooks, J. Browse, S. M. Burrows, K. S. Carslaw, J. A. Huffman, C. Judd, W. P. Kilthau, R. H. Mason, G. McFiggans, L. A. Miller, J. J. Najera, E. Polishchuk, S. Rae, C. L. Schiller, M. Si, J. V. Temprado, T. F. Whale, J. P. S. Wong, O. Wurl, J. D. Yakobi-Hancock, J. P. D. Abbatt, J. Y. Aller, A. K. Bertram, D. A. Knopf, and B. J. Murray. 2015. A marine biogenic source of atmospheric ice-nucleating particles. *Nature* 525(7568):234-238. DOI: 10.1038/nature14986.

Wisthaler, A., and C. J. Weschler. 2010. Reactions of ozone with human skin lipids: Sources of carbonyls, dicarbonyls, and hydroxycarbonyls in indoor air. *Proceedings of the National Academy of Sciences of the United States of America* 107(15):6568-6575. DOI: 10.1073/pnas.0904498106.

WMO (World Meteorological Organization). 2014. *Scientific Assessment of Ozone Depletion: 2014. Global Ozone Research and Monitoring Project–Report No. 55*. Geneva, Switzerland: WMO.

Womack, A. M., B. J. M. Bohannan, and J. L. Green. 2010. Biodiversity and biogeography of the atmosphere. *Philosophical Transactions of the Royal Society B-Biological Sciences* 365(1558):3645-3653. DOI: 10.1098/rstb.2010.0283.

Woodcock, A. H. 1950. Condensation Nuclei and Precipitation. *Journal of Meteorology* 7(2):161-162. DOI: 10.1175/1520-0469(1950)007<0161:Cnap>2.0.Co;2.

Wooster, M. J., B. Zhukov, and D. Oertel. 2003. Fire radiative energy for quantitative study of biomass burning: derivation from the BIRD experimental satellite and comparison to MODIS fire products. *Remote Sensing of Environment* 86(1):83-107. DOI: 10.1016/s0034-4257(03)00070-1.

Wu, S. L., L. J. Mickley, E. M. Leibensperger, D. J. Jacob, D. Rind, and D. G. Streets. 2008. Effects of 2000-2050 global change on ozone air quality in the United States. *Journal Of Geophysical Research-Atmospheres* 113(D6). DOI: 10.1029/2007jd008917.

Xue, H. W., and G. Feingold. 2006. Large-eddy simulations of trade wind cumuli: Investigation of aerosol indirect effects. *Journal of the Atmospheric Sciences* 63(6):1605-1622. DOI: 10.1175/Jas3706.1.

Yasmeen, F., N. Sauret, J. F. Gal, P. C. Maria, L. Massi, W. Maenhaut, and M. Claeys. 2010. Characterization of oligomers from methylglyoxal under dark conditions: a pathway to produce secondary organic aerosol through cloud processing during nighttime. *Atmospheric Chemistry and Physics* 10(8):3803-3812.

Yasunari, T. J., R. D. Koster, W. K. M. Lau, and K. M. Kim. 2015. Impact of snow darkening via dust, black carbon, and organic carbon on boreal spring climate in the Earth system. *Journal Of Geophysical Research-Atmospheres* 120(11):5485-5503. DOI: 10.1002/2014JD022977.

Yin, J. H. 2005. A consistent poleward shift of the storm tracks in simulations of 21st century climate. *Geophysical Research Letters* 32(18). DOI: 10.1029/2005gl023684.

Yoshino, A., Y. Sadanaga, K. Watanabe, S. Kato, Y. Miyakawa, J. Matsumoto, and Y. Kajii. 2006. Measurement of total OH reactivity by laser-induced pump and probe technique - comprehensive observations in the urban atmosphere of Tokyo. *Atmospheric Environment* 40(40):7869-7881. DOI: 10.1016/j.atmosenv.2006.07.023.

You, Y., L. Renbaum-Wolff, M. Carreras-Sospedra, S. J. Hanna, N. Hiranuma, S. Kamal, M. L. Smith, X. L. Zhang, R. J. Weber, J. E. Shilling, D. Dabdub, S. T. Martin, and A. K. Bertram. 2012. Images reveal that atmospheric particles can undergo liquid-liquid phase separations. *Proceedings of the National Academy of Sciences of the United States of America* 109(33):13188-13193. DOI: 10.1073/pnas.1206414109.

You, Y., M. L. Smith, M. J. Song, S. T. Martin, and A. K. Bertram. 2014. Liquid-liquid phase separation in atmospherically relevant particles consisting of organic species and inorganic salts. *International Reviews in Physical Chemistry* 33(1):43-77. DOI: 10.1080/0144235X.2014.890786.

Young, C. J., R. A. Washenfelder, P. M. Edwards, D. D. Parrish, J. B. Gilman, W. C. Kuster, L. H. Mielke, H. D. Osthoff, C. Tsai, O. Pikelnaya, J. Stutz, P. R. Veres, J. M. Roberts, S. Griffith, S. Dusanter, P. S. Stevens, J. Flynn, N. Grossberg, B. Lefer, J. S. Holloway, J. Peischl, T. B. Ryerson, E. L. Atlas, D. R. Blake, and S. S. Brown. 2014. Chlorine as a primary radical: evaluation of methods to understand its role in initiation of oxidative cycles. *Atmospheric Chemistry and Physics* 14(7):3427-3440. DOI: 10.5194/acp-14-3427-2014.

Zannoni, N., V. Gros, M. Lanza, R. Sarda, B. Bonsang, C. Kalogridis, S. Preunkert, M. Legrand, C. Jambert, C. Boissard, and J. Lathiere1. 2015. OH reactivity and concentrations of Biogenic Volatile Organic Compounds in a Mediterranean forest of downy oak trees. *Atmospheric Chemistry and Physics Discussions* 15:22017-22095. DOI: 10.5194/acpd-15-22047-2015.

Zelenyuk, A., J. Yang, E. Choi, and D. Imre. 2009. SPLAT II: An Aircraft Compatible, Ultra-Sensitive, High Precision Instrument for In-Situ Characterization of the Size and Composition of Fine and Ultrafine Particles. *Aerosol Science and Technology* 43(5):411-424. DOI: 10.1080/02786820802709243.

Zhang, R. Y., G. H. Li, J. W. Fan, D. L. Wu, and M. J. Molina. 2007. Intensification of Pacific storm track linked to Asian pollution. *Proceedings of the National Academy of Sciences of the United States of America* 104(13):5295-5299. DOI: 10.1073/pnas.0700618104.

Zhang, R., A. Khalizov, L. Wang, M. Hu, and W. Xu. 2011. Nucleation and Growth of Nanoparticles in the Atmosphere. *Chemical Reviews* 112(3):1957-2011. DOI: 10.1021/cr2001756.

Zhang, R., G. Wang, S. Guo, M. L. Zamora, Q. Ying, Y. Lin, W. Wang, M. Hu, and Y. Wang. 2015a. Formation of Urban Fine Particulate Matter. *Chemical Reviews* 115(10):3803-3855. DOI: 10.1021/acs.chemrev.5b00067.

Zhang, R. Y., G. H. Wang, S. Guo, M. L. Zarnora, Q. Ying, Y. Lin, W. G. Wang, M. Hu, and Y. Wang. 2015b. Formation of Urban Fine Particulate Matter. *Chemical Reviews* 115(10):3803-3855. DOI: 10.1021/acs.chemrev.5b00067.

Zhang, Y. P., B. J. Williams, A. H. Goldstein, K. Docherty, I. M. Ulbrich, and J. L. Jimenez. 2014. A Technique for Rapid Gas Chromatography Analysis Applied to Ambient Organic Aerosol Measurements from the Thermal Desorption Aerosol Gas Chromatograph (TAG). *Aerosol Science and Technology* 48(11):1166-1182. DOI: 10.1080/02786826.2014.967832.

Zhou, X. L., H. L. Gao, Y. He, G. Huang, S. B. Bertman, K. Civerolo, and J. Schwab. 2003. Nitric acid photolysis on surfaces in low-NOx environments: Significant atmospheric implications. *Geophysical Research Letters* 30(23). DOI: 10.1029/2003gl018620.

Zhu, C., B. Xiang, L. Zhu, and R. Cole. 2008. Determination of absorption cross sections of surface-adsorbed HNO3 in the 290-330 nm region by Brewster angle cavity ring-down spectroscopy. *Chemical Physics Letters* 458(373-377).

Zhu, L., J. M. Nicovich, and P. H. Wine. 2005. Kinetics studies of aqueous phase reactions of Cl atoms and Cl-2(-) radicals with organic sulfur compounds of atmospheric interest. *Journal of Physical Chemistry A* 109(17):3903-3911. DOI: 10.1021/jp044306u.

Zielinski, S. 2016. The Size of the California Methane Leak Isn't the Scariest Part of the Story. *Smithsonian*, February 26, 2016. Available at http://www.smithsonianmag.com/science-nature/size-california-methane-leak-isnt-scariest-part-story-180958234/#qUus3MVA6q2uvQ59.99, accessed April 15, 2016.

Appendix A—Committee Biographies

Dr. Robert A. Duce (Co-Chair)
Texas A&M University, College Station
Dr. Robert A. Duce is University Distinguished Professor Emeritus of Oceanography and Atmospheric Sciences and retired Dean of the College of Geosciences at Texas A&M University. He was also Dean of the Graduate School of Oceanography, University of Rhode Island. His research focuses on atmospheric and marine chemistry, including the global cycling of trace elements, and he was awarded the Rosenstiel Award in 1990. He was recently chair of the National Academies of Sciences, Engineering, and Medicine's Ocean Studies Board, served on the Board on Atmospheric Sciences and Climate and chaired or co-chaired several National Research Council committees. Dr. Duce is a Fellow of the American Geophysical Union, American Meteorological Society, and American Association for the Advancement of Science, International Union of Geodesy and Geophysics, and the Oceanography Society. He is past president of the Oceanography Society, the International Association of Meteorology and Atmospheric Sciences, the International Commission on Atmospheric Chemistry and Global Pollution, and the International Council for Science Scientific Committee on Oceanic Research. He is also past chair and current member of the United Nations Group of Experts on the Scientific Aspects of Marine Environmental Protection, and he served on the National Sea Grant Advisory Board. He earned his Ph.D. in Inorganic and Nuclear Chemistry from the Massachusetts Institute of Technology in 1964.

Dr. Barbara J. Finlayson-Pitts (Co-Chair)
University of California, Irvine
Dr. Barbara J. Finlayson-Pitts is a Professor of Chemistry and Distinguished Professor at the University of California, Irvine where she co-directs AirUCI, a collaboration of about two dozen faculty working on different facets of atmospheric chemistry and its impacts. She is a Fellow of American Geophysical Union, American Association for the Advancement of Science and the Royal Society of Chemistry, and is a member of the American Academy of Arts & Sciences as well as the National Academy of Sciences. Awards include the American Chemical Society Award for Creative Advances in Environmental Science and Technology, the Tolman Medal of the Southern California ACS and the Haagen-Smit Clean Air Award from the California Air Resources Board. Her research is centered on obtaining a molecular level understanding of reactions that are known, or have the potential, to occur in the atmosphere. Her research includes reactions of airborne sea salt particles and reactions of oxides of nitrogen and organics in thin films on surfaces. Current areas of focus include the atmospheric chemistry of neonicotinoid pesticides, and mechanisms of formation and growth of particles in air and the implications for air quality and climate in the future. She obtained her Ph.D. in Chemistry from the University of California, Riverside, in 1973.

Dr. Tami Bond
University of Illinois, Urbana-Champaign
Dr. Tami Bond is the Nathan M. Newmark Distinguished Professor at the University of Illinois at Urbana-Champaign in Civil and Environmental Engineering, and an Affiliate Professor in

Atmospheric Sciences. Dr. Bond's research addresses the aerosol chemistry, physics, and optics that govern the environmental impacts of particles from combustion. Her work includes laboratory studies of aerosol behavior, field measurements of emissions from small combustion sources, development of global emission inventories, and future emission projections. Dr. Bond is a Fellow of American Geophysical Union and a member of the American Association for Aerosol Research and has authored or co-authored more than 60 scientific papers. She earned her Ph.D. in Atmospheric Sciences, Civil Engineering and Mechanical Engineering at the University of Washington. Dr. Bond is a 2014 John D. and Catherine T. MacArthur Fellow.

Dr. William H. Brune
Pennsylvania State University
Dr. William H. Brune is a Distinguished Professor of Meteorology at Pennsylvania State University. His research interests include atmospheric photochemistry from Earth's surface to the stratosphere; atmospheric aerosol particle formation and aging; uncertainty and sensitivity analysis for atmospheric chemistry models and measurements; and new measurement strategies for atmospheric oxidation processes. He received his Ph.D. from Johns Hopkins University.

Dr. Annmarie Carlton
Rutgers, The State University of New Jersey
Dr. Annmarie Carlton is an Associate Professor at Rutgers University, The State University of New Jersey. Her research interests include atmospheric modeling for air quality and climate with emphasis on atmospheric aqueous chemistry; formation of secondary organic aerosol through cloud processing; aerosol-cloud interactions; biogenic and anthropogenic influences on climate and air quality; and atmospheric processing of pollution. Dr. Carlton received the 2015–2016 Board of Trustees Award for Excellence in Research and the 2009 Distinguished Alumnus for early career distinction from Rutgers. Dr. Carlton worked for the U.S. Environmental Protection Agency in New York, N,Y and Research Triangle Park, NC. She received her Ph.D. from Rutgers University, The State University of New Jersey.

Dr. Allen H. Goldstein
University of California, Berkeley
Dr. Allen H. Goldstein is a Professor in the Department of Civil and Environmental Engineering and in the Department of Environmental Science, Policy, and Management, at the University of California, Berkeley where he served as department chair from 2007–2010. He is currently co-Chair of the International Global Atmospheric Chemistry Program (IGAC). His research program encompasses anthropogenic air pollution, biosphere-atmosphere exchange of radiatively and chemically active trace gases, and development and application of novel instrumentation to investigate the organic chemistry of earth's atmosphere. He engages in field measurement campaigns, controlled laboratory experiments, and modeling activities covering indoor, urban, rural, regional, intercontinental, and global scale studies of ozone, aerosols, and their gas phase precursors. Dr. Goldstein was elected a fellow of the American Geophysical Union, selected as a Miller Foundation Researcher Professor, and a Fulbright Senior Scholar in Australia. He has published more than 280 peer-reviewed scientific papers and holds a patent for On-Line Gas Chromatographic Analysis of Airborne Particles. He earned his B.A. and B.S. degrees from the University of California, Santa Cruz, in politics and chemistry, and his Ph.D. in Chemistry from Harvard University in 1994.

Dr. Colette Heald
Massachusetts Institute of Technology
Colette L. Heald is an Associate Professor in the Departments of Civil and Environment Engineering & Earth and Planetary Sciences at the Massachusetts Institute of Technology (MIT). Her primary research interests are exploring biosphere-atmosphere exchange processes relevant to atmospheric chemistry, and the lifecycle and impacts of atmospheric aerosols. Her research focuses on integrating global modeling tools with observations. Dr. Heald has authored or co-authored more than 70 peer-reviewed scientific articles. She is a recipient of the Macelwane Medal and is a Fellow of the American Geophysical Union. She received her undergraduate degree in Engineering Physics from Queen's University in Canada in 2000, and her Ph.D. in Earth and Planetary Science from Harvard University in 2005.

Dr. Scott C. Herndon
Aerodyne Research, Inc.
Dr. Scott C. Herndon is a Physical Chemist and Principal Scientist in the Center for Atmospheric and Environmental Chemistry at Aerodyne Research, Inc. Since joining Aerodyne in 1999, his research interests have focused on the development and utilization of laboratory and field trace gas and fine particle instrumentation, together with modeling studies, to characterize and elucidate atmospheric processes relevant to stratospheric ozone depletion, urban and regional air quality and climate change. He has led more than 20 field measurement campaigns to characterize and quantify air pollutant emission sources and map ambient pollution concentrations using suites of advanced, real-time spectroscopic and mass spectrometric instrumentation deployed on the Aerodyne Mobile Laboratory and on a range of research aircraft and ships. Most recently Dr. Herndon has developed an improved dual tracer release ratio method to quantify methane emissions from oil and gas production and transmission facilities and other sources in the US and Mexico. He is the author or co-author of more than 50 archival publications addressing atmospheric science and physical chemistry issues. He earned his Ph.D. in Physical Chemistry at the University of Colorado.

Dr. Dylan B. Jones
University of Toronto
Dr. Dylan Jones is a Professor in the Department of Physics at the University of Toronto. His research is focused on integrating measurements of atmospheric composition with global three-dimensional models of chemistry and transport to develop a better understanding of how pollution influences the chemical and dynamical state of the atmosphere. His research also uses chemical data assimilation and inverse modeling techniques to quantify surface fluxes of trace constituents that are important for air quality and the global carbon cycle. Dr. Jones is a member of the GEOS (Goddard Earth Observing System)–Chem model Steering Committee. He held a Tier II Canada Research Chair from 2004-2014 and received an Ontario Early Researcher Award in 2007. He received his B.A. in Physics and Astronomy and Astrophysics from Harvard University in 1990, his M.Sc. in Applied Physics from Harvard University in 1994, and his Ph.D. in Earth and Planetary Sciences from Harvard University in 1998.

Dr. Athanasios Nenes
Georgia Institute of Technology
Dr. Athanasios Nenes is a Professor, Georgia Power Scholar, Cullen-Peck Fellow and Johnson Faculty Fellow at the Georgia Institute of Technology. He is also an affiliate scientist at the Foundation for Research and Technology Hellas, and the National Observatory of Athens, Greece. Through a combination of theory, modeling and observations, his research focuses on aerosol thermodynamics, aerosol-cloud interactions and their impacts on the hydrological cycle and climate; biogeochemical cycling of nutrients, and aerosol impacts on marine productivity and the carbon cycle. He is an author on more than 210 peer-reviewed manuscripts, and is developer of the ISORROPIA aerosol thermodynamic equilibrium codes and is co-inventor of the Continuous Flow Streamwise Thermal Gradient CCN (Cloud Condensation Nuclei) chamber. He served as Secretary of the Atmospheric Science Section of the American Geophysical Union, the Board of Directors of the American Association for Aerosol Research, and on the Committee on Nucleation and Atmospheric Aerosols. He is recipient of the Atmospheric Sciences Section Ascent Award, American Geophysical Union, Kenneth T. Whitby and Sheldon K Friedlander Awards from the American Association for Aerosol Research, and the Henry G. Houghton Award from the American Meteorological Society. He received his Ph.D. in Chemical Engineering from the California Institute of Technology.

Dr. Kimberly A. Prather
University of California, San Diego
Kimberly Prather is the Distinguished Chair in Atmospheric Chemistry and holds a joint appointment in the Department of Chemistry and Biochemistry and Scripps Institution of Oceanography at University of California, San Diego since 2001. Early in her career, she developed novel on-line single particle mass spectrometers that are used worldwide to determine the sources of aerosols in air pollution. Her group uses these instruments to better understand the sources of air pollution and understand the role of aerosols in climate change, including aircraft-based measurements of aerosol impacts on cloud properties and precipitation. She has published more than 180 peer-reviewed scientific papers and holds a number of patents on on line mass spectrometry methods. She served two terms on the National Academy of Sciences, Engineering, and Medicine’s Board for Atmospheric Science and Climate. She is an elected fellow of the American Academy of Arts and Science, American Geophysical Union, and Association for the Advancement of Arts and Sciences. She received her Ph.D. in Physical Chemistry from University of California, Davis in 1990.

Dr. Michael J. Prather
University of California, Irvine
Dr. Michael J. Prather is Professor of Earth System Science at the University of California, Irvine. His research focuses on: the simulation of the physical, chemical, and biological processes that determine atmospheric composition; the development of detailed numerical models of photochemistry and atmospheric radiation; and overal testing of global chemical transport models that describe ozone and other trace gases. Post-Ph.D., Dr. Prather was a research fellow at Harvard University and then a scientist at the Goddard Institute for Space Studies, including also managing NASA HQ (National Aeronautics and Space Administration Head Quarters) programs on upper atmosphere and aviation impacts. A fellow of the AGU (American Geophysical Union), AAAS (American Association for the Advancement of

Science), and a member of the Norwegian Academy of Science and Letters, he served from 1997 through 2001 as Editor-in-Chief of Geophysical Research Letters. He received a B.A. in mathematics from Yale University, a B.A. in physics from the University of Oxford, and a Ph.D. in astronomy and astrophysics from Yale University. Dr. Prather has participated in key United Nations environmental efforts, including the international ozone assessments (1985, 1988, 1989, 1991, 1994, 2010, 2014) and climate assessments IPCC (Intergovernmental Panel on Climate Change): 1992, 1995, 1999, 2001, 2007, 2013, 2014). Dr. Prather has served on numerous National Research Council committees, most recently as a member of the Assessment of NASA's (National Aeronautics and Space Administration) Earth Science Programs. He also previously served on the Committee on Methods for Estimating Greenhouse Gas Emissions, the Panel on Climate Variability and Change of the 2007 decadal survey on Earth science and applications from space, and the Committee for Review of the U.S. Climate Change Science Program Strategic Plan.

Dr. Allison Steiner
University of Michigan, Ann Arbor
Dr. Allison Steiner is an Associate Professor at the University of Michigan, Ann Arbor. Her research interests focus on understanding biosphere-atmosphere interactions across a range of spatial and temporal scales, including improved understanding of primary emissions and their fate in the atmosphere and changes in atmospheric chemistry on climatological time scales. She is the recipient of the University of Michigan Henry Russel Award in 2013 and the American Geophysical Union Atmospheric Sciences section Ascent Award in 2015. She received her B.S. in Chemical Engineering from Johns Hopkins University and her Ph.D. in Atmospheric Science from Georgia Institute of Technology.

Dr. Christine Wiedinmyer
National Center for Atmospheric Research
Dr. Christine Wiedinmyer is a Scientist III in the Atmospheric Chemistry Observations & Modeling Laboratory of the National Center for Atmospheric Research. Dr. Wiedinmyer's research emphasizes the identification and quantification of various emission sources and determining the transport and fate of pollutants in the atmosphere using models and observations. Her research interests include evaluating ways in which climate, technology, and policy impact air quality. Dr. Wiedinmyer was the 2014 Walter Orr Roberts Lecturer in Interdisciplinary Sciences from the American Meteorological Society. She received her B.S.E. in Chemical Engineering from Tulane University and her M.S. and Ph.D. in Chemical Engineering from The University of Texas at Austin.

Dr. Lei Zhu
New York State Department of Health
Dr. Lei Zhu is a Research Scientist at the Wadsworth Center, New York State Department of Health, and a professor in the Department of Environmental Health Sciences at University at Albany. Dr. Zhu's research program has been designed to investigate and reveal what controls the atmosphere's energy balance and how chemical reactions impact composition, pollutant and oxidant formation in Earth's environment. Her research interests include kinetics and photochemistry of homogeneous and heterogeneous atmospheric reactions, atmospheric application of cavity ring-down spectroscopy and its novel variants, and atmospheric application of time-resolved FT-IR (Fourier Transform Infrared Spectroscopy). Dr. Zhu received her Ph.D. in Physical Chemistry from Columbia University in 1991. She was an Enrico Fermi Scholar at Argonne National Laboratory from 1991 to 1993.

Appendix B—Community Input Gathering

The Committee obtained a broad swath of input, guidance, and suggestions from the atmospheric chemistry community on important potential research topic and programmatic priorities for the coming decade. Six public town hall meetings were held throughout 2015 in several locations across the United States: Boulder, CO (March), Boston, MA (April), Washington, DC (April), Irvine, CA (May), Atlanta, GA (June), and Waterville Valley, NH (August). In addition, an online "virtual town hall"[36] was established to solicit input from those who could not attend an in-person meeting. In all, more than 250 atmospheric chemistry research community members provided input to this study, either at one of the in-person meetings or via the website.

Participants were asked to provide feedback for the following five questions:

1. What are the important areas of scientific research that could transform the understanding of atmospheric chemistry over the coming decade?
2. What research linkages of atmospheric chemistry with other disciplines as well as with national or international research portfolios could produce transformational science over the next decade?
3. How can advances in atmospheric chemistry, either alone or in tandem with other disciplines, play a critical role in addressing major societal challenges over the next decade?
4. What infrastructure, new approaches, or other community capabilities, need to be maintained or developed to support advances in these topics? (You might consider shared models, facilities, platforms, instrumentation, or computing, but are not limited to these.)
5. Do you have other comments pertinent to the Committee's Statement of Task?

Input received by the Committee to these five questions formed the basis for the Committee's discussions of future priorities in atmospheric chemistry research. The Committee sifted through this input, categorized it, and ultimately developed our own list of priority areas for research, as discussed in Chapter 5.1.

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[36] The Future of Atmospheric Chemistry Research Virtual Town Hall: http://nas-sites.org/atmchem/.

Appendix C—Analysis of Funding Trends for Atmospheric Chemistry Research

In order to understand the trends that are currently occurring in the field of atmospheric chemistry and the research emphases of different federal departments, the Committee requested information from five government agencies that support atmospheric chemistry research: the National Science Foundation (NSF), the National Oceanic and Atmospheric Administration (NOAA), the Environmental Protection Agency (EPA), the Department of Energy (DOE), and the National Aeronautics and Space Administration (NASA). The recent trends in spending by these major atmospheric chemistry research funding agencies are shown in Figure 6.2. Figure C.1 further examines these trends for NSF research spending in terms of the Atmospheric Chemistry (ATC) program, the National Center for Atmospheric Research's (NCAR's) Atmospheric Chemistry Observations and Modeling (ACOM) Laboratory, and "other" NSF program and deployment funding (see figure legend for details) related to atmospheric chemistry research.

In addition, the Committee examined trends in the awards granted in atmospheric chemistry research by NSF. For these NSF awards, the total number of atmospheric chemistry research projects funded, the combined award amount, and the median award amount (Figure C.2) have not changed substantially over the past two decades. Year to year variability makes it challenging to discern any trends from this spending, but overall, total funding spent on Atmospheric Chemistry and the median budget per project (considering inflation) have not increased robustly in this time period.

The Committee also examined how support from the various agencies is divided among types of research approaches, including laboratory, field, and modeling studies. It is noted that there is often overlap of types of research within a given study, but these classifications indicate the general trends in the field as a whole. From 2006–2014 the NSF Atmospheric Chemistry program has fairly consistently spent roughly half of its budget on field projects, 25 percent on lab studies, 15 percent on modeling, 5 percent on instrument development, and 5 percent on "other" projects (see Figure 6.3). Between 2008–2015 the EPA Science to Achieve Results (STAR) program for atmospheric science research (not including graduate fellowships) has devoted about 10 percent of its funds on laboratory research (see Figure C.3a), with the rest split about evenly between field and modeling research. Note that while total STAR funding appears to have decreased over this time period, Atmospheric Science program funding has remained fairly constant, despite the drop in 2008 (modeling studies were funded less that year than in 2007 or 2009 to accommodate health priorities). Overall, NOAA Oceanic and Atmospheric Research (OAR) funding for research has modestly increased from 2005–2015. On average, OAR spends about 35 percent of its funds on field studies, 20 percent on modeling work, which both appear to have risen in recent years, 5 percent on laboratory research, and the remaining 40 percent on facilities/infrastructure and "other" research (see Figure C.3b). While only a partial breakdown of DOE Climate and Environmental Sciences Division (CESD) funding figures was available for the 2005–2015 period, it is estimated that experimental lab work is relatively constant at approximately 25 percent of the budget, modeling funds also account for about 25 percent, and field observations comprise the remaining 50 percent of CESD funds.[37] While we review only a few agencies, overall, most fluctuations in funding appear to be temporary and do not likely indicate substantial changes in research priorities within atmospheric chemistry.

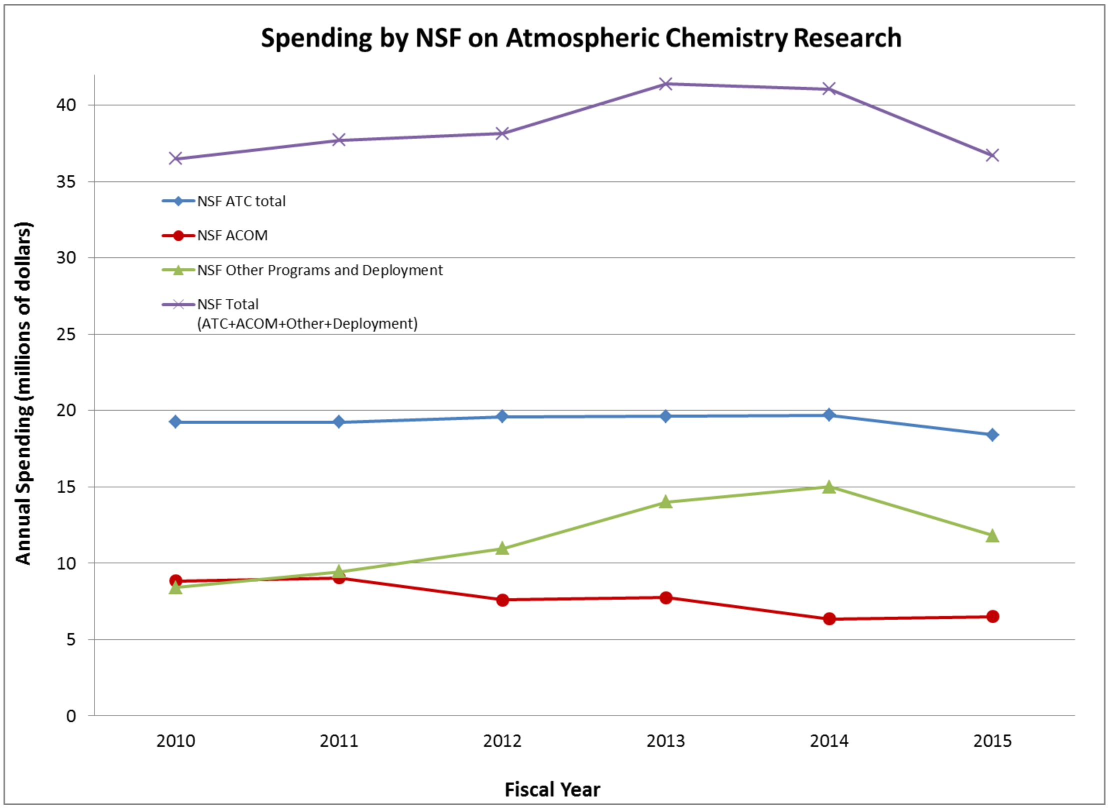

FIGURE C.1 Annual funding in millions of U.S. dollars for programs within NSF that support atmospheric chemistry research, including the Atmospheric Chemistry (ATC) program (blue line), the National Center for Atmospheric Research's (NCAR's) Atmospheric Chemistry Observations and Modeling (ACOM) Laboratory (red line), "other" NSF program and deployment (green line), and the sum of these categories (purple line). "Other" NSF program and deployment funding comes from the NSF Deployment Pool[38] and funding for atmospheric chemistry research from other NSF directorates, including the Directorate for Mathematical & Physical Sciences (Environmental Chemical Sciences program, and Division of Chemistry through the Centers for Chemical Innovation program); the Directorate for Engineering (Environmental Engineering and Environmental Sustainability programs); and the Directorate for Geosciences (Chemical Oceanography, Arctic Natural Sciences, and Antarctic Ocean and Atmospheric Sciences programs).

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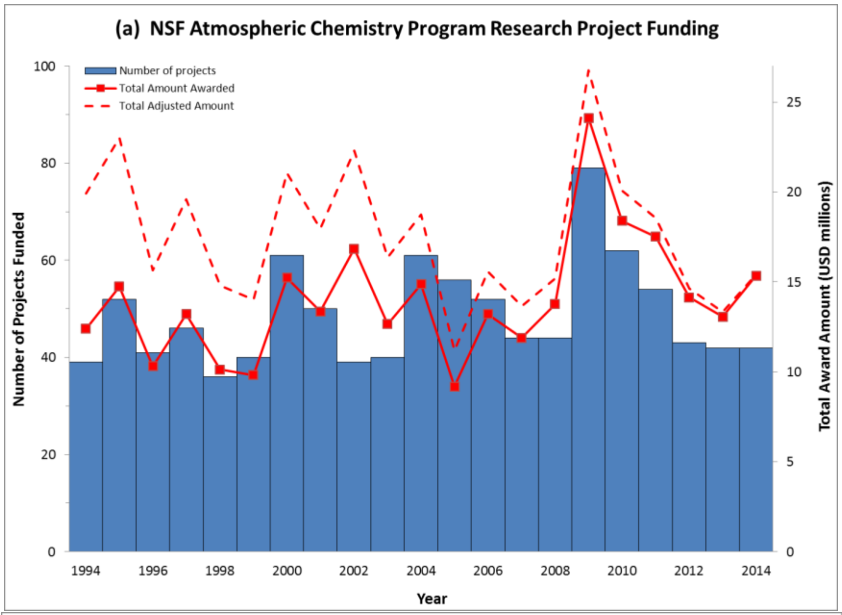
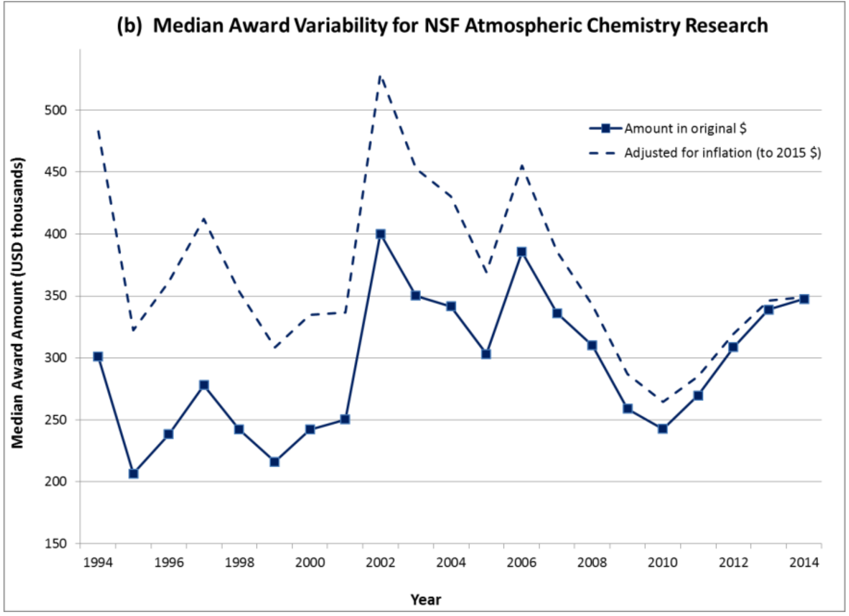
[37] Personal communication, Ashley Williamson, December 2015.
[38] https://www.eol.ucar.edu/facilities-instruments.



(a) NSF Atmospheric Chemistry Program Research Project Funding



(b) Median Award Variability for NSF Atmospheric Chemistry Research

FIGURE C.2 (a) Number of atmospheric chemistry research projects funded (blue bars and the left side axis) and total NSF Atmospheric Chemistry (ATC) program funds awarded (millions of U.S. dollars shown with red squares and adjusted for inflation [in 2015 dollars] shown with red dashed line on the right side axis), and **(b)** thousands of U.S. dollars awarded to atmospheric chemistry researchers between 1994–2014. Values were determined according to the year of the grant's "Date Awarded," and the majority of grants spanned a 3–5-year period. Note the peak in number of projects funded in 2009 corresponds to a one-year increase provided by the Recovery Act stimulus funding. NSF grant data for the years 1994–2014 (awarded through the ATC program) were obtained through the research.gov database of research spending.[39]

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[39] Caveats with research.gov data: "Research Spending and Results displays grant awards active as of FY 1994 and beyond for NSF and active as of FY 2007 and beyond for NASA. Please note information may not be complete for awards that date prior to the Federal Funding Accountability and Transparency Act of 2006 (awards not active as of October 2006 and beyond). It does not provide award information for loans or contracts."

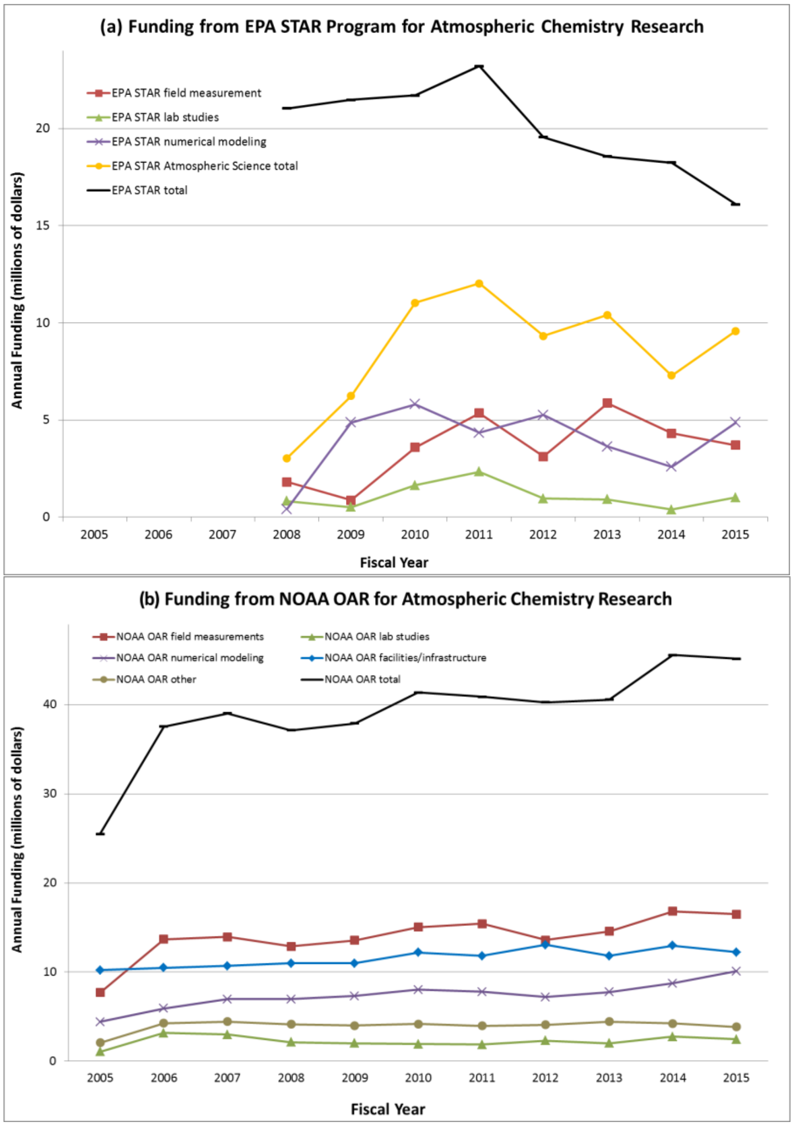

FIGURE C.3 Annual funding in millions of U.S. dollars for the **(a)** EPA Science to Achieve Results (STAR) program total and atmospheric science budget (not including graduate fellowships) and **(b)** NOAA Oceanic and Atmospheric Research (OAR) atmospheric chemistry research that has been allocated to field measurements (red lines), laboratory research (green lines), numerical modeling studies (purple lines), instrument development or facilities and infrastructure (dark blue lines), and "other" projects[40] (brown lines) between fiscal years 2005–2015. EPA Atmospheric Science funding total is shown by a yellow line, and EPA STAR (including disciplines other than atmospheric sciences) and NOAA OAR atmospheric chemistry research total funding over the time period specified are shown by black lines. Aggregate budget and funding information were provided through personal communication with representatives from EPA and NOAA; and data were subject to availability. Note the different scales on graphs.

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[40] For NOAA, "other" does not include facilities or infrastructure support.