This PDF is available from The National Academies Press at http://www.nap.edu/catalog.php?record_id=18683

FROM THE ARCHIVES

# Problems Related to Interplanetary Matter (1961)

Pages
111

Size
6 x 9

ISBN
0309300347

Subcommittee on Nuclear Geophysics; Committee on Nuclear Science; National Research Council

Find Similar Titles

More Information

Visit the National Academies Press online and register for...

- Instant access to free PDF downloads of titles from the
  - NATIONAL ACADEMY OF SCIENCES
  - NATIONAL ACADEMY OF ENGINEERING
  - INSTITUTE OF MEDICINE
  - NATIONAL RESEARCH COUNCIL
- 10% off print titles
- Custom notification of new releases in your field of interest
- Special offers and discounts

Distribution, posting, or copying of this PDF is strictly prohibited without written permission of the National Academies Press. Unless otherwise indicated, all materials in this PDF are copyrighted by the National Academy of Sciences.

To request permission to reprint or otherwise distribute portions of this publication contact our Customer Service Department at 800-624-6242.

Copyright © National Academy of Sciences. All rights reserved.

NATIONAL ACADEMY OF SCIENCES
1863–2013
Celebrating 150 Years of Service to the Nation

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

Nuclear Science Series
Report Number 33

Subcommittee on
Nuclear Geophysics .

# Problems Related to Interplanetary Matter

Proceedings of an Informal Conference

Highland Park, Illinois

June 20-22, 1960

NATIONAL ACADEMY OF SCIENCES
LIBRARY
MAY 3 1961
NATIONAL RESEARCH COUNCIL

Publication 845
National Academy of Sciences—National Research Council
Washington, D. C.
1961

NATIONAL ACADEMIES LIBRARY
16648

$2.00

Copyright © National Academy of Sciences. All rights reserved.

Library of Congress
Catalog Card Number 61-60013

Copyright © National Academy of Sciences. All rights reserved.

# FOREWORD

The fourth conference sponsored by the Subcommittee on Nuclear Geophysics of the Committee on Nuclear Science, National Academy of Sciences—National Research Council, was held at Highland Park, Illinois, June 20-22, 1960. The topic of this conference was "Problems Related to Interplanetary Matter," a theme chosen in response to the increasing interest of geoscientists in problems related to outer space. The subcommittee believed that the participation of scientists of different disciplines in such a meeting would be of benefit to all who are working in the fields of the geosciences. This type of meeting was in line with the general aim of the subcommittee to stimulate interest in problems related to nuclear geophysics.

It has been the policy of the subcommittee to conduct the meetings as informally as possible, in order to maximize the discussion and interchange of ideas. To encourage the freedom of discussion, it was made clear at the outset that direct quotations would be kept to a minimum in the published proceedings of the conference.

A new method for reporting the meeting was used. The proceedings were recorded by two qualified workers in the scientific field under discussion, Robert A. Fish and Gordon Goles of the Enrico Fermi Institute for Nuclear Studies. Subsequently, the written reports were circulated to the various speakers for review and emendation as necessary. The final report thus represents the speakers' final judgment of the content of the papers though not necessarily their style of writing. Questions and responses have in many cases been modified and incorporated into the text for purposes of clarity and continuity. In some cases, the editors have added their own comments; these are enclosed in brackets.

As usual, the valuable assistance and advice of the Division of Physical Sciences of the Academy-Research Council contributed greatly to the success of the conference. Financial support from the United States Air Force is gratefully acknowledged.

Subcommittee on Nuclear Geophysics:
S. Epstein, Chairman

| | |
|---|---|
| L. T. Aldrich | P. M. Hurley |
| J. R. Arnold | M. G. Inghram |
| L. F. Curtiss | A. O. Nier |



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

## PARTICIPANTS

W. M. Alexander
National Aeronautics and
Space Administration
Goddard Space Flight Center
8719 Colesville Road
Silver Spring, Maryland

Edward Anders
Enrico Fermi Institute
University of Chicago
Chicago 37, Illinois

James R. Arnold
University of California
La Jolla, California

Wallace S. Broecker
Lamont Geological Observatory
Palisades, New York

A. G. W. Cameron
California Institute of Technology
Pasadena, California

Samuel Epstein
California Institute of Technology
Pasadena, California

E. L. Fireman
Smithsonian Astrophysical
Observatory
Cambridge, Massachusetts

Robert A. Fish
Enrico Fermi Institute
University of Chicago
Chicago 37, Illinois

Paul W. Gast
Department of Geology
University of Minnesota
Minneapolis 14, Minnesota

Gordon Goles
Enrico Fermi Institute
University of Chicago
Chicago 37, Illinois

Albert R. Hibbs
Jet Propulsion Laboratory
California Institute of Technology
Pasadena, California

Patrick M. Hurley
Department of Geology
Massachusetts Institute of
Technology
Cambridge, Massachusetts

Mark G. Inghram
Department of Physics
University of Chicago
Chicago 37, Illinois

Truman P. Kohman
Department of Chemistry
Carnegie Institute of Technology
Pittsburgh 13, Pennsylvania

A. O. C. Nier
Department of Physics
University of Minnesota
Minneapolis, Minnesota

Stanislaw Olbert
Massachusetts Institute of
Technology
Cambridge, Massachusetts

George W. Reed, Jr.
Argonne National Laboratory
Argonne, Illinois



Copyright © National Academy of Sciences. All rights reserved.

John H. Reynolds
Department of Physics
University of California
Berkeley 4, California

Oliver Schaeffer
Chemistry Department
Brookhaven National Laboratories
Upton, Long Island, New York

Peter Signer
Department of Physics
University of Minnesota
Minneapolis, Minnesota

Heinz Stauffer
University of California
La Jolla, California

Hans Suess
University of California
La Jolla, California

Anthony Turkevich
Enrico Fermi Institute
University of Chicago
Chicago 37, Illinois

J. A. Van Allen
University of Iowa
Iowa City, Iowa

George W. Wetherill
Department of Terrestrial Magnetism
Carnegie Institution of Washington
5241 Broad Branch Road
Washington, D. C.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

CONTENTS





Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683





Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# THE ORIGIN OF THE SOLAR SYSTEM*

A. G. W. Cameron
Mount Wilson and Palomar Observatories**
Pasadena, California

With the growing astronomical evidence that the Galaxy is many billions of years older than the solar system, it becomes necessary to revise our ideas about the processes which enriched the interstellar medium in heavy elements. It is no longer very likely that the heavy elements of the solar system were built in the sort of enormous neutron fluxes which would be expected in supernovae of Type I if the light curves of these supernovae are due to the decay of $Cf^{254}$ (Burbidge, Hoyle, Burbidge, Christy, and Fowler 1956). Between the time of formation of the Galaxy and the formation of the solar system enough of these supernovae would have occurred to produce heavy element concentrations in the interstellar medium at least a thousand times too large to be compatible with today's solar and meteoritic abundances.

As soon as one is willing to accept less powerful neutron sources, a variety of possibilities suggest themselves, so that it becomes difficult to be sure which stellar sources have been mainly responsible for the synthesis of the heavy elements. We start with neutron capture on a slow time scale, in which heavy elements are slowly built up in a capture chain starting with the abundant elements of the iron peak. I now believe that the $Ne^{22}(\alpha, n)Mg^{25}$ reaction is the principal source of these neutrons. Once the heavy elements have been synthesized on a slow time scale, subsequent reactions taking place on intermediate and fast time scales can add neutrons quickly and produce neutron-rich isobars. I believe that a variety of reactions associated with carbon thermonuclear reactions are responsible for producing these neutrons. It seems most likely that the products of the fast time scale will be ejected in Type I supernova explosions, although with very much smaller abundances than in the californium theory.

*This research was supported by the Air Force Office of Scientific Research, ARDC, under contract AF 49(638)-21.

**Permanent address: Atomic Energy of Canada, Ltd., Chalk River, Ontario.



Copyright © National Academy of Sciences. All rights reserved.

Among the nuclei which are synthesized by neutron capture on a fast time scale is $I^{129}$. This has recently become a particularly interesting nuclide because Reynolds (1960a) found an excess amount of its decay product, $Xe^{129}$, in the Richardton meteorite.

At first it appeared that there was one part of excess $Xe^{129}$ per million parts of $I^{127}$. Since there is also about one part of lithium, beryllium, or boron isotope per million oxygen atoms, it seemed possible that the $I^{129}$ might have been produced by spallation by high energy protons from the Sun during the formation of the solar system. However, since the iodine content of Richardton is now found to be a factor ~25 lower than the first estimate (Goles 1960, Reynolds 1960b), this hypothesis becomes difficult to maintain. Instead, we must consider the $I^{129}$ to have survived the interval between the cessation of nucleogenesis in that part of the interstellar medium from which the Sun was formed, and the cooling of the meteorite parent bodies sufficiently to retain xenon. Assuming the galaxy to be $10^{10}$ years older than the solar system, and rate of nucleogenesis 4.6 billion years ago to be half the average rate of the preceding $10^{10}$ years, then the above interval turns out to be $1.0 \times 10^8$ years.

This interval seems surprisingly small for many complex processes to take place. It therefore becomes of interest to estimate the time required for the distribution of fresh radioactivity to the interstellar medium, for the formation of the star cluster of which the Sun was a member, for the contraction of the Sun, and for the formation and cooling of the planets (or at least of the meteorite parent bodies).

The gas and dust in the plane of the galaxy form a disk 200 parsecs thick. This interstellar medium is composed of relatively dense clouds (about 10 hydrogen atoms per $cm^3$) which move through the intercloud medium (of density 20 to 100 times smaller) with velocities of the order of 10 km/sec. The old Population I stars, of which Type I supernovae are probably members, form a layer with a thickness about four times that of the gas and dust. Hence about one Type I supernova in four explodes in the interstellar medium, and about 90 percent of these will be located in the intercloud regions. The products of their explosions will expand to fill a sphere of about 40 parsecs diameter in about 10 million years. To be conservative we will neglect any further diffusion of the products into the interstellar medium.

If the clouds were formed by an isotropic compression of the interstellar medium, the internal magnetic pressure would be very much greater than any likely external pressure, and the clouds would soon expand again. To be stable, the clouds must have formed by compression of gas along the magnetic lines of force. A mechanism which seems to



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

cause this has been suggested by Hoyle and Ireland (1960); if a section of the interstellar magnetic field loops up into the galactic halo, material will fall down the lines of force until it has sufficient kinetic energy to overpower magnetic restraints. The material thus compressed will contain products from supernovae which have exploded as much as $10^9$ years ago, but the probability is fairly high that some of the material is of recent origin. I estimate that, subject to a probable error of about a factor of five, the material collected will have an amount of $I^{129}$ equal to that which one would calculate for a continuous synthesis model with a decay time for uniform mixing of 10 million years. The time required to form the cloud by compression is about 20 million years.

In order that such a cloud can condense further to form stars, the level of ionization in it must fall very low so that the gas becomes decoupled from the magnetic field, and the temperature must become small enough so that Jeans' gravitational instability criterion is satisfied. According to this criterion, twice the thermal energy of the gas cloud plus the potential energy of gravitation must be less than zero. Therefore, for a uniform spherical gas cloud, contraction can take place only for temperatures below

$$T = \frac{M^{2/3}\, n^{1/3}}{30}$$

which means that T must be less than about $7^{\circ}$ K if the cloud has a typical mass M of about 1000 solar masses and a density n of about 10 particles per $cm^3$.

The principal heating mechanisms are collisions with other clouds, photoionization by starlight, and ionization by cosmic rays. The principal cooling mechanisms are electron-ion collisions, collisional excitation of $H_2$ molecules, and molecule-grain collisions. It is likely that cosmic rays will be excluded if the clouds are magnetically isolated from the surrounding medium. Starlight can be excluded if there is a large effective grain density at the surface of the cloud. It may be that collisions with clouds will cause evaporation of some large grains and recondensation of small grains, and the optical activation of these and other small grains due to radiation damage. It is tempting to ascribe to these small grains the quantum mechanical properties suggested by Platt (1960), so that they can absorb radiation of wavelength ~400 times their diameters.

Collisions between clouds will raise the gas kinetic temperature to a few thousand degrees, perhaps sufficient to dissociate $H_2$ molecules. Reassociation will take place by collisions with grains, and hydrogen molecule cooling will quickly lower the temperature to about $40^{\circ}$ K. Further cooling depends on molecule-grain collisions; we can only guess at the rate here because of our almost complete ignorance of the grain



Copyright © National Academy of Sciences. All rights reserved.

properties. Temperatures as low as $7^{\circ}$ K can probably be reached after total cooling times of 20 to 40 million years, but this is a highly uncertain estimate.

However, collisions between clouds occur on the average about every 10 million years. Hence the clouds will be reheated usually before the temperature falls low enough for star formation to take place. Star formation thus requires a statistical fluctuation resulting in a somewhat greater than average interval between cloud collisions. This will add a variable time probably of the order of 30 million years to the $I^{129}$ interval.

When Jeans' instability criterion is satisfied, the cloud will contract at a rate which rapidly becomes a free fall. The reason for this is that cooling remains sufficiently rapid during the contraction, i.e., once Jeans' criterion becomes satisfied it stays well-satisfied. The free fall time is

$$t = 1.11\ (R^3/GM)^{1/2} = 1.63 \times 10^{15}\ n^{-1/2}\ \text{sec},$$

which is about 16 million years for n = 10. At the start, the effective gravity G is less owing to thermal pressures, so the effective free fall time may be taken as about 25 million years. Note that the above formula does not involve the mass of the cloud, so that subunits can condense out of the cloud as soon as they satisfy Jeans' criterion. The final subdivisions will have masses of solar order. The result of these considerations so far is to give a predicted $I^{129}$ interval in the range $0.4$ to $2.2 \times 10^8$ years, with a probable value near $1.1 \times 10^8$ years. This would be decreased for more massive clouds than the one considered, and increased for less massive clouds. But it is already of the correct order of magnitude. We would thus begin to feel uncomfortable if the remaining stages of stellar contraction and planet formation should take a similar time.

The free fall collapse stops when the protosun has a radius of about $3 \times 10^3$ astronomical units. At this point the medium becomes opaque to grain radiation. Further contraction is luminosity controlled, with density in the protosun becoming a smooth function of the radius and with a substantial central condensation taking place. At this point also the requirement of the conservation of angular momentum will make the protosun gravitationally unstable at the equator, so that further contraction must be accompanied by mass loss.

The contraction from $3 \times 10^3$ a.u. to 100 a.u. probably takes less than one million years. At this point a dynamical instability sets in owing to the operation of the following energy-absorbing processes: the dissociation of $H_2$ molecules, the ionization of hydrogen, and the double ionization of helium. A free fall collapse takes place which causes the protosun



Copyright © National Academy of Sciences. All rights reserved.

to shrink from 100 a.u. to about 0.34 a.u. in a period of ~200 years. About half a solar mass is shed in the form of a nebula during this time. The sun will require about another 70 million years to reach the main sequence, but as we shall see this time is not to be added to the $I^{129}$ interval. The important stages in the evolution of the planets will take place while the Sun has a radius of ~0.34 a.u. and a surface temperature of about 410° K.

The nebula probably does not form gaseous "protoplanets" directly, as Kuiper (1951) has suggested, because even protoplanets stable against tidal disruption (Roche criterion) would still suffer thermal disruption (Jeans' criterion) unless the radius of the protoplanets was at least six times greater than the "homogeneous" height of the nebula.

Detailed calculations show that in the portion of the nebula shed between 0.3 and 30 a.u., part of the gas has been heated in excess of 1000° K. This is sufficient to destroy all traces of the original interstellar grains. Cooling is a relatively slow process, and the vaporized gases will recondense on a relatively small number of centers, as in a cloud chamber in very slow expansion. It is to be expected that solid bodies the size of boulders will be formed. These will tend to fall toward the plane of the ecliptic and to accumulate into larger bodies. The terrestrial planets represent a very inefficient collection of the total amount of solid material available.

The time scale for this planet accumulation is probably very small compared to $10^8$ years, and therefore it probably does not add appreciably to the $I^{129}$ interval.

There are three stages in the development of the protosun at which magnetic forces are very important. These are: (a) in the early stages of luminosity-controlled contraction when a unified rate of angular velocity is being produced, (b) the escape of strong magnetic fields from the nebula which is shed during the dynamical collapse, and (c) following the dynamical collapse when an outer solar convection zone can build an external dipole field which can interact with the nebula to brake the Sun's rotation. At each stage an intense acceleration of charged particles may take place, thus producing extensive nuclear spallation throughout the nebula (support for this view is found in the lithium-rich spectrum of T Tauri type protostars--Herbig 1956). It might be possible to produce by spallation some $10^{-6}$ atoms of $Al^{26}$ per Si atom and $10^{-6}$ $Be^{10}$ atoms per oxygen atom. According to the calculations of Fish, Goles, and Anders (1960) these abundances of short-lived radioactivities could have melted the asteroids, which is one of the steps in their model for the synthesis of the meteorites.



Copyright © National Academy of Sciences. All rights reserved.

Arnold: How can we interpret the fact that some meteorites show essentially no excess $Xe^{129}$, even though they may have iodine abundances as high or higher than in Richardton (e.g., Beardsley: Goles 1960, Reynolds 1960b)? The different xenon results could reflect different time scales for the formation of the meteorites, different diffusive properties of the meteorites, or differences in the thermal histories.

Cameron: In my model all the time scales for formation of the meteorite parent bodies are the same; the subsequent history may be different.

Anders: Some calculations by Goles and Fish reveal that the cooling times for melted asteroids containing the complement of $K^{40}$ which would have been present $4.5 \times 10^9$ years ago are already comparable with the Richardton I-Xe age, so that the amount of time elapsed between nucleosynthesis and the formation of solid objects in the solar system is very uncertain on the short end. Therefore, differences in the $Xe^{129}$ content are likely to reflect principally the position of the meteorites in their parent object.

Arnold: Then in your model the I-Xe "age" refers to an event which took place later than any of the events discussed by Cameron, and so the interpretation of these ages is still quite uncertain.

## REFERENCES

Burbidge, G. R., Hoyle, F., Burbidge, E. M., Christy, R. F., and Fowler, W. A. (1956) Phys. Rev. 103, 1145.

Fish, R. A., Goles, G. G., and Anders, E. (1960) Astrophys. J., 132, 243.

Goles, G. G. (1960) J. Geophys. Res., in press.

Herbig, G. (1956) in STELLAR POPULATIONS, ed. O'Connell (Interscience, New York).

Hoyle, F., and Ireland, J. G. (1960) Mon. Not. Roy. Astronom. Soc. 120, 173.

Kuiper, G. P. (1951) in ASTROPHYSICS, A TOPICAL SYMPOSIUM, ed. Hynek (McGraw-Hill Book Co., Inc., New York).

Platt, J. R. (1960) Lowell Obs. Bulletin 105, 278 - Proceedings of a Conference on the "Polarization of Starlight in the Interstellar Medium."

Reynolds, J. R. (1960a) Phys. Rev. Letters, 4, 8.

Reynolds, J. R. (1960b) unpublished work.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# COMMENTS ON THE TIME DEPENDENCE OF NUCLEOSYNTHESIS

Truman P. Kohman
Department of Chemistry
Carnegie Institute of Technology

The discovery by Reynolds of excess $Xe^{129}$ in meteorites, about which we have just heard, has opened extremely important new avenues to the study of the early events of the solar system. Earlier today we heard from Cameron of a remarkably complete and detailed account of nucleosynthesis and planetary system formation which would account for Reynold's observations and many other facts. Another noteworthy feat of deductive inference has been accomplished by Anders and his collaborators (Goles, Fish and Anders, 1960; Fish, Goles and Anders, 1960), who give convincing arguments for a relatively intense but short-lived source of heat in the meteorite parent bodies. Calculations of the speaker (Kohman 1960) have shown, however, that the continuous nucleosynthesis rapid-mixing model cannot supply it at the right time (Kohman 1956). One way out of this difficulty is Cameron's suggestion of medium-lived radioactivity induced by a high flux of high-energy particles in the solar nebula, but such a radical postulate is not really necessary.

Calculations have been made for a "mixed nucleosynthesis" model, in which the bulk of the stable and long-lived nuclides are made more-or-less continuously over a long period of time, and shortly prior to the formation of the solar system a "spike" of freshly synthesized material from a near-by supernova, to the extent of 1 - 10 percent of the total, is added. Then both the $I^{129}$ and the heating can be accounted for very nicely.

Under this model, about 100 million years more than Cameron assumed [~200 m. y. in all] is available for the processes of condensation of the solar system to the point where the meteoritic parent bodies had cooled sufficiently to retain Xe.

Cameron: These models depend very sensitively on the mixing time assumed. If mixing requires two or three half-lives of $I^{129}$, then it should be possible to get local enrichment of this nuclide. The problem is, in a sense, a philosophical one: should we try to construct a general model, assuming complete mixing, or should we work with the special case of the arrival of just what is needed at just the right time?



Copyright © National Academy of Sciences. All rights reserved.

Kohman: In order to avoid invoking a rare coincidence to account for the solar system, would it not be possible to assume that the "hot breeze" from the local supernova had some necessary correlation, causal or otherwise, with the initiation of condensation?

Cameron: The trouble with that suggestion is that it appears to be necessary, from Jeans' criterion, for the galactic cloud to remain undisturbed for a long period while condensing into protostars. Any large-scale disturbance of the cloud would probably result in its dispersal.

Anders: On the other hand, the more massive stars in such a condensing cloud would evolve very quickly, within about 10 million years, and would then explode and inject freshly-synthesized material into the cloud.

Cameron: I would expect that such events would result in the disruption of the cluster by sweeping out the nebular gas and dust and thereby reducing the total mass so that the remaining stars would no longer be gravitationally bound.

## REFERENCES

Fish, R. A., Goles, G. G., and Anders, E. (1960) Astrophys. J. 132, 243.

Goles, G. G., Fish, R. A., and Anders, E. (1960) Geochimica et Cosmochimica Acta, 19, 177.

Kohman, T. P. (1960) Paper presented at Cleveland Meeting of American Chemical Society, April 1960; to be published in J. Chem. Ed.

Kohman, T. P. (1956) Ann. N.Y. Acad. Sci. 62, 503.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# XENON IN STONE METEORITES

John H. Reynolds
Department of Physics
University of California at Berkeley

[REYNOLDS reviewed briefly the negative results of attempts to find excess $Xe^{129}$ in Beardsley (chondrite) and Nuevo Laredo (achondrite), and discussed his positive results on Richardton (chondrite) (Reynolds 1960a) and Murray (carbonaceous chondrite) (Reynolds 1960b).] Table 1 gives the relative abundance of isotopes in the anomalous component of Xe from Richardton, Murray, Mighei and Orgueil. These values are derived by subtracting Xe of terrestrial composition, normalized to $Xe^{136}$, from the total meteoritic Xe as given by peak heights. This normalization is arbitrarily chosen so that the anomalous abundances are all positive. It seems likely that a considerable fraction of the so-called terrestrial component is in fact atmospheric Xe adsorbed on the sample and apparatus. No outgassing of the crucible was included in the procedure; the extraction system is being redesigned in order to make it possible to include such a step.

TABLE 1

Anomalous Components of Meteoritic Xenon
(Percent Abundance)

| Isotope | Richardton | Murray | Mighei | Orgueil |
|---|---|---|---|---|
| 124 | 0.33±0.09 | 0.64±0.05 | 0.62±0.11 | 0.72±0.24 |
| 126 | 0.23±0.02 | 0.56±0.10 | 0.52±0.10 | 0.52±0.25 |
| 128 | 2.5 ±0.3 | 6.52±0.25 | 6.05±0.50 | 6.45±0.41 |
| 129 | 81.5±3.5 | 46.6±3.4 | 54.3±4.4 | 49.4±4.4 |
| 130 | 2.3±0.4 | 7.33±0.81 | 5.67±0.60 | 6.08±0.68 |
| 131 | 8.2±2.0 | 25.12±2.7 | 22.5±2.9 | 24.03±3.0 |
| 132 | 3.6±2.3 | 11.59±3.4 | 9.52±3.37 | 11.63±2.5 |
| 134 | 1.4±1.0 | 1.69±1.3 | 0.83±1.30 | 1.16±1.31 |
| 136 | 0 | 0 | 0 | 0 |



Copyright © National Academy of Sciences. All rights reserved.

In general, there appear to be two types of Xe mass spectra observed in meteorites. The "anomalous" spectra are, except for variable amounts of $Xe^{129}$, identical within experimental error with that of Murray, and when normalized to terrestrial Xe (choosing $Xe^{132}$ as the reference isotope) indicate that there may have been a mass fractionation effect in the Earth's atmosphere. The two heaviest Xe isotopes, $Xe^{134}$ and $Xe^{136}$, do not seem to follow such a fractionation curve, however [see especially Reynolds (1960b) for discussion of this point]. The second type of spectrum, of which Beardsley and Pesyanoe are examples, appears to be terrestrial in isotopic composition. Both Beardsley and Pesyanoe have very little Xe, so that the observed spectra may be due to the blank.

Kohman: If one attributes all this variation to some effect which is a function of mass, then there is a deficiency of $Xe^{129}$ in Murray [and the other carbonaceous chondrites], rather than an excess.

Reynolds: That is correct, but then $Xe^{134}$ and $Xe^{136}$ are in substantial excess in the meteorite. There are difficulties in accounting for the excess of these isotopes as fission products, since their amounts do not fit the yield curve for spontaneous fission of $U^{238}$.

Anders: Kuroda (1960) has treated the problem by normalizing to $Xe^{130}$, which is a shielded isotope and therefore should be unaffected by fission, and by suggesting that neutron-induced fission or fission of extinct transuranic elements such as $Pu^{244}$ could have accounted for most of the observed differences between the Earth's atmosphere and the meteoritic Xe. [Kuroda's idea is that the U/Xe ratio for the Earth is higher than for the meteorites, so that atmospheric xenon has been enriched in those isotopes of Xe which are fission products--especially by spontaneous fission of $Pu^{244}$.]

Reed: Is this scheme self-consistent; that is, can one normalize to $Xe^{128}$, which is also shielded, and get similar results?

Anders: Yes, within the accuracy of the data. Kuroda's treatment reproduces the second-order peak at $Xe^{132}$ which one would expect from the data on transuranic spontaneous fission. Also, the secondary anomalies in meteoritic Xe are considerably reduced, so that they could possibly be accounted for by spallation.

Reynolds: In view of this discussion, it is very important to find out whether all Xe from "xenon-rich" meteorites has an anomalous component closely similar to that of Murray, which one would certainly expect from Kuroda's arguments, or whether there is any Xe of strictly terrestrial composition in such meteorites. This would be a crucial test of



Copyright © National Academy of Sciences. All rights reserve

Kuroda's hypothesis. Meteorites such as Beardsley, Pesyanoe and Nuevo Laredo should also be investigated further, in a system with small blanks.

Table 2 presents a survey of results on excess $Xe^{129}$ to date. The $Xe^{129}/Xe^{132}$ ratios in the table are the maximum observed. Data obtained by other workers at Heidelberg and Minnesota are also included. The ratios sometimes display considerable variability from one sample to another of the same meteorite, which may be due to variance in the blank, or to real variations among samples [the iodine results reported by Goles for some of these meteorites also display large variance among different aliquots]. Sample sizes, while ranging up to ~6 g, were normally about 1.5 g, so that some sampling errors could be expected.

TABLE 2

$Xe^{129}/Xe^{132}$ Ratios For Meteorites

| | | |
|---|---|---|
| Indarch | 3.4 | (Heidelberg: 3.1) |
| Richardton | 1.48 | (Minnesota: 1.4) |
| | | (Heidelberg: 1.25) |
| Murray | 1.09 | |
| Elenovka | 1.08 | (tentative) |
| Mighei | 1.08 | |
| Orgueil | 1.06 | |
| Kyushu | 1.05 | (tentative) |
| Atmosphere | 0.98 | |

The absolute amounts of excess $Xe^{129}$, using the normalization to $Xe^{136}$ for subtracting a terrestrial component, were determined by isotopic dilution with a $Xe^{128}$ spike made by irradiation of KI. These amounts, in cc STP/g, are:

| | |
|---|---|
| Richardton | $0.13 \pm 0.01 \times 10^{-9}$ |
| Indarch | $2.54 \pm 1.0 \times 10^{-9}$ |
| Murray | $1.09 \pm 0.11 \times 10^{-9}$ |

The Indarch result is provisional; it was hurried to completion, at the expense of accuracy, so as to be reported at this meeting. However, it is clear that Indarch, while exceptionally rich in excess $Xe^{129}$, has a total Xe content much smaller than Murray (Murray has ~ $40 \times 10^{-9}$ cc STP/g total Xe, while Indarch has roughly $6 \times 10^{-9}$ cc STP/g Xe). The Xe in Murray has been shown (Reynolds 1960b) to be primordial.



Copyright © National Academy of Sciences. All rights reserved.

The Ar from Murray and the other carbonaceous chondrites also appears to be primordial. The $Ar^{40}/Ar^{36}$ ratios in these meteorites are:

| | |
|---|---|
| Murray: | 8.5 |
| Orgueil: | 5 |
| Mighei: | ~17. |

These ratios are clearly very different from the atmospheric value of ~300. While the isotopic compositions of Ar in these carbonaceous chondrites seem very similar, the total amounts may differ. [Note that Orgueil has a higher K content (730 ppm) than Murray (380 ppm) according to Edwards, and so must have either a much shorter K-Ar age or much more $Ar^{36}$ than Murray. REYNOLDS reported 260 ppm K in Murray, and a K-Ar age of ~3 A. E., which would strengthen the argument above.]

Cameron: What information do you have on primordial $He^3$ and $Ne^{20}$? These isotopes are very important, $He^3$ because it can only be made in a limited number of ways, and $Ne^{20}$ because it is made in cool stars and the intergalactic material seems to have been greatly enriched in it since the solar system was formed.

Reynolds: The He in Pesyanoe, assuming its U content is low enough so that there was no contribution of radiogenic $He^4$, is primordial. (The $He^3$ is about ten times as abundant as $Ne^{21}$, implying a negligible amount of cosmogenic He.) The $He^3/He^4$ ratio in Pesyanoe is ~0.004 according to our measurements, so that this is probably the primordial $He^3/He^4$ ratios. Zahringer and Gentner (1960) have come to a similar conclusion from their work on Kapoeta.

Anders: Some Russian work has been published on Pesyanoe claiming $\sim 10^{-6}$ g U/g. However, I put a sample of Pesyanoe directly into a scintillation counter and looked for γ-radiation from the $U^{238}$ decay chain. I would estimate $10^{-7}$ g U/g as an upper limit: possibly much less is present.

Reynolds: Then 0.0004 for the primordial $He^3/He^4$ ratio would seem to be a good value. I have results on Murray (0.0005) and Orgueil (0.0004) which agree with this, but the agreement may be fortuitous. The $Ne^{21}/Ne^{20}$ ratios for these carbonaceous chondrites are:

| | |
|---|---|
| Murray: | 0.034 |
| Orgueil: | 0.013 |

Much of this data is summarized in Figure 1. The profiles of primordial noble gases in stone meteorites are compared with cosmic (Suess-Urey) and atmospheric noble gases. (The data on Kapoeta and Abee



Copyright © National Academy of Sciences. All rights reserved.

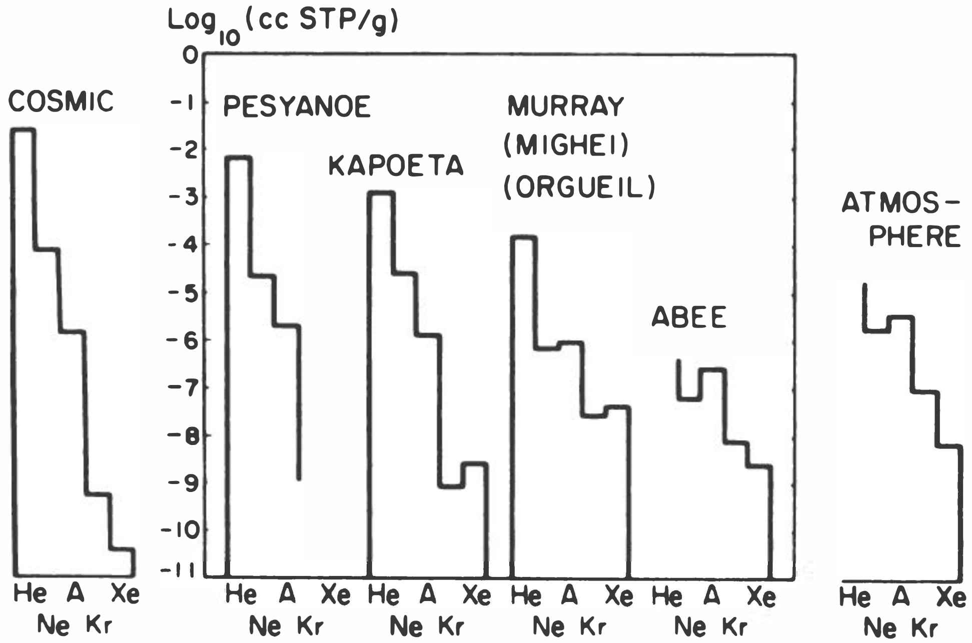


Figure 1. Primordial noble gases in stone meteorites.

are from Zahringer; Pesyanoe data from Russian work.) There are marked similarities between Pesyanoe, Kapoeta and the cosmic profile on the one hand, and Murray, Abee and the atmospheric profile on the other.

[Cameron and Suess discussed the Ne/Ar ratios at some length, pointing out possible reasons for variations between the given profiles. Suess suggested that it is important to keep in mind the effects of adsorption, desorption and diffusion in changing the profiles, e.g., by inducing a loss of Ne relative to Ar or Kr relative to Xe, which could account for some of the features of the profiles. Anders indicated that such separation processes would be critically dependent upon temperature, among other parameters. For further discussion of some of these points, see the paper by Stauffer in this volume.]

Finally, it now seems likely that the K-Ar age of Richardton is greater than that of Beardsley. Using the results of recent potassium analyses, Richardton has a K-Ar age of 4.47 A.E., compared to 4.30 A.E. for Beardsley according to Geiss and Hess. Within experimental error, these values are the same, but at least it is no longer necessary to try to



Copyright © National Academy of Sciences. All rights reserved.

explain why Beardsley should appear to be older than Richardton and yet have retained no excess $Xe^{129}$.

## REFERENCES

Kuroda, P. K. (1960) Nature 187, 36.

Reynolds, J. H. (1960a) Phys. Rev. Letters 4, 8.

Reynolds, J. H. (1960b) Phys. Rev. Letters 4, 351.

Zähringer, J., and Gentner, W. (1960) Z. fur Naturfor. 15a, 600.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# PRIMORDIAL ARGON AND NEON IN STONE METEORITES

Heinz Stauffer
School of Science and Engineering
University of California at La Jolla

The abundance and isotopic composition of argon and neon in carbonaceous chondrites and ureilites (diamond-bearing achondrites) have been measured. A detailed description of the experimental procedure is to be published by Stauffer (1960). The results are given in Table 1.

The striking results are the unusually high abundances of the isotopes of $Ar^{36}$ and $Ar^{38}$. The ratios $Ar^{36}/Ar^{38}$ are all very close to that of atmospheric argon. However, the low values for the ratios $Ar^{40}/Ar^{36}$ show that only a small fraction of the isotopes $Ar^{36}$ and $Ar^{38}$ could be due to atmospheric contamination, and therefore their extraterrestrial origin is proved. No nuclear processes are known to produce them in meteorites in such quantities and in such a ratio. It is concluded, therefore, that they are primordial.

The isotopic composition of cosmogenic neon in stone meteorites has been found to be about $Ne^{20}/Ne^{21}/Ne^{22} = 0.9/0.93/1.0$. The results of Table 1 show in each case an excess of $Ne^{20}$ and $Ne^{22}$. Except for Mokoia, an addition of atmospheric neon to the cosmogenic fraction could explain the measured isotopic ratios. However, for the carbonaceous chondrites atmospheric contamination can be excluded by comparing the observed $Ar^{40}/Ne^{20}$ ratios with the corresponding atmospheric ratio, and therefore the excess neon is considered to be primordial.

Table 2 shows the effect of adsorbed atmospheric argon. The large differences in the $Ar^{40}/Ar^{36}$ ratios between the two different samples of Pesyanoe and between the aliquots of the Goalpara sample are due to different amounts of adsorbed atmospheric argon. The abundances of primordial argon agree.

Several heating experiments have been carried out. The results are listed in Table 3. For a detailed discussion of the experimental procedure and the results, see Stauffer (1960). Summarizing, we may conclude:



Copyright © National Academy of Sciences. All rights reserved.

## TABLE 1

Results of Ar and Ne Measurements
Absolute amounts given in $10^{-8}$ cc STP/gm

| Meteorite | $Ar^{40}/Ar^{36}$ | $Ar^{36}/Ar^{38}$ | $Ar^{40}$ | $Ar^{38}$ | $Ar^{36}$ | $Ne^{20}/Ne^{22}$ | $Ne^{21}/Ne^{22}$ | $Ne^{21}$ |
|---|---|---|---|---|---|---|---|---|
| **Ureilites:** | | | | | | | | |
| Novo Urei | 1.09 | 5.33 | 260 | 44.7 | 238 | 1.33 | 0.87 | 2.25 |
| Goalpara | $\leq$11.1 | 5.14 | 440 | 7.7 | 39.6 | 0.956 | 0.88 | 8.55 |
| **Carbonaceous Chondrites:** | | | | | | | | |
| Felix | 19.5 | 5.15 | 3280 | 32.6 | 168 | 1.22 | 0.89 | 13.9 |
| Lance | 11.8 | 5.33 | 2050 | 32.6 | 174 | 2.53 | 0.74 | 1.34 |
| Mokoia | 43.0 | 5.20 | 1490 | 6.7 | 34.6 | 11.9 | 0.124 | 3.25 |
| Ivuna | 6.64 | 5.35 | 640 | 18.0 | 96.5 | 8.5 | 0.16 | 0.4 |
| Murray | 3.65 | 5.35 | 500 | 25.6 | 137 | 9.12 | 0.167 | 1.00 |
| Murray* | $<$8.54 | 5.39 | 653 | 14.2 | 76.5 | 7.19 | 0.245 | 2.22 |

*Results published by Reynolds (1960).

NOTES:
1. All results corrected for blanks.
2. Errors in $Ar^{40}/Ar^{36}$ ratios smaller than 5%.
3. Errors in all other isotopic ratios smaller than 2%.
4. Errors in absolute amounts smaller than 5%.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 2

Variation in Adsorbed Ar-content

| Sample | | 40/36 | $Ar^{40}$ | $Ar^{36}$ | $Ar^{36}/Ar^{38}$ |
|---|---|---|---|---|---|
| Pesyanoe I | Sample obtained from Russia | 28.7 | 4590 | 160 | 5.17 |
| Pesyanoe II | Sample obtained from Smithsonian Collection | 25.1 | 3880 | 155 | 5.25 |
| Goalpara | Two different aliquots of the same sample | 11.1 | 440 | 39.6 | 5.14 |
| | | 16.5 | 651 | 39.5 | 5.05 |

Absolute amounts given in $10^{-8}$ cc STP/gm

1. Predegassing the samples for several hours at about 100° C under continuous pumping releases the adsorbed atmospheric argon in the case of Lance. We may expect that to be true for all carbonaceous chondrites. All samples listed in Table 1 have been predegassed. Therefore, the $Ar^{40}$ abundances given in Table 1 are equal to the abundances of radiogenic $Ar^{40}$, except for the sample Goalpara, where a considerable fraction of the adsorbed argon survives the predegassing.

2. Radiogenic argon diffuses out more readily than primordial argon. The diffusion coefficients have been calculated, using the same mathematical model as Goles, Fish and Anders (1960). They are listed in Table 4.

3. The primordial argon is much more strongly bound than the radiogenic. It is enclosed within the matrix of the crystal lattice.

4. The same is true for cosmogenic neon, since its diffusion coefficient is smaller than that of radiogenic argon.

5. No accurate value for the diffusion coefficient of primordial neon could be given because of high blanks. It seems, however, that there are no large differences between the diffusion coefficients of primordial and cosmogenic neon.

6. Inhomogeneities in the content of primordial rare gases occur.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

## TABLE 3

### Heating Experiments

| Sample | Experiment | $\frac{Ar^{40}}{Ar^{36}}$ | $Ar^{40}$ | $Ar^{36}$ | $\frac{Ne^{20}}{Ne^{21}}$ | $Ne^{20}$ | $Ne^{21}$ |
|---|---|---|---|---|---|---|---|
| Lance I | 2 hr at 105° C | 270 | 320 | 1.2 | | | |
| | 2 hr at 315° C | 140 | 150 | 1.1 | | | |
| | completely reacted | 11.7 | 2010 | 171.8 | 3.42 | 4.58 | 1.34 |
| | Total | 14.1 | 2480 | 174 | | | |
| Lance II predegassed | 1 hr at ~1000° K | 33.3 | 850 | 25.5 | 2.9 | 0.93 | 0.32 |
| | completely reacted | 8.15 | 1210 | 148.5 | 3.46 | 3.29 | 0.95 |
| | Total | 11.84 | 2060 | 174 | 3.32 | 4.22 | 1.27 |
| Lance III | Sample completely reacted, not pre-degassed | 13.65 | 2410 | 176.5 | 6.03 | 7.60 | 1.26 |
| Goalpara predegassed | 1 hr at ~1000° K | 281 | 450 | 1.6 | | | 0.92 |
| | completely reacted | 6.33 | 126 | 19.9 | 1.01 | 7.63 | 7.55 |
| | Total | 26.8 | 576 | 21.5 | | | 8.47 |

Absolute amounts given in $10^{-8}$ cc STP/gm



Copyright © National Academy of Sciences. All rights reserved.

TABLE 4

Diffusion Coefficients Calculated From Heating Experiments

| Sample | Conditions | $Ar^{40}$ | | $Ar^{36}$ | | $Ne^{21}$ | |
|---|---|---|---|---|---|---|---|
| | | F | $\frac{D}{a^2}$ sec$^{-1}$ | F | $\frac{D}{a^2}$ sec$^{-1}$ | F | $\frac{D}{a^2}$ sec$^{-1}$ |
| Lance | 1 hr 1000° K | 41.3 | $5.3 \times 10^{-6}$ | 14.7 | $5.6 \times 10^{-7}$ | 25 | $1.8 \times 10^{-6}$ |
| Goalpara | 1 hr 1000° K | 78 | $3 \times 10^{-5}$ | 7.4 | $1.4 \times 10^{-7}$ | 11 | $3.1 \times 10^{-7}$ |
| Chondrites* | 1 hr 973° K | 14 | $5.15 \times 10^{-7}$ | | | | |

F = Fraction expelled, in % of total content.
* = Value calculated from heating experiments by Geiss and Hess (1958).

The only potassium analyses on carbonaceous chondrites are those published by Edwards (1955). Using his values and the present argon analyses, the following Ar/K ages have been calculated: Felix 4.5 A.E.; Mokoia 3.4 A.E.; Murray 1.9 A.E.; Ivuna 1.4 A.E. The Ar/K age of Pesyanoe is given as 4.2 A.E. by Gerling and Levskii (1956). No potassium content is known for Novo Urei, and the value for Goalpara given by Edwards (1955) is very uncertain as is the content of radiogenic $Ar^{40}$.

From Table 5 it can be seen that a large loss of rare gases compared to the silicon, as well as a fractionation between them, must have occurred in the history of meteoritic material. Furthermore, the degree of such fractionation shows differences of several factors of ten between different samples. Comparing the results of the heating experiments with the Ar/K ages and the values in Table 5, it can be concluded:

1. The high Ar/K age of Felix shows that no loss of radiogenic argon and--because of the difference in diffusion coefficients--no loss of primordial argon occurred since the formation of this meteorite. Only minor losses of primordial neon could have occurred. The observed fractionation must have happened before or during the formation of the meteorite.

2. The other carbonaceous chondrites have lost a large fraction of the radiogenic argon, about 50 percent for Mokoia and about 90 percent for Ivuna. Here we expect losses of primordial gases too, though it is difficult to estimate their extent. However, the observed differences between the diffusion coefficients of primordial argon and primordial neon cannot alone account for the large fractionation.



Copyright © National Academy of Sciences. All rights reserve

## TABLE 5

Primordial Ar and Ne
Absolute amounts given in $10^{-8}$ cc STP/gm

| Sample | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| | prim. $Ar^{36}$ | prim. $Ne^{20}$ | $\left(\frac{Ne^{20}}{Ar^{36}}\right)$ prim. | $\log\left(\frac{\text{prim. } Ar^{36}}{\text{silicon}}\right)$ | $\left(\frac{Ar^{36}}{Ar^{38}}\right)$ prim. | $\left(\frac{Ne^{20}}{Ne^{22}}\right)$ prim. |
| Novo Urei | 238 | 1.3 | .005 | -7.8 | 5.36 | |
| Goalpara | 39 | 1.0 | .026 | -8.6 | 5.88 | |
| Lance | 174 | 3.3 | .019 | -7.85 | 5.36 | |
| Felix | 167 | 5.6 | .034 | -7.9 | 5.40 | |
| Ivuna | 96.5 | 20.8 | .22 | -7.95 | 5.37 | 9.8 |
| Abee(1) | 25 | 6.2 | .25 | -8.75 | 5.50 | |
| Murray | 137 | 53.7 | .39 | -7.9 | 5.37 | 10.6 |
| Atmospheric | | | .52 | -9.7 | 5.35 | 10.2 |
| Murray(2) | 76 | 63.2 | .83 | -8.15 | 5.47 | 9.2 |
| Mokoia | 34 | 309 | 9.1 | -8.6 | 5.39 | 13.2 |
| Pesyanoe | 159 | 1886 | 11.9 | -8.1 | 5.46 | 12.2 |
| Kapoeta(1) | 107 | 2400 | 22.4 | -8.1 | 5.20 | 14.0 |
| Cosmic | | | 61.4 | -0.9 | 6.07 | 14.6 |

(1) Calculated from the results published by Zähringer and Gentner (1960).
(2) Calculated from the results published by Reynolds (1960).
Col. 1 and 5: Correction for cosmogenic argon calculated from the $Ne^{21}$ contents, with $(Ne^{21}/Ar^{38})_{cosmogenic} = 8$ and $(Ar^{36}/Ar^{38})_{cosmogenic} = 0.6$. The cosmic ratio is taken from Suess (1949).
Col. 2 and 6: Correction for cosmogenic neon calculated with a cosmogenic ratio $Ne^{20}/Ne^{21}/Ne^{22} = 0.9/0.93/1.0$. The cosmic ratio is taken from Suess (1949).
Col. 3 and 4: Cosmic ratios calculated from Suess-Urey (1956) abundance tables.
Col. 4: Ratios calculated from the silicon values given by Wiik (1956) for the carbonaceous chondrites and by Urey and Craig (1953) for ureilites. The terrestrial ratio is taken from Suess (1949).



Copyright © National Academy of Sciences. All rights reserved.

The degree of fractionation may depend on differences in the properties of the rare gases. Processes such as adsorption and, at low temperatures, condensation are known to give large fractionation. Also, a selective trapping of the rare gases in the crystal lattice, or at dislocations, may cause a large fractionation.

Small isotopic effects are expected in all possible mechanisms of fractionation, between rare gases, due to the differences in the mass of the isotopes. It can be seen from Table 5 that in some cases the primordial isotopic ratios differ from the atmospheric values. The errors are rather high, due to the uncertainty in the correction for the cosmogenic rare gases. However, for the samples Mokoia and Pesyanoe, the primordial $Ne^{20}/Ne^{22}$ ratios are undoubtedly higher than the atmospheric value, and in Kapoeta it almost agrees with the cosmic ratio, calculated by Suess (1949) from the distribution of the rare gases in the Earth's atmosphere. Correspondingly, these samples show the smallest fractionation, and they have almost a cosmic $Ne^{20}/Ar^{36}$ ratio.

## REFERENCES

Edwards, G. (1955) Geochimica et Cosmochimica Acta 8, 285.

Geiss, J., and Hess, D. C. (1958) Astrophys. J. 127, 224.

Gerling, E., and Levskii, L. (1956) Dokl. Akad. Nauk. S.S.S.R. 110, 750.

Goles, G., Fish, R. A., and Anders, E. (1960) Geochimica et Cosmochimica Acta 19, 177.

Reynolds, J. H. (1960) Phys. Rev. Letters 4, 351.

Stauffer, H. (1960) to be published in Geochim. et Cosmochim. Acta.

Suess, H. E. (1949) J. Geology 57, 600.

Suess, H. E., and Urey, H. C. (1956) Rev. Mod. Phys. 28, 53.

Urey, H. C., and Craig, H. (1953) Geochim. et Cosmochim. Acta 4, 36.

Wiik, H. B. (1956) Geochim. et Cosmochim. Acta 9, 279.

Zähringer, J., and Gentner, W. (1960) Zeitschr. f. Naturf., in press.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# COSMIC RAY AGES OR IRON METEORITES*

Oliver A. Schaeffer
Chemistry Department
Brookhaven National Laboratory

By the cosmic ray age of a meteorite is meant the length of the time interval between the formation of the meteorite in its preatmospheric form and its collision with the earth. The knowledge of such ages is of interest in the understanding of the origin of meteorites and the past history of the solar system. To measure the cosmic ray exposure age one needs to measure the time-integrated content and rate of production of a cosmic ray produced nuclide which was not present in the meteorite before being exposed to cosmic rays. The most easily measured products are the stable rare gases. In order to obtain the production rate it is necessary to measure the cosmic ray induced activity of a nuclide with a half life short compared to the cosmic ray age and long compared to the time since its arrival at the earth. It is then only necessary to know the relative production rate of the radioactive and stable products. A simple case exists if the stable product is produced predominantly by the decay of the radioactive nuclide, then the two production rates are of course equal. In addition, it is not necessary to make a correction for the dependence of the relative production rates on the energy of the cosmic ray particles.

There are two cases where a relatively long lived activity decays to a stable rare gas, namely $Cl^{36}$, $Ar^{36}$ and T, $He^{3}$. Of course the production rates of $He^{3}$ and T or of $Ar^{36}$ and $Cl^{36}$ are not equal as in each case some of the rare gas is produced directly by the action of cosmic rays. Thus, if one computes an exposure age it is necessary to make a correction for the directly produced material.

A cosmic ray exposure age can also be computed by measuring a radioactive nuclide of nearly the same mass, but not necessarily decaying to a rare gas nuclide, e.g., $Ar^{39}$, $Ar^{38}$ is such a pair. In this case one must have rather complete information on the cosmic ray production rate of the two nuclides.

---

* Research performed under the auspices of the U. S. Atomic Energy Commission.



Copyright © National Academy of Sciences. All rights reserved.

In general, in order to interpret the radioactive and rare gas measurements it is necessary to have knowledge of the cosmic ray production ratios of various nuclides from meteoritic material.

Production ratios can be determined from high energy proton irradiation of target elements of representative materials. In the case of nickel-iron meteorites, iron itself furnishes a good target material. In the case of the stone meteorites, there is no single element which can reasonably approximate the meteorite, so in the case of stones a number of target materials should be studied as well as stone meteorites themselves.

In comparing the results of thin target irradiations to the products in meteorites one must exercise some caution. In the meteorite, a large number of low energy secondaries are formed which in turn produce radioactive and stable rare gas products. In a thin target, however, the secondary production is, to a large extent, missed. If a given ratio of products depends on the energy of the bombarding particles, a comparison of meteorite results to thin target data can be misleading. For this reason, it is important to determine cosmic ray ages from the ratio of two nearly equal mass nuclides as such a ratio is usually not too sensitive to the energy.

Let us turn to some of the bombardment results. It has been found that the production ratios of the argon isotopes in the energy interval 200 Mev to 6 Bev are constant to within 5% and are $Ar^{36}:Ar^{37}:Ar^{38}:Ar^{39}::1:33:8:4.2$. These ratios are in good agreement with the predictions of nuclear evaporation theories. On the other hand the production ratio $He^3+T/He^4$ (which is to be compared to $He^3/He^4$ ratios in meteorites) varies considerably with proton energy, from 0.15 at 200 Mev to 0.30 at 6 Bev. Finally the ratio $He^3+T/Ar^{38}$ varies from 10 at 400 Mev to 23 at 3 Bev. A cosmic ray age dependent on a ratio of $Ar^{39}/Ar^{38}$ should be much more reliable than an age dependent on a $T/He^3$ ratio or a $T/Ar^{38}$ ratio.

In Table 1 are listed the rare gas ratios in several iron meteorites. It is seen that the meteorite results are in the same range as the proton production ratios quoted above. The $Ar^{38}/Ar^{36}$ ratio is essentially constant at 1.6 as would be expected from the lack of dependence of this ratio on energy. The corresponding proton production rate is, however, five times as large. This is due to the fact that evidently only 1/5 of the $Ar^{36}$ is directly produced by cosmic rays and 4/5 of the $Ar^{36}$ must come from the decay of $Cl^{36}$. From this result, one concludes that a cosmic ray age based on the ratio $Ar^{36}/Cl^{36}$ should be practically independent of the energy of the bombarding particle and hence the depth of the sample in the original meteorite.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 1

Cosmogenic Nuclides in Typical Iron Meteorites

| Meteorite | $He^3$ ($10^{-8}$ cc/g) | $He^3/He^4$ | $He^3/Ar^{38}$ | $Ar^{38}/Ar^{36}$ | $Cl^{36}$ (dpm/kg) | $Cl^{36}$-$Ar^{36}$ age |
|---|---|---|---|---|---|---|
| Williamstown | 480 | 0.25 | 18 | 1.5 | 3.3 | 2200 m.y. |
| Canyon Diablo | 95 | 0.30 | 23 | 1.6 | 6.8 | 160 m.y. |
| Carbo* | 350 | 0.24 | 19 | 1.6 | 3.6 | 1200 m.y. |
| Sikhote-Alin | 130 | 0.27 | 32 | 1.5 | 7.8 | 170 m.y. |

* Rare gas analyses from results of Nier et al., University of Minnesota.

The $He^3/He^4$ ratio in the meteorites listed in Table 1 runs from 0.24 to 0.30. This ratio shows the energy dependence of the cosmic ray production ratios. The samples of Carbo are evidently from near the center of the preatmospheric meteorite and that of Canyon Diablo from near the surface. This is also shown by the $He^3/Ar^{38}$ ratios. The $Cl^{36}$ values listed in Table 1 also show the same behavior. For this reason a cosmic ray age calculated from a $T/He^3$ ratio depends on the depth in the original meteorite.

In additon to determining cosmic ray exposure ages a study of cosmic ray produced activities can also lead to new knowledge about the cosmic rays themselves. For example, the constancy of cosmic rays in time can be tested by measuring several cosmic ray produced radioactive nuclides with different half-lives. The long lived isotope will yield an average value of cosmic ray flux over a long period, while the short lived isotope will give an average flux for more recent time. Such a convenient set of isotopes is $Ar^{39}$, $t_{1/2}$ = 325 y; $Cl^{36}$, $t_{1/2}$ = 308,000 y and $K^{40}$, $t_{1/2} = 1.3 \times 10^9$ y. The $Ar^{39}$ activity will give a mean value of the flux for the last $10^3$ years, the $Cl^{36}$ for the last $10^6$ years and the $K^{40}$ (as cosmic ray exposure ages are shorter or comparable to the $K^{40}$ half life), for the exposure age of the meteorite. In order to make a comparison the relative production rates must be known. One way of comparing is to use the above cited results and compute exposure ages for the three radioactivities. The comparison of the three ages will then furnish a test for the constancy of cosmic ray flux in time. Such a comparison is afforded by comparing the $Ar^{39}$-$Ar^{38}$ and $Ar^{36}$-$Cl^{36}$ exposure ages of Sikhote-Alin and the $Ar^{36}$-$Cl^{36}$ and $K^{40}$-$K^{41}$ exposure ages of Carbo. The results are listed in Table 2.

It is seen that cosmic ray flux appears to be constant in time when such averages are compared. This observation then removes one of the assumptions implicit in the determination of cosmic ray exposure ages.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 2

Constancy of Cosmic Rays From Cosmic Ray Exposure Ages

| Meteorite | $Cl^{36}$-$Ar^{36}$ age m. y. | $Ar^{39}$-$Ar^{38}$ age m. y. | $K^{40}$-$K^{41}$ age m. y. |
|---|---|---|---|
| Sikhote-Alin | 170 | 150 | --- |
| Carbo | 1200 | --- | 1360* |

* Determination at Max-Planck-Institut für Chemie by Hintenberger et al.

In order to measure the exposure age of a number of meteorites it would be of interest if an age could be obtained from the rare gas content of the meteorite; then, a large number of meteorites could be measured because of the small sample size (the order of 0. 1 g) required for a mass spectrometric measurement compared to the relatively large sample size (the order of 100 g) required for a measurement of the low level activities.

It would be possible to use the rare gases alone if one had some way to make a correction for the change in cosmic ray flux with depth inside a meteorite. One way of accomplishing this is to make use of the observation that some of the rare gas ratios vary with depth in the meteorite. From Table 1 it is seen that $He^3/He^4$ or $He^3/Ar^{38}$ would be a suitable ratio.

Table 3 presents data on rare gases in several meteorites. There are a few cases where the ratios observed are very different from the general trend. These variations are most likely due to impurities in the irons, e. g., the low $He^3/Ne^{21}$ ratio for Tucson is probably due to production of $Ne^{21}$ from Mg, which is known to be present in minute fosterite inclusions. If one assumes that corrections due to such effects are not important for rough age calculations, and assumes that the $He^3$ production rate implied by the data in Table 1 is applicable, estimates of $He^3$ ages may be made. Figure 1 represents a histogram of these ages. A peak in the distribution near 200 million years is apparent, as well as a long tail out to 2 billion years. This pattern may be interpreted as a reflection of the collision probabilities for meteoroids (so that fresh surfaces are exposed) and of the mean lifetimes for collision with the Earth.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 3

Rare Gas Contents of Iron Meteorites

| Meteorite | $He^3$ ($10^{-8}$ cc/g) | $He^3/Ne^{21}$ | $He^3/Ar^{38}$ | $He^3/He^4$ | $Ne^{21}/Ar^{38}$ |
|---|---|---|---|---|---|
| Toluca | 100 | 125 | 42 | 0.34 | 0.33 |
| Casas Grande | 130 | 130 | 36 | 0.35 | 0.28 |
| Arispe | 270 | 117 | 30 | 0.29 | 0.26 |
| Sikhote-Alin | 130 | 87 | 32 | 0.27 | 0.36 |
| Washington Co. | 193 | 94 | 26 | 0.07 | 0.28 |
| Williamstown | 480 | 80 | 18 | 0.25 | 0.23 |
| Odessa | 220 | 96 | 18 | 0.31 | 0.19 |
| Tucson | 13 | 16 | 14 | 0.09 | 0.88 |
| Smithland | 58 | 94 | 13 | 0.28 | 0.14 |
| Canyon Diablo* | 95 | 95 | 13 | 0.30 | 0.13 |
| Forsyth | 48 | 83 | 13 | 0.32 | 0.16 |
| Santa Catherina | 57 | 111 | 12 | 0.25 | 0.11 |
| Santa Rosa | 47 | 107 | 12 | 0.26 | 0.11 |
| Tombigbee | 4.7 | 61 | 7.3 | 0.14 | 0.12 |
| Canyon Diablo* | 4.2 | 38 | 9.5 | 0.10 | 0.25 |

* Two individual pieces of Canyon Diablo, one obtained from B. Mason, New York Museum of Natural History and the other from Wards Natural Science Foundation, Rochester, New York.

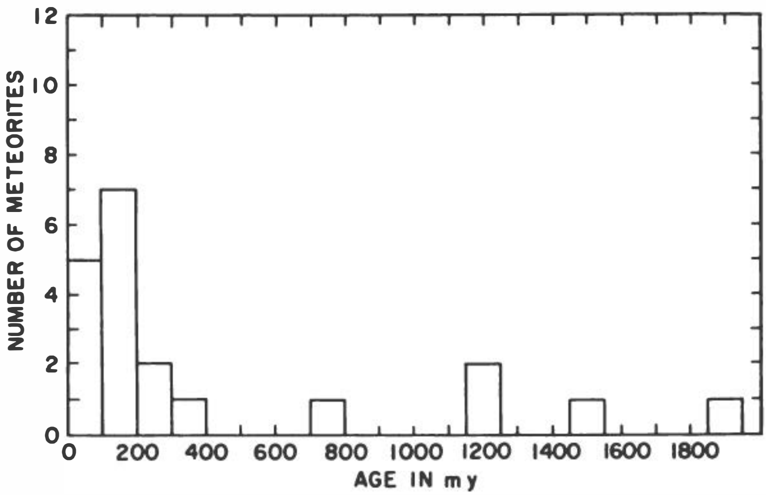


Figure 1. Cosmic ray exposure ages of iron meteorites.



Copyright © National Academy of Sciences. All rights reserved.

Cameron: The age distribution could also reflect differing erosion rates, due to variations in the semi-major axes of the orbits of these meteorites. If erosion is principally by dust swept from the asteroid belt to ward the Sun (by the Poynting-Robertson effect), one would expect approximately a $r^{-1}$ dependence of the dust concentration on distance from the Sun, and perhaps essentially no dust beyond the asteroids.

Fireman: The age of greatest interest is the oldest one, because it at least puts an upper limit on the amount of dust in the regions where the meteorite resided. It appears that this upper limit is significantly lower than the dust concentration in the region of the inner planets estimated from the intensity of the zodiacal light.

Anders: In this connection, it is important to remember that a critical parameter is the orbital inclination. An orbit with high inclination will result in lengthening both the erosional lifetime, since the dust is presumably strongly concentrated in the plane of the ecliptic, and the lifetimes for collision with other meteoroids and with the Earth.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# ARGON-37, ARGON-39 AND TRITIUM IN RECENT METEORITE FALLS

E. L. Fireman
Smithsonian Astrophysical Observatory
Cambridge, Massachusetts

Most of the work on radioactivity in meteorites has been concerned with nuclides of relatively long half-lives simply because the samples available were too old for short-lived activities to be present at measurable levels. Within the past few months, however, three freshly fallen meteorites have been obtained and studied. These are the Hamlet chondrite (fell October 13, 1959, in Indiana), the Aroos iron (fell November 25, 1959, in the USSR) and the Bruderheim stone (fell March 4, 1960, in Alberta). Data on $Ar^{37}$, $Ar^{39}$ and $H^3$ in Hamlet, Aroos, and Benton (fell in 1949; used as a blank for $Ar^{37}$), as well as information on irradiated targets can now be reported; results on Bruderheim are not yet complete.

The content of $Ar^{37}$ (34 day half-life) provides information on the cosmic ray flux during the last fifty or so days before the meteorite struck the Earth. If one had a reliable estimate of the orbit of a freshly fallen meteorite, one could consider the meteorite to be a kind of space probe for cosmic rays. Unfortunately, visual observations of meteorite falls can do no more than supply a rough indication of the direction in which the meteorite was traveling when it collided with the Earth. However, there is one accurately determined orbit, obtained with wide-angle rotating-shutter cameras, that of the Luhy chondrite which fell in Czechoslovakia on April 25, 1959 (Cepleca 1959). This meteorite had an orbit with aphelion 4.1 A.U., perhelion 0.79 A.U., and inclination of $10^{o}$. Its period was 3.8 years. If this orbit is typical of Hamlet and Aroos, then $Ar^{37}$ integrates the cosmic-ray flux over the region of space quite close to the Earth, $H^3$ integrates over seven or eight orbital periods, and $Ar^{39}$ (325 year half-life) would integrate over more than a hundred periods.

The experimental procedures used were essentially the same as those reported previously (Fireman and DeFelice 1960). The total argon activity from the Hamlet and Aroos samples gave evidence of a complex decay curve, which was resolvable into the expected components. The data is summarized in Table 1. The errors quoted are at the 95 percent confidence level, derived from the least-squares analyses used to correlate the raw data. Since no decline in the argon activity from the Benton meteorite was found, only $Ar^{39}$ was still present in this meteorite.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 1

Summary of Data
(Extrapolated to Time of Fall Where Necessary)

| Meteorite or Target | Date of Fall | Type | $Ar^{39}$ Decays/g day | $Ar^{37}/Ar^{39}$ | $H^3$ Decays/g day | $H^3/Ar^{39}$ |
|---|---|---|---|---|---|---|
| Hamlet | 10-13-59 | Chondrite | 8.0 ± 0.5 | 2.3 ± 0.2 | 230 ± 30 | 29 ± 6 |
| Hamlet | 10-13-59 | Chondrite | 8 ± 2 | 2.0 ± 0.5 | 200 ± 50 | 25 ± 10 |
| Hamlet Target | (10-13-59) | Irradiated Chondrite | 800 ± 100 | 1.2 ± 0.3(1) | (500 ± 30) x $10^3$ | 24 ± 5(1) |
| Benton | 1-16-49 | Chondrite | 11.8 ± 0.6 | --- | 340 ± 30 | 29 ± 4 |
| Aroos | 11-25-59 | Iron | 23 ± 2 | 1.4 ± 0.3 | 50 ± 10 | 2 ± 0.5 |
| Iron Target | --- | Irradiated terrestrial iron | --- | 0.8 ± 0.1(2) | --- | 20 ± 5(3) |

(1) Production ratios.
(2) O. A. Schaeffer and J. Zähringer (1958), Z. Naturf. 13a, 347.
(3) E. L. Fireman and J. Zähringer (1957), Phys. Rev. 107, 1695.

The $H^3/Ar^{39}$ ratios in the Hamlet meteorite, including that from the irradiated Hamlet sample, agree quite well among themselves. (The production ratios listed in Table 1 for the irradiated targets are derived by correcting the observed activities to saturation levels, and should therefore be directly comparable with the extrapolated activity ratios in the meterorites exposed to cosmic rays only.) The $Ar^{37}/Ar^{39}$ activity (or production) ratios do not agree so well, however. The higher $Ar^{37}/Ar^{39}$ ratio in Hamlet than that in the irradiated Hamlet target may indicate a cosmic ray flux that is higher near the Earth than a few A. U. out from the Sun. It is also possible that this anomaly may reflect production of $Ar^{37}$ from $Ca^{40}$ by relatively low energy reactions.

The serious discrepancy between the ratio of $H^3/Ar^{39}$ in Aroos and that in the iron target requires comment. A similar, but even more striking effect was observed in Sikhote Alin. A possible explanation is that $H^3$ is lost during the passage of these iron meteorites through the atmosphere. It would not be necessary to heat the interiors of iron meteorites very much in order to cause loss of hydrogen by diffusion.

Anders: This suggestion is very unconventional, since extremely high diffusion rates seem to be required. Would it be feasible to test this idea experimentally, by irradiating an iron meteorite, heating it to a few hundred degrees for a few seconds, and looking for escaped $H^3$?



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

Fireman: We have started to do this on several occasions, but always got sidetracked. I feel, however, that some such explanation must apply, since I believe our experimental results cannot be that far wrong for the $H^3$ in Aroos. We have done other samples by the same method with no trouble. Certainly, the target presents no problem.

Reynolds: We have some data, obtained accidentally, which supports your comment about production of $Ar^{37}$ in low-energy reactions. In one of our pile irradiations of Richardton, the sample was placed in a large flux of fast neutrons, so that enough argon was formed for us to be able to see it on the mass spectrometer. The $Ar^{37}/Ar^{39}$ atomic ratio was about seven in that case. Apparently, half as much $Ar^{36}$ as $Ar^{37}$ was also formed, presumably from the $Ca^{40}$ by (n, nα).

Van Allen: There is an appreciable flux of solar protons, presumably varying as the inverse square of the distance from the Sun. Could they be involved in this $Ar^{37}$ excess?

Fireman: Not for production of $Ar^{37}$ from Fe, but they probably contribute to the type of production from Ca under discussion, as well as to $H^3$ production.

Arnold: Once again the question of ablation during infall is important. If 10 cm or more of material is removed, the effects of the solar protons should not be observable. If very little material is ablated, the solar protons could contribute to the $Ar^{37}$.

Olbert: There is another effect which should be taken into account. The cosmic-ray flux, for low energies, is affected by the eleven-year solar cycle. This could be very important for $Ar^{37}$ and $H^3$, but not, of course, for $Ar^{39}$. Variations by factors of two or larger in the flux have been observed, and measurements of the type reported should be correlated with these changes.

## REFERENCES

Cepleca, Z. D., et al (1959), Bull. Astron. Inst. Czech. 4, 10.

Fireman, E. L. and DeFelice, J. (1960), Geochim. et Cosmochim. Acta 18, 183.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# THE DISTRIBUTION OF RARE GASES IN IRON METEORITES

Peter Signer and Alfred O. Nier
School of Physics
University of Minnesota

This investigation is an attempt to deduce information on the production mechanism of cosmogenic nuclides in iron meteorites from the variation in the concentration of cosmogenic rare gases.

In order to determine the variations in the concentration of the cosmogenic rare gases, 35 samples, taken from locations scattered over a cross section of the 480 kg iron meteorite Grant (fine Octahedrite) were analyzed. In each sample the concentration of $Ar^{38}$, $Ar^{36}$, $Ne^{22}$, $Ne^{21}$, $Ne^{20}$, $He^{4}$ and $He^{3}$ was determined in one single extraction process, using a statically operated mass spectrometer.

Special techniques were used to prevent contamination of the gas mixture extracted from small meteoritic samples (200 - 400 mg) by rare gases from the atmosphere. For He, this problem could be solved completely. The $Ne^{20}$ was affected, in some runs, by air-contamination to the extent of a few percent, whereas for $Ne^{22}$, $Ne^{21}$, $Ar^{36}$ and $Ar^{38}$ contamination was negligible. The lowest measured ratio for $Ar^{38}/Ar^{40}$ was 0.52, which after correction for air-argon becomes 0.4. This is an upper limit for the ratio of cosmogenic $Ar^{38}$ and $Ar^{40}$, because the $Ar^{40}$ may still be contaminated with radiogenic $Ar^{40}$ from the decay of $K^{40}$.

The concentration of the cosmogenic Ar, Ne and He has been found to vary systematically with the location of the sample. Contours of equal concentration could be constructed, as shown for four isotopes in Figure 1. The isopleths for helium agree within the limits of the procedure with those reported earlier by Hoffman and Nier (1958) and Fireman (1959). It is therefore assumed, that the isopleths of Ar and Ne also are closed curves of slightly elliptical shape.

In the subsequent analysis of the variation of the concentration it will be assumed that the isopleths are circles and the straight line in Figure 1, extending from the position of the minimum concentration to the upper right in each diagram (referred to as "reference radius") is a



Copyright © National Academy of Sciences. All rights reserved.

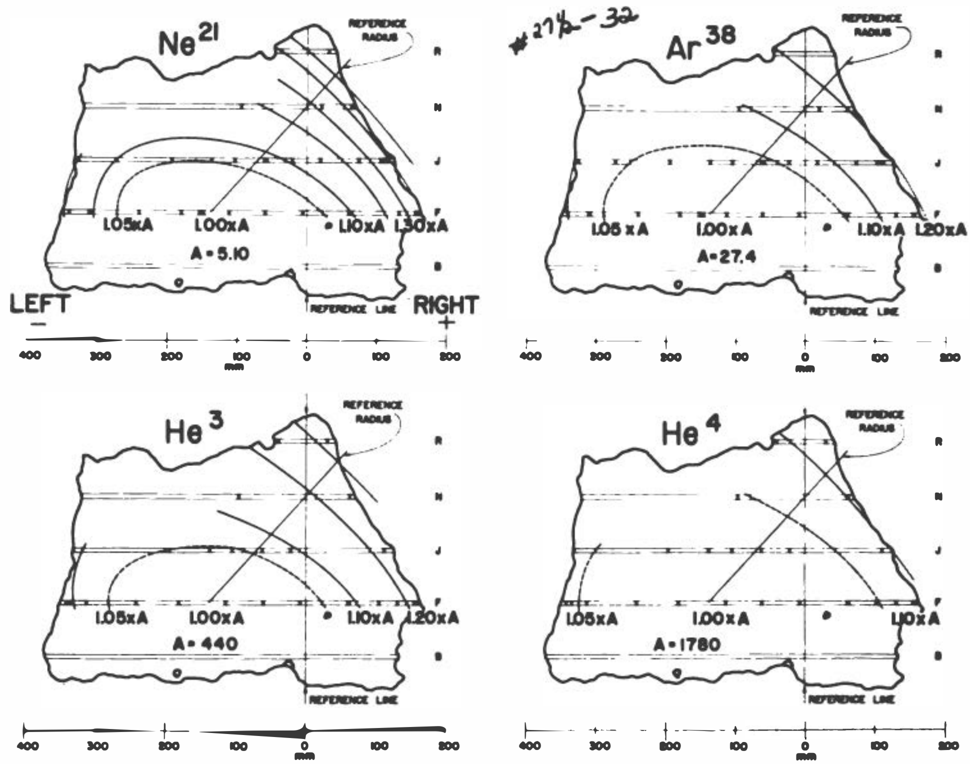


Figure 1. Contours of equal concentration for $Ne^{21}$, $Ar^{38}$, $He^{3}$ and $He^{4}$. Crosses indicate the locations of the measured samples. Measurements were made only in the regions with the lowest concentration and the highest gradient in concentration of the He. Therefore, the curves are not drawn as closed contours. The reference radius is assumed to to be perpendicular to the outer isopleths.

radius of the spherical isopleths. From the diagrams in Figure 1, the increase of the concentration along the reference radius can be constructed. Figure 2 shows this radial increase for $Ar^{38}$, $Ne^{21}$, $He^{4}$ and $He^{3}$. To show the difference in the relative increase, the curves in Figure 2 are normalized to the concentration of each isotope found in the center. $Ne^{21}$ exhibits the most variation with depth, $He^{3}$ and $Ar^{36}$ vary less but roughly the same, whereas $He^{4}$ is the least variable nuclide.

It should be noted that the $Ar^{36}$ was found to increase slightly more than the $Ar^{38}$. At the position of the minimum concentration, $Ar^{36}/Ar^{38}$ is 0.61 and at the post-atmospheric surface it is about 6 percent higher.



Copyright © National Academy of Sciences. All rights reserved.

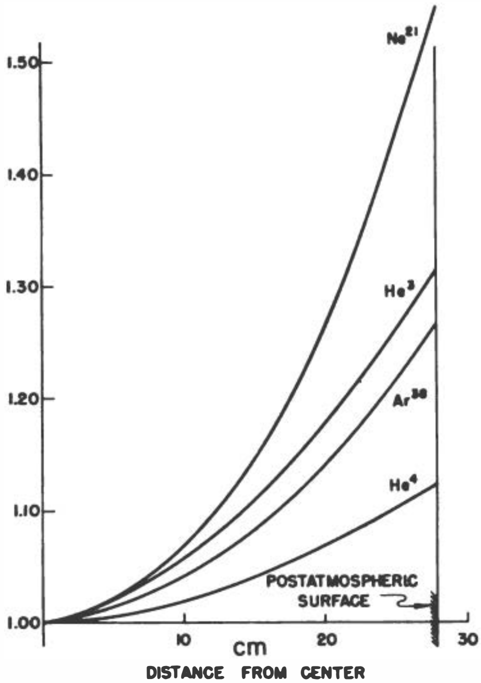


Figure 2. Increase of the $Ne^{21}$, $He^3$, $Ar^{38}$ and $He^4$ concentration along the reference radius. Amounts are normalized to the respective minimum amount.

The abundance of the three Ne-isotopes was found to be, within the limits of error, constant over the whole cross section. ($Ne^{20}/Ne^{21} = 0.96 \pm 0.05$ and $Ne^{22}/Ne^{21} = 1.06 \pm 0.05$).

The precise physical formulation of the production mechanism of the cosmogenic nuclides is not yet possible because of lack of a complete understanding of some of the processes involved. Martin (1953) derived a formula for the production rates of cosmogenic nuclides which has been slightly modified by Ebert and Wänke (1957).

The equation given by the latter authors can be integrated so as to be valid for a spherical target exposed to an isotropic radiation. Accordingly, the number of cosmogenic nuclides of a species "i" produced in a volume is given by an expression of the form

$$P_i(r) = A_i\left[\int_0^{2\pi}\int_0^{\pi} e^{-k_a x}\sin\theta\, d\theta\, d\phi - B_i\int_0^{2\pi}\int_0^{\pi} e^{-k_s x}\sin\theta\, d\theta\, d\phi\right]$$

Where:

X describes the position of the volume under consideration relative to the pre-atmospheric surface.

$k_a$, $k_s$ are parameters which indicate the attenuation of the primary and secondary particles. These parameters are the same for all species "i".

$A_i$, $B_i$ are parameters composed of quantities such as individual production cross sections for the different species; absorption cross sections for primaries and secondaries (also contained in $k_a$ and $k_s$); the number of secondaries produced per primary interaction, and finally the radiation dosage.



Copyright © National Academy of Sciences. All rights reserved.

The basic assumptions made in deriving the stated equation for the production rates are the following ones:

A). The intensity of the primary and secondary particles in an infinite-plane target decreases exponentially with depth, if the incident particles are unidirectional.

B). The energy spectrum of the primaries and secondaries does not change with depth.

C). In each primary interaction a definite number of secondary particles are produced.

D). Wide angle scattering is negligible.

E). Tertiary particles are negligible.

F). The production of the cosmogenic nuclides by both primary and secondary particles can be described by average production cross sections for each species "i".

G). The pre-atmospheric shape of the meteorite was a sphere and its size did not change during the exposure to the cosmic radiation.

For more details refer to the publications by Martin (1953), Ebert and Wänke (1957) and also Signer and Nier (1960).

The parameters $k_a$ and $k_s$ in equation (1) are the same for all species "i" whereas $A_i$ and $B_i$ have to be determined individually for each nucleus. Values of $k_a$, $k_s$, $A_i$, $B_i$ (i = 38, 21, 4, 3) were found by adjusting these parameters to give the best agreement between the experimental data in Figure 2 and the amounts computed with equation (1). Figure 3 shows the agreement between the calculated values (solid curves) and experimental data (dots). The close agreement provides evidence that at least in this case the production mechanism proposed by Martin and modified by Ebert and Wänke is adequate to explain the experimental data.

The fitting process also yields the pre-atmospheric radius (R = 40 cm) of the spherical meteorite. The points on the post-atmospheric surface lay at a value of 0.7 for r/R.

With the parameter values determined in the fitting process it is possible to calculate average values for the absorption cross sections. The present investigation, however, allows only the calculation of production cross section ratios. Absolute values may be obtained if further information such as radiation dosage or a single production cross section is available.



Copyright © National Academy of Sciences. All rights reserved.

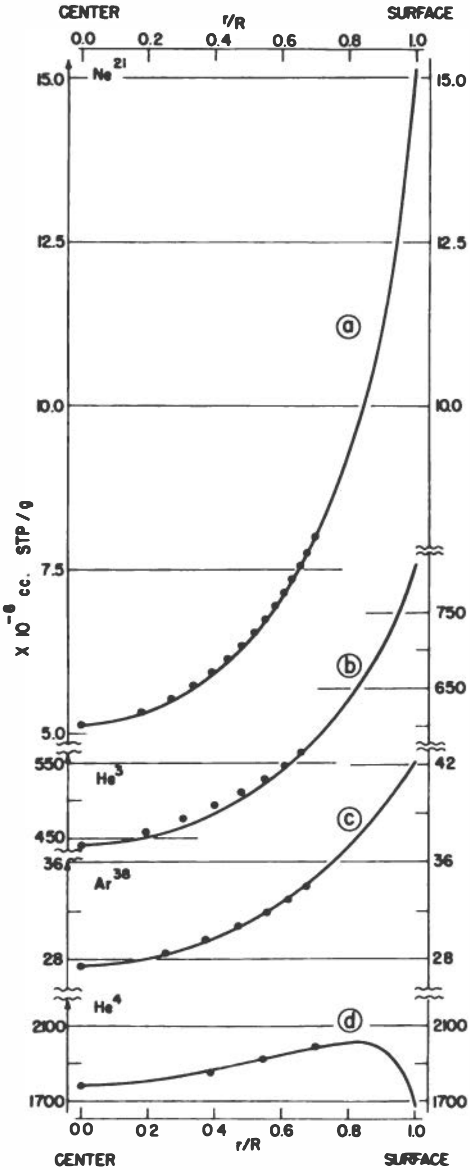

Figure 3. Computed radial increase (solid line) compared with experimentally determined increase (dots). Note the position of the dots, corresponding to the post-atmospheric surface, lying at r/R = 0.7.

The agreement of the values found here and those determined by Hoffman and Nier, as shown in Table 1, is satisfactory considering the limited precision of the whole procedure. Table 2 gives the ratios of the secondary to primary produced nuclides. It is interesting to note, that the secondary produced amount is biggest for $He^4$, less for $He^3$ and $Ar^{38}$ and negligible for $Ne^{21}$. This fact can be seen also from Figure 2, where the $Ne^{21}$ decreases most with depth, because of the insignificant secondary production. A comparison of production cross sections with results of target-studies may have only a limited validity. The values determined here are averages over the whole energy-range of the cosmic radiation, whereas targets are irradiated monoenergetically.

Having determined the parameters in equation (1) it is of interest to carry through the integration for some non-spherical bodies. Such a computation was made for prolate and oblate ellipsoids of revolution having masses of 2000 kg and a ratio of axes of 1:2. The calculations were made for points along the axis of symmetry. The results agreed closely with those for a spherical body having the same mass and indicated that the assumption of a spherical shape is a reasonable approximation.

A knowledge of the parameters also permits one to compute the relative amounts of nuclides produced by primary and secondary particles. Figure 4 shows the variation of the primary production (long dashes)



Copyright © National Academy of Sciences. All rights reserved.

TABLE 1

Comparison of Ratios of Production Cross Sections as Deduced from the Parameters*

| | $\sigma_{p3}/\sigma_{p4}$ | $\sigma_{s3}/\sigma_{s4}$ | $\sigma_{p3}/\sigma_{s3}$ | $\sigma_{p4}/\sigma_{s4}$ |
|---|---|---|---|---|
| Present Work | 0.67 | 0.13 | 4.8 | 1.0 |
| Hoffman & Nier | 0.50 | 0.14 | 5.0 | 1.4 |

* The production cross sections for nuclides of the species "i" for primary and secondary particles are designated by $\sigma_{pi}$ and $\sigma_{si}$, respectively.

TABLE 2

Relative Contribution of the Secondary-produced Nuclides

| $\sigma_{s3}/\sigma_{p3}$ | $\sigma_{s4}/\sigma_{p4}$ | $\sigma_{s21}/\sigma_{p21}$ | $\sigma_{s38}/\sigma_{p38}$ |
|---|---|---|---|
| 0.21 | 1.0 | 0.0 | 0.30 |

and the secondary production (short dashes) as well as the total concentration (solid line) along a radius in a spherical meteoroid having the mass of Grant (2000 kg).

The parameters determined in the fitting process can be used for further applications. One can, for example, calculate the way the concentrations of the four nuclides $Ar^{38}$, $Ne^{21}$, $He^4$ and $He^3$ vary in spherical bodies of different mass. Figure 5 demonstrates the decrease of the concentrations along a radius from the surface to the center of spherical meteoroids of $10^2$, $10^3$, $10^4$, $10^5$ kg and $\infty$ mass. The amounts were normalized to the respective amounts calculated to be found in infinitely small meteoroids.

The results of these calculations can be presented in different ways in order to analyze the variation of nuclide-ratios with depth in hypoththetical spherical meteoroids. One of the possible presentations is given in Figure 6. Here, the $He^4/Ar^{38}$ ratio is given as a function of the $He^4/Ne^{21}$ ratio along a radius in spherical meteoroids of $10^2$, $2 \times 10^2$,



Copyright © National Academy of Sciences. All rights reserved.

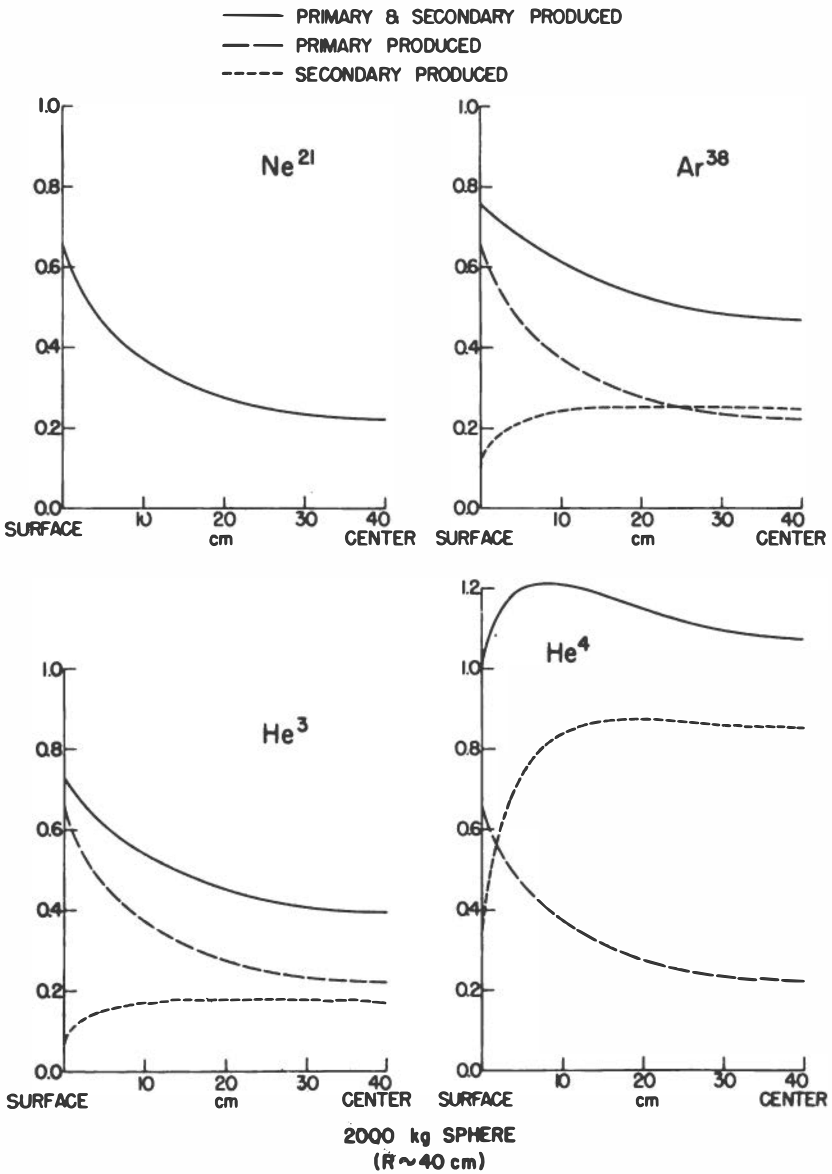


Figure 4. Contribution of the primary (long dashes) and secondary (short dashes) produced nuclides in a spherical meteoroid of 2000 kg mass, as computed with the parameters giving best agreement with measurements on Grant. The total amounts are given by the solid lines. The amounts are normalized to the respective amount in an infinitesimally small meteoroid.



Copyright © National Academy of Sciences. All rights reserved.

$2 \times 10^3$, $2 \times 10^4$, and $2 \times 10^5$ kg mass. The big dots on the ends of the curve at the lower left correspond to values found on the surface of the respective meteoroids whereas the big dots on the upper right end of each curve represent the value at the center. The small dots on the curves represent values at relative depths corresponding to r/R = 0.8, 0.6, 0.4, and 0.2 (surface 1.0; center 0.0). The crosses indicate seven arbitrarily chosen Grant samples. These are given to illustrate the agreement of the calculated curve ($2 \times 10^3$ kg) for Grant with the experimental data.

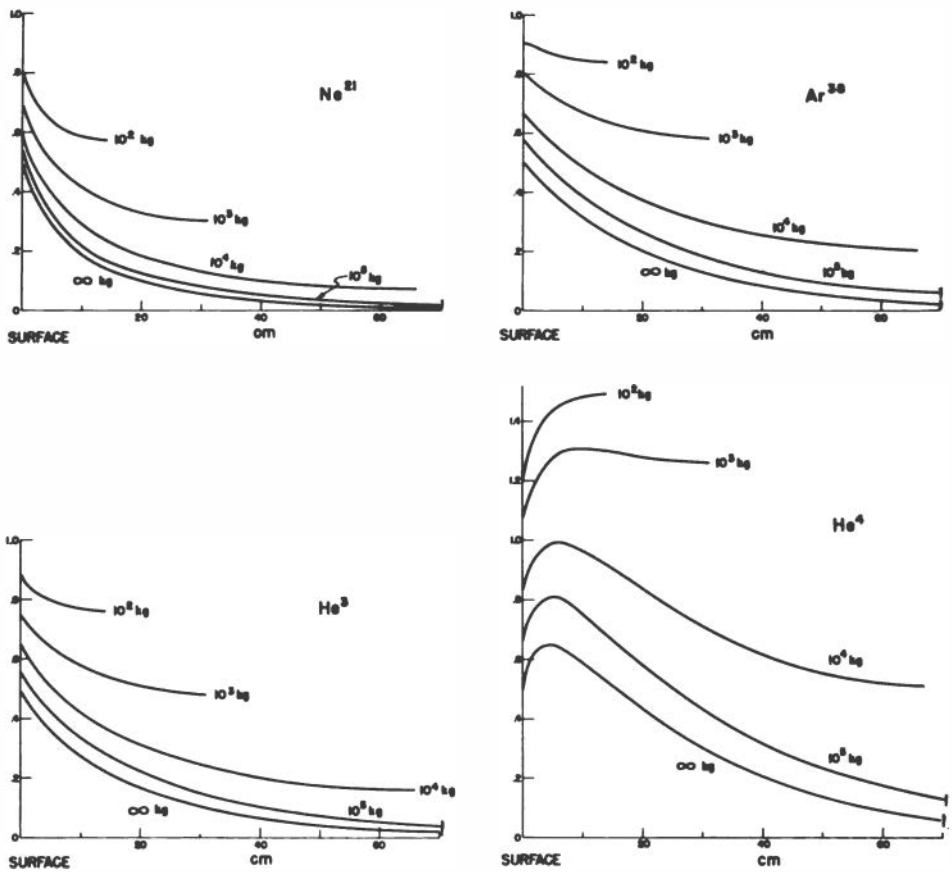


Figure 5. Radial variation of the concentration of the four isotopes $Ne^{21}$, $He^3$, $Ar^{38}$ and $He^4$ for spherical meteorites of different masses as predicted by the assumed production mechanism. The amounts were normalized as in Figure 4.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

According to the assumptions made, ratios found in meteorites of nearly spherical shape should lie close to the family of curves in Figure 6. A deviation can only be explained by a chemical composition for the particular meteorite different from that in Grant or by an irradiation having a very much different energy spectrum than that to which Grant was exposed. The latter explanation can, with good reasons, be excluded.

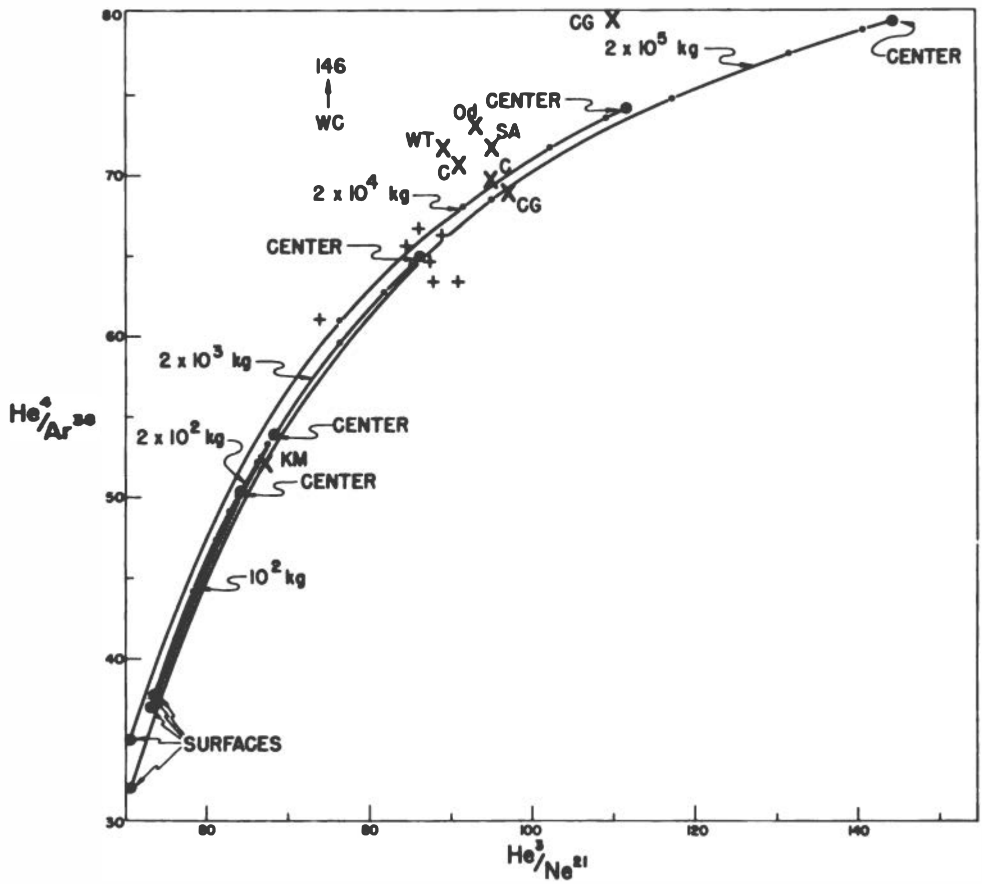


Figure 6. $He^4/Ar^{38}$ as a function of $He^3/Ne^{21}$ in spherical meteorites of different masses. Surface points lie at the left end of the curves, center points at the right end. Intermediate points correspond to r/R values of 0.8, 0.6, 0.4 and 0.2. Crosses (+) indicate some arbitrarily chosen samples of Grant and the symbols (X) represent some measurements of other iron meteorites.



Copyright © National Academy of Sciences. All rights reserved.

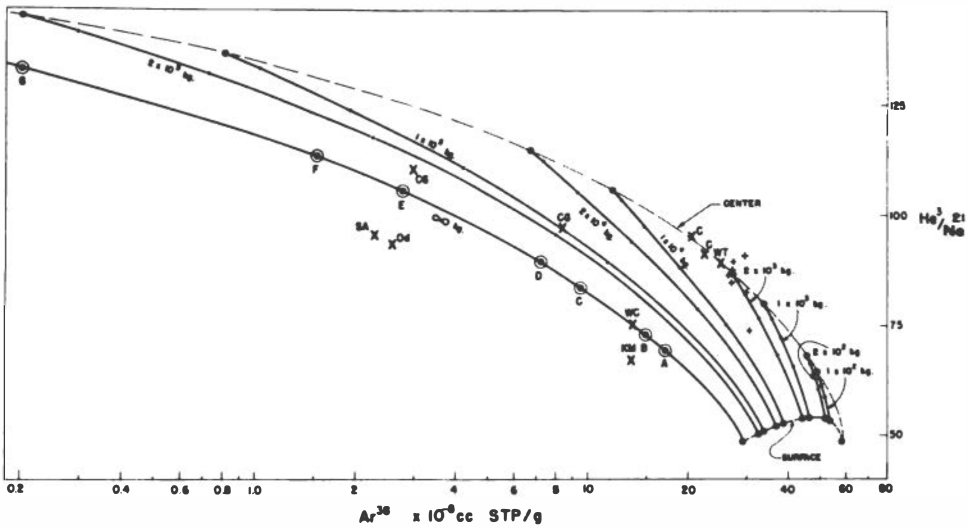


Figure 7. $He^3/Ne^{21}$ as a function of the $Ar^{38}$ concentration in spherical meteorites exposed to the same radiation (with respect to radiation dosage and energy spectrum) as Grant. The mass is used as parameter. Values found in the center lie on the dashed line designated as "center" and surface-values on the one designated "surface." The dots on the "mass-lines" correspond to depths with r/R values of 0.2, 0.4, 0.6 and 0.8. The crosses (+) and (X) have the same meaning as in Figure 6.

The symbol (X) shows some preliminary measurements on other meteorites, where the meaning of the designating letters is given in Table 3. With the exception of Washington County (WC) all the measurements lie within the experimental error, on the family of curves. We conclude from this fact, that excluding Washington County*, all the meteorites listed have a chemical composition similar to Grant.

Figure 7 is a graph useful for investigating the size and radiation dosage of a meteoroid as well as the position from which the analyzed sample was removed. The $He^3/Ne^{21}$ ratio is given as a function of the

---

* Schaeffer and Fisher (1959) have pointed out that Washington County has an exceptionally high $He^4$ content, probably resulting from U and Th decay. The present work points also to a high $Ne^{20}$ content which would suggest the presence of primordial gas in this meteorite. (The presence of Kr and Xe have not yet been investigated.) No primordial gas seems to have been reported for any other iron meteorite.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 3

Conclusions for Some Iron Meteorites Based on the Presented Model*

| Meteorite (& symbols used in Figs. 6 and 7) | Recovered Mass kg | Model-Mass kg | $Ar^{38}$-$He^{3}/Ne^{21}$ | | $Ne^{21}$-$He^{3}/He^{4}$ | | $Ar^{38}$-$He^{3}/He^{4}$ | | $He^{3}$-$Ne^{21}/Ar^{38}$ | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | | r/R | d | r/R | d | r/R | d | r/R | d |
| Williamstown (WT) | 54 | 5000 | 0.2 | 1.0 | 0.5 | 1.0 | 0.0 | 1.1 | 0.5 | 1.0 |
| Carbo (C) | 454 | 5000 | 0.3<br>0.0 | 1.0<br>1.0 | 0.3<br>0.0 | 1.0<br>1.0 | 0.2<br>0.0 | 0.9<br>0.9 | 0.4<br>0.1 | 1.0<br>1.0 |
| Washington County (WC) | 5.7 | 1000 | 0.5 | 0.5 | --- | --- | --- | --- | 0.3 | 0.4 |
| Keen Mountain (KM) | 6.7 | 800 | 0.2 | 0.3 | 0.0 | 0.4 | 0.0 | 0.4 | 0.0 | 0.3 |
| Casas Grandes (CG) | 1550 | $2x10^4$ | 0.6<br>0.3 | 0.7<br>0.4 | 0.7<br>0.5 | 0.5<br>0.3 | 0.7<br>0.5 | 0.5<br>0.3 | 0.6<br>0.5 | 0.6<br>0.3 |
| Odessa (Od) | >1000 | $2x10^5$ | 0.8 | 0.3 | 0.8 | 0.3 | 0.9 | 0.3 | 0.9 | 0.2 |
| Sikhote-Alin (SA) | $>3x10^4$ | $2x10^5$ | 0.8 | 0.3 | 0.8 | 0.3 | 0.8 | 0.2 | 0.8 | 0.3 |

* The "model-mass" is the pre-atmospheric mass indicated by the interpretation of the experimental data in the four graphs $Ar^{38}$-$He^{3}/Ne^{21}$ (given in Fig. 7), $Ne^{21}$-$He^{3}/He^{4}$, $Ar^{38}$-$He^{3}/He^{4}$ and $He^{3}$-$Ne^{21}/Ar^{38}$ (not given here). The radiation dosage d, relative to that of Grant and the relative depth r/R have been determined in the four graphs too. The comparison shows the precision of the interpretation.

Copyright © National Academy of Sciences. All rights reserved.

$Ar^{38}$ concentration in spherical meteoroids of different masses, assuming that all these hypothetical bodies were exposed to radiation similar (with respect to dosage as well as to energy distribution) to that irradiating Grant. Values measured at the center of spherical meteoroids should lie on the dashed curve designated with "center," whereas surface values are expected to lie on the curve "surface." The curves between two big dots indicate the variation of the ratio $He^3/Ne^{21}$ with the $Ar^{38}$ concentration in a meteoroid of a particular mass. The small dots represent again the values corresponding to a relative depth of $r/R = 0.8$; 0.6; 0.4 and 0.2. The solid line to the left gives the functional relationship in an infinitely large meteoroid. The points A, B, . . . G correspond to values found in this "meteoroid" at a depth equal to the radius of the $10^2$, $2 \times 10^2$, . . . $10^5$ kg meteoroids, respectively. The crosses represent the same Grant samples as in Figure 6 and indicate again the agreement of the experimental data with the calculation. As long as the radiation dosage and chemical composition of a meteoroid of any size is the same as for Grant, analysis of a sample of it should give rise to a point which lies in the restricted area shown in the figure.

These requirements seem to be fulfilled in the case of Carbo. Two samples were measured on Carbo, one removed from the radiation center and one close to the pre-atmospheric surface (Fireman 1958; Hoffman and Nier 1959). The two measurements lead to points (X, designated with C) well on the "mass-line" for a 5000 kg sphere, one at the center and the other at a relative depth of about $r/R = 0.3$.

Points, such as the ones representing measurements on Sikhote-Alin, Odessa or Keen Mountain, can be explained by assuming that their radiation dosage was smaller than that of Grant. Note that the point for Washington County is reasonably located in this graph, indicating that only the $He^4$ of the four isotopes investigated here is not explainable by purely cosmogenic origin.

Without any further information or assumptions, the graph shown in Figure 7 allows a determination of the radiation dosage with an uncertainty of about a factor of two. The use of similar graphs correlating other combinations of isotopes provides restricting criteria. Furthermore, logical reasons may be used to limit the uncertainty in the interpretation.

Table 3 gives some conclusions based on the graph in Figure 7 as well as three similar graphs, as indicated in the table. It should be remembered that these conclusions are all based on the many assumptions entering into the model. Future investigation may show if these assumptions were justified or if the model has to be modified.



Copyright © National Academy of Sciences. All rights reserved.

Epstein: Is there any difficulty in applying this method of analysis to Canyon Diablo?

Signer: No fundamental difficulty, except that the content of cosmogenic rare gases of some fragments of Canyon Diablo may be a little low to be measured with much accuracy. Many fragments of the meteorite are known, of course, and it would be interesting to see if they lead to a consistent picture in the model presented here.

Arnold: It would be tempting to apply the infinite radius model to Canyon Diablo.

Signer: Yes--the best tests of this method of analysis would be with samples of very large meteorites and of cosmic dust.

Anders: How would erosion of the meteorite in space affect the analysis?

Signer: One would have to combine curves for the concentration in objects of several different sizes. The result would be a flattening of the concentration curves, especially near the post-atmospheric surface.

[Anders, Arnold and Fireman were concerned about the disagreement between Signer's suggestion of a 12 cm loss of material from the reference axis during flight through the atmosphere and recent metallurgical arguments that this mass loss could not have exceeded 2 - 6 cm. On the basis of Signer's foregoing comment it has to be remembered that Signer's 40 cm preatmospheric radius for Grant is based on many assumptions as, for example, those of spherical shape and no alteration in the size during the irradiation. Therefore, if space erosion is indeed significant, no real disagreement may exist at all.]

Anders: It should be possible to employ this method of analysis to determine at last whether the cavities in iron meteorites are preatmospheric. Some of them are considerably larger than the typical scale of cosmic ray interaction (~15 cm).

Signer: A serious difficulty here is that the holes automatically cause drastic departure of the meteorite from the assumed spherical shape. The effects would also be considerably smeared by virtue of the isotropy of the flux.

Fish: The largest perturbation in the concentration contours would be expected at the very bottoms of the holes. A study of the shape of the contours near these holes might be revealing even if a detailed analysis were not practical.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

## REFERENCES

Ebert, K. H., and Wänke, H. (1957) "Ueber die Einwirkung der Hoehenstrahlung auf Eisenmeteorite," Z. f. Naturforsch. 12a, 766-773.

Fireman, E. L. (1958) "Distribution of helium-3 in the Carbo meteorite," Nature, 181, 1725.

Fireman, E. L. (1959) "The distribution of helium-3 in the Grant meteorite and a determination of the original mass," Planet. Space Sci. 1, 66-70.

Hoffman, J. H. and Nier, A. O. (1958) "Production of helium in iron meteorites by the action of cosmic rays," Phys. Rev. 112, 2112-2117.

Hoffman, J. H. and Nier, A. O. (1959) "The cosmogenic $He^3$ and $He^4$ distribution in the meteorite Carbo," Geochim. et Cosmochim. Acta, 17, 32-36.

Martin, G. R. (1953) "The origin of meteoritic helium and the age of meteorites," Geochim. et Cosmochim. Acta, 3, 288-309.

Schaeffer, O. A. and Fisher, D. E. (1959) "Cosmogenic noble gases in Washington County meteorite," Nature, 183, 660-661.

Signer, P. and Nier, A. O. (1960) "The distribution of cosmic-ray produced rare gases in iron meteorites," J. Geophys. Research (in press).



Copyright © National Academy of Sciences. All rights reserved.

# COSMIC RAY PRODUCTION OF RADIOACTIVE NUCLIDES IN IRON METEORITES

J. R. Arnold
School of Science and Engineering
University of California at La Jolla

Much new information is becoming available on the concentration of various long-lived radioactive nuclides (such as $Be^{10}$, $Al^{26}$, $Cl^{36}$, $K^{40}$, and $Mn^{53}$) produced by cosmic rays in iron meteorites. A number of short-lived species are also currently being measured in the recently fallen iron meteorite Aroos. With the aid of this information and of data on stable isotopes Dr. Honda, Dr. Lal, and I have tried to shed new light on the old problem of the time dependence of the intensity of the cosmic radiation. I am going to discuss some of this information and the problems which have arisen in its interpretation. The following table presents some data (Honda, Shedlovsky, and Arnold 1960) on the activity ratios of Be, Al, K, and Mn in four iron meteorites.

| Nuclide | $Be^{10}$ | $Al^{26}$ | $K^{40}$ | $Mn^{53}$ |
|---|---|---|---|---|
| Half-life | $2.7 \times 10^6$ y | $7.4 \times 10^5$ y | $1.3 \times 10^9$ y | $2 \times 10^6$ y |
| Grant<br>Absolute disintegration rate (dpm/kg) | 4.03±0.27 | 4.29±0.27 | 4.6±0.5 | 299±11 |
| Williamstown / Grant | 0.87±0.11 | 0.80±0.13 | 1.1±0.3 | 0.95±0.15 |
| Odessa / Grant | 0.51±0.04 | 0.32±0.04 | 0.4±0.15 | 0.66±0.03 |
| Canyon Diablo / Grant | 0.21±0.04 | 0.20±0.06 | | 0.31±0.04 |



Copyright © National Academy of Sciences. All rights reserved.

The samples are as follows:

Grant--saw cuttings from the cut through the meteorite which was made preliminary to the distribution studies on rare gases (Fireman 1959, Hoffman and Nier 1958). These distribution studies tell us that the 500 kg specimen we possess was a single individual in space.

Williamstown--a slice from the single 30 kg specimen recovered.

Odessa--an individual of this large meteorite fall.

Canyon Diablo--again a single small individual from a large fall. We expect that the production rates in these two meteorites will depend very much on the specimen chosen.

Our chemical procedures have been distinguished by the use of unusually large samples in order to permit comparatively accurate measurements, and by a serious and apparently successful effort to recycle to constant specific activity in each case. We have also been able because of the level of activity observed to apply certain auxiliary tests such as the measurement of beta absorption curves. The accuracy is of course still low compared to that obtainable in rare gas work, but some significant features are evident in the data:

1. The disintegration rates in Williamstown and Grant are practically alike for all activities. For the species $Be^{10}$, $Al^{26}$, $Mn^{53}$ this similarity may be taken to mean that both meteorites have built steady state abundances. Such an interpretation is manifestly not valid for $K^{40}$, which has a far longer half-life. Presumably the nearly equal potassium decay rates mean that Williamstown and Grant had approximately the same cosmic ray ages.

2. The ratios of the short-lived isotopes of Odessa/Grant and Canyon Diablo/Grant are all less than one, as one would expect for internal samples from large meteorites when they are compared to samples from a smaller meteorite ($K^{40}$ activities once again need not conform to the other ratios).

3. The absolute disintegration rate of $Mn^{53}$ is approximately two orders of magnitude higher than those of $Al^{26}$ and $Be^{10}$ in all cases.



Copyright © National Academy of Sciences. All rights reserved.

4. The ratio of $Mn^{53}$ activity to that of $Be^{10}$ or $Al^{26}$ is substantially higher in both Odessa and Canyon Diablo than in Williamstown or Grant.*

Of the models which have been developed to calculate production rates of isotopes in meteorites the most widely used is that of Martin (1953) and Ebert and Wänke (1957). Here the buildup of secondaries with depth was taken into account using the geometric interaction cross section of the primaries, and assuming a constant number of secondaries (about 3) per primary interaction. The cross section for the production of a particular species is determined at the average energy of the primary radiation (usually 3 Bev). This cross section is multiplied by the primary flux at a given depth. Cross sections at some average secondary energy (usually 300 Mev) are used to predict the production rate by secondaries. This method is satisfactory in making comparisons of production rates of isotopes which have no important production below 300 Mev. However, no plausible set of cross sections can be found in this model to account for the production of so much $Mn^{53}$ or other species close to the target nucleus as is actually observed. The physical basis of this model is admittedly primitive; its great advantage is convenience and simplicity.

Fireman and coworkers (1958) developed an ingenious method in connection with their studies of tritium and argon-37. A long iron bar is bombarded at various energies in high energy machines to deduce the production rate as a function of depth in iron and of energy, in a collimated beam. One then integrates the observed depth effect over angle, which yields a production rate as a function of depth in a meteorite. For accurate results this method would require integration over the primary energy spectrum. The most serious difficulty would appear to be the loss of secondaries which are emitted at large angles to the beam in the machine experiments. This is particularly important for low energy products.

Nuclear emulsion studies of the cosmic ray primary and secondary particles within the atmosphere are a potential source of pertinent information. Especially important are the experiments of Shapiro and coworkers (1951) who flew large blocks of lead of various shapes at balloon altitudes. They placed emulsions at different positions in the blocks and studied the star size distribution. The block which most closely approximated a sphere was about 15 cm in radius. Despite the fact that the

---

* It should be noted that the production of $Al^{26}$ by spallation of sulfur and phosphorus in an iron meteorite may be significant compared to the production from iron, because of the much higher cross section and lower energy threshold in these elements. The variation in the sulfur and phosphorus content may be a source of variation in the $Al^{26}$ data.



Copyright © National Academy of Sciences. All rights reserved.

dimensions were much in excess of the mean interaction length neither the total number of stars nor the star size distribution changed very much with depth. Flatter shapes showed depth effects, but the change with depth was not great. This is particularly striking since the primary flux comes only from above in these cases. These results would not be predicted either by the Martin model or by the use of the iron bar target data. Admittedly they should be checked with iron blocks of various dimensions, but they do seem consistent with the fact that one keeps finding small meteorites which all show about the same radioactivity level.

Many studies have been made of star size distributions in emulsions in the free atmosphere at various depths. Measurements of this type have also been made using mono-energetic beams of protons between about 100 Mev and 6 Bev. From a combination of these two sets of data it is in principle possible to deduce the energy spectrum of primary and secondary particles at any atmospheric depth. In practice this method is not capable of high precision, but the differential spectrum deduced is probably not in error by as much as a factor of two at any energy, and should be more accurate than this when broad energy ranges are considered. Other information such as the primary energy spectrum and the secondary production spectrum of Camerini et al (1950) are helpful in inferring the functional form of the distribution.

Shapiro's studies show that the total star production in emulsion inside a lead block is enhanced over that in emulsion in the free atmosphere, but that the shape of the energy distribution is quite similar. The same should be true for iron.

The energy distribution thus determined may be used together with the excitation functions for various nuclides to predict relative and absolute production rates. By measurement of short-lived isotopes from freshly fallen meteorites it is possible to check the validity of this technique under conditions where the cosmic ray intensity is known to have been constant within the accuracy of the predictions (at present no better than 30 percent). The abundance ratios of stable isotopes or of pairs of long lived isotopes of similar half-life may also be used to check the technique.

Our current program includes the measurement for this purpose of numerous short-lived isotopes in the meteorite Aroos.

The higher ratio of $Mn^{53}$ to $Al^{26}$ observed in Odessa and Canyon Diablo is easy to understand on the basis of such a model. Since $Mn^{53}$ is a "low energy nuclide" and $Be^{10}$ and $Al^{26}$ are "high energy nuclides" the ratio should be elevated as the energy spectrum is degraded. The lower



Copyright © National Academy of Sciences. All rights reserved.

production rates in Odessa and Canyon Diablo would lead one to expect that they represent greater depth; thus the results seem entirely reasonable.

This new model seems to represent a useful advance over earlier ones. Presumably it is capable of being made exact as energy distributions and excitation functions are improved. Preliminary calculations for the production rates of radioactive and stable isotopes are in gratifying agreement with experiment. The agreement is gratifying, that is, if the ablation of a meteorite like Grant was of the order of a few centimeters or less. But if one assumes an ablation of 12 cm in Grant (see Signer's paper in this connection) then the theoretical production rates are low by factors of the order two. We cannot yet prove that this discrepancy is significant.

Within the accuracy of these preliminary calculations the cosmic ray flux has not altered significantly over the lifetime of species such as $Be^{10}$, $Al^{26}$, $Cl^{36}$, and $Mn^{53}$. In the case of $K^{40}$ a steady state abundance would not be expected since the apparent cosmic ray ages of the meteorites so far studied are less than $10^9$ years. From the point of view of the constancy of the cosmic ray flux, $K^{40}$ is by far the most interesting isotope. Its usefulness would be enhanced if meteorites of cosmic ray age more than one billion years could be found. The longer the time over which a meteorite has actually been bombarded, the greater the difference between the behavior of $K^{40}$ and that of a stable species.

Erosion in space, continued over the lifetime of the meteorite, can also be studied using radioactive isotopes. It seems obvious that not enough erosion can have occurred in the iron meteorites studied over the lifetimes of the million year species to seriously alter their activity ratios. In the case of $K^{40}$ one might expect to see signs of an apparently increasing flux with time. It would be difficult but not impossible to distinguish this from a true variation in cosmic ray intensity with time.

## REFERENCES

Camerini, U., Fowler, P. H., Lock, W. O., Muirhead, H. (1950) Phil. Mag. 41, 413.

Ebert, K. H., and Wänke, H. (1957) Z. Naturforsch 12a, 766.

Fireman, E. L. (1958), in "Cosmological and Geological Implications of Isotope Ratio Variations," Publication 572 of the National Academy of Sciences—National Research Council (Washington, D. C.).



Copyright © National Academy of Sciences. All rights reserved.

Fireman, E. L. (1959) Planet. Space Sci. 1, 66.

Hoffman, J. H. and Nier, A. O. (1958) Phys. Rev. 112, 2112.

Honda, M., Shedlovsky, J. P., and Arnold, J. R. (1960) Geochim. et Cosmochim. Acta (in press).

Martin, G. R. (1953) Geochim. et Cosmochim. Acta 3, 288.

Shapiro, M. M., Stiller, B., Birnbaum, M., and O'Dell, F. W. (1951) Phys. Rev. 83, 455. (See also Phys. Rev. 86, 86 and 84, 160).

Shapiro, M. M. (1951) Phys. Rev. 83, 456.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# COSMOGENIC NUCLIDES IN THE HAMLET METEORITE

Anthony Turkevich
Enrico Fermi Institute for Nuclear Studies
University of Chicago

The Hamlet meteorite, which fell at Hamlet, Indiana, October 13, 1959, is a small (~2.5 kg) chondrite, apparently quite ordinary, of roughly pyramidal shape with an approximately square base. Gross gamma-ray spectra were taken on a portion of the whole meteorite, using a 3" x 3" crystal and 200 channel analyzer. In the spectra taken two weeks after infall, a 500-Kev peak and an 800-Kev peak were clearly discernible, along with a number of less prominent peaks. Gross spectra taken later indicate that the 800-Kev peak decays appreciably with time, while the 500-Kev peak is essentially constant.

Under the assumption that the point of the pyramid defines the front of the meteorite during infall, analyses of various cosmogenic nuclides were made and are in progress on material from the base, presumably near the original surface. Preliminary data is available on 16-day $V^{48}$, 26-day $Cr^{51}$, the sum of 77-day $Co^{56}$ and 71-day $Co^{58}$, 267-day $Co^{57}$, ~300-day $V^{49}$, and 300-day $Mn^{54}$ activities. The "absolute" activities for the V, Cr, Co and Mn nuclides mentioned range from about 5 to about 100 dpm/kg. The tentative nature of this data does not warrant a detailed listing of the numbers, which will be published later in final form, but some comments on ratios of activities may be made.

To facilitate our understanding of the production of the cosmogenic nuclides in Hamlet, a proton irradiation of a 5.2 g sample of the meteorite was performed at the Brookhaven cyclotron, using the external beam. The sample was mounted directly behind a six-inch thick block of iron; a smaller block of iron was placed behind the sample. According to $Na^{24}$ activity produced in Al foils, a dose of $\sim 10^{14}$ protons, nominally 3 Bev in energy, was received by 'hot' Hamlet. The sample was then fused in the course of extracting noble gases, so that the silicate and iron phases were separated. The gamma-ray spectra of each of these phases were examined, and identifications of the various peaks observed were attempted. It appears that all the Co activities are in the iron phase, although a small fraction of these nuclides must have been formed from oxidized iron in the silicate phase. Peaks assignable to Cr were seen in both phases, indicating a partitioning of this element, while an intense peak, presumably in



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

large part due to annihilation radiation from $Na^{22}$ and to $Be^{7}$ gamma radiation, was observed in the spectrum of the silicate phase. Apparently, extraction of trace elements into phases appropriate to their lithophilic or siderophilic character was reasonably complete during fusion of the sample.

If one compares the activity ratio of $Co^{57}/(Co^{56} + Co^{58})$, corrected to time of fall, in Hamlet with the same ratio in the 'hot' Hamlet, an interesting discrepancy comes to light. This activity ratio in the cosmic-ray irradiated specimen is about 1.3 times that in the sample irradiated in the Brookhaven cyclotron. If the cyclotron bombardment represents a valid duplication of the conditions of cosmic-ray irradiation (see the paper by Schaeffer in this volume for comments on this assumption), one would expect that, for a constant long-term cosmic-ray bombardment, the ratio in Hamlet should be ~4 times the value in 'hot' Hamlet. This conclusion is derived from a simple consideration of the effect of saturation of the activity levels.

There are a number of ways of interpreting this discrepancy. Perhaps the most likely hypothesis is that the cyclotron irradiation, for one or another of the reasons discussed by Schaeffer, is <u>not</u> a good "mock-up" of the cosmic irradiation. Another possibility, however, is that the cosmic irradiation of Hamlet was more nearly an instantaneous irradiation than a continuous one. Before any serious discussion of these hypotheses can be undertaken, much more data must be gathered. Note especially that the production of Co isotopes from Fe or Ni has a low energy-threshold, so that production by secondaries may be important. Also, the effect of working with material which was presumably quite near the pre-atmospheric surface should be given detailed consideration. Finally, the cosmogenic Co isotopes may have been in large part produced by meson interactions with Ni, which would make comparison with the cyclotron irradiation even more questionable.

Nevertheless, it is clear that this approach is potentially capable of yielding important information on cosmic-ray fluxes and their variations with time and in different regions of the solar system.



Copyright © National Academy of Sciences. All rights reserved.

# THE HIGH ENERGY COSMIC RAY SPECTRUM

S. Olbert
Department of Physics
Massachusetts Institute of Technology

In the cosmic ray spectrum above about two Bev it is meaningful to set down a single energy spectrum as typical: in contrast to the wildly fluctuating energy spectrum at lower energies, which will be discussed by Meyer and by Van Allen, the spectra above two Bev are stable within about 20 percent. The energy dependence of the high energy cosmic ray spectra for particles of different charge are shown in the following table:

| Designation | Charge Z | J(E) (no/m$^2$/ster/sec) | Range of Validity (Bev/nucleon) | Notes |
|---|---|---|---|---|
| P | 1 | 4000 $E^{-1.15}$ | 2 - 20 | Steepens gradually: $E^{-2}$ > $10^6$ Bev. No cut-off at $10^{18}$ ev. |
| α | 2 | 460 $E^{-1.6}$ | 1.5 - 8 | |
| L (Li, Be, B) | 3, 4, 5 | ~50% of M? | --- | Difficult to measure--see text. |
| M (C, N, O, F) | 6, 7, 8, 9 | 24 $E^{-1.6}$ | 3 - 8 | Little is known about spectrum at higher energies. |
| H | ≥ 10 | 16 $E^{-2.0}$ | 3 - 8 | |

For protons many detecting devices have been employed: Geiger-Müller counters, proportional counters, ion-chambers, Cerenkov counters, cloud chambers, etc. At low energies the Earth's magnetic field provides a spectrometer. At higher energies the cosmic ray events are too rare to be detected directly, so scintillation counters are employed to detect electrons produced in the air shower. In this case, the number of electrons passing any level of the atmosphere provides a good measure of the energy of the primary without too much dependence on the model of multiple meson production which is assumed. [The discussion of measuring techniques will



Copyright © National Academy of Sciences. All rights reserved.

not be given in as much detail as OLBERT provided--those readers interested in techniques may consult review articles such as that of Singer (1958) ].

It is important to note that there is no indication of any cut-off in the proton cosmic ray spectrum, even at $10^{18}$-$10^{19}$ ev; this point will be of importance later. The errors probably do not exceed 5 - 15 percent in the spectral region from about 3 - 20 Bev and at the highest energies the errors are probably not more than a factor of two; in between there are some gaps which are not as precisely defined.

Heavy and medium weight particles in the 2 - 20 Bev range have been studied almost exclusively by means of photographic emulsion techniques. One attempts to identify the particles from their tracks--by delta-ray counting for example--but it must be recognized that the mass spectrum of particles is undoubtedly modified by fragmentation in the residual atmosphere above the detecting station. Various authors have published relative abundances of C, N, and O nuclei. Some of their results are quoted below, along with the Suess-Urey cosmic abundances and Aller's (1960) latest solar abundances:

| C | N | O | |
|---|---|---|---|
| 24 | 16 | 6 | Cosmic Rays (various authors) |
| 27 | 32 | 14 | |
| 39 | 9 | 37 | |
| 41 | 4 | 8 | |
| 31 | 25 | 24 | |
| 1 | 2 | 6 | Suess-Urey |
| 9.3 | 2.4 | 25 | Aller |

The cosmic-ray results are in fairly good agreement with one another. They diverge considerably from the Suess-Urey cosmic abundances, particularly in the C:N ratio, but are not appreciably at variance with Aller's solar abundances. [Aller notes that the f-value for oxygen is not well known, and that better f-values and model atmosphere calculations are required for carbon.] At higher energies little is known about relative abundances or energy spectra of the medium and heavy weight nuclides due to the scarcity of such events. It does appear, however, that the total medium-weight particles are about 2-1/2 to 3 orders of magnitude less abundant than protons, and that the heavy weight nuclides are about 3-1/2 orders of magnitude less abundant throughout the energy range from 1 Bev up to a few hundred Bev. [No significant disagreement with either the Suess-Urey or Aller abundances.]

The greatest uncertainties, both in energy spectrum and identification, are at Z = 3, 4, 5. First of all, these particles are far harder to



Copyright © National Academy of Sciences. All rights reserved.

detect on emulsions than the heavier particles. Secondly, so few of these particles are detected that they could all have been produced by fragmentation of heavier nuclei in the atmosphere; it is still an open question whether there are any Li, Be, B nuclei in the primary cosmic-ray flux at all, although current opinion favors the existence of a primary ratio of L to M particles of perhaps 50 percent. These nuclei are especially worthy of attention when cosmic-ray spectrometers are carried above the atmosphere. If nuclei are also sought which are almost exclusively fragmentation products--$H^2$, $H^3$, $He^7$--it should be possible to settle the debate on whether the light- and medium-weight cosmic ray nuclei are really only fragments of, say, iron. It is also important, of course, to try to improve air-shower detection systems so that nuclear interactions with μ-mesons can be studied, and so that other components of showers can be studied which appear with lower fluxes than the electrons.

Time delay circuits set up in connection with the numerous scintillation counters used in air shower research are used to define the angle of attack of primary cosmic rays. By plotting angles of incidence vs. sidereal time it has been possible to seek point sources of cosmic rays in the galaxy with better than $5^\circ$ precision. Except for anisotropy in the direction of the sun, however, the galactic cosmic ray flux appears to be extremely isotropic. Cocconi (1956) has found that the degree of anisotropy δ, where

$$\delta = \frac{\text{Maximum Galactic flux - Minimum Galactic Flux}}{\text{Average Flux}}, \qquad (1)$$

is less than $10^{-3}$ for particles up to $10^{15}$ ev and less than 0.01-0.02 for particles up to $10^{18}$ ev.

There is a possibility of gaining valuable information on the structure and composition of our Galaxy by looking for pure electron-photon showers caused by energetic γ-rays that arose when a high energy particle struck quiet matter at some distant point in the Galaxy. These pure photon showers would be free of all particles but electrons and photons because the probability of photonuclear reactions is truly negligible compared to ionizations. They could easily be detected by circuits set to detect electrons in anticoincidence with nucleons. The frequency of these showers would serve to measure the interstellar galactic density, since the production probabilities are reasonably well known. The γ-ray flux, unlike the ordinary cosmic ray flux, should be highly anisotropic; it should be largely confined to the dish of the Galaxy and concentrated toward the galactic center. [Information on material densities in the galactic center would be especially valuable, since it is in this region that dynamical determinations of the mass density have lowest weight. The interest in this problem is heightened by the recent observation that neutral hydrogen is streaming from the galactic nucleus (van Woerden, Rougoor, and Oort 1957).]



Copyright © National Academy of Sciences. All rights reserved.

Certainly this technique could provide some important information not available to radio or optical astronomy.

The origin of cosmic rays is not yet settled. Certainly the sun cannot be producing the high energy flux since particles of more than $10^{13}$ ev would not be deflected enough by magnetic fields within the solar system to appear as an isotropic flux. Rather it appears that particles are ejected into interstellar space by various objects--flare stars, novae, supernovae, etc.--and then accelerated by collisions with hydromagnetic waves "magnetic clouds" in the galactic magnetic field (Fermi, 1954). Acceleration is the result of these collisions because head-on collisions, which transmit energy to the particles, are statistically favored compared to overtaking collisions, which remove energy.

The general form of the acceleration equation is

$$\frac{dE}{dn} = \alpha E - \beta(E) \qquad (2)$$

where n is the number of collisions, $\alpha$ is a parameter describing the efficiency of acceleration ($\alpha$ is of order $k\frac{V^2}{C^2}$ where k = 4/3 for a random velocity distribution of ionized particles in a gas cloud) and $\beta$ is a collision-loss parameter which decreases with increasing energy. It is easy to understand why a power law spectrum results from such an equation if injection is continuous, and it is also plain that the highest energy particles will be the oldest. Variations in wave structure, relative velocities and wave spacing, magnetic field topology, damping parameters and the energies of injected particles have been employed to explain the observed energy spectrum for primary cosmic rays. The isotropy of the flux can also be explained as a result of the trapping of cosmic ray particles by the Galactic magnetic field and the resulting diffusion of the particles throughout the galactic halo.

Nonetheless this picture is incomplete in several respects. First, it is clear from equation (2) that a critical energy of injection is required before the Fermi acceleration mechanism can begin to be effective. For protons the critical energy is only 1 Mev, a reasonable enough injection energy, but for iron nuclei the critical energy for acceleration is 400 Mev, which poses a real injection problem. Second, since the index of the power law spectrum of cosmic rays is known, the parameter $\alpha$ is fixed; the resulting value of $\alpha$ demands relative magnetic cloud velocities the order of 100 km/sec. Such relative velocities are about ten times larger than would be anticipated on other grounds. Third, cosmic ray particles with energies in excess of $10^{18}$ ev cannot be trapped even in the galactic magnetic field, so apparently their source is extra-galactic. (Galaxies may simply exchange between themselves particles which they cannot trap.)



Copyright © National Academy of Sciences. All rights reserved.

There is no obvious reason why the spectrum of particles above this critical energy for galactic trapping should fit smoothly to the spectrum of trapped particles, but it does.

Other theories of the origin of cosmic rays have been proposed; Ginzburg (1958), for example, suggests that the cosmic ray flux is a result solely of supernovae acceleration and diffusion throughout the galactic halo; this averts the first difficulty mentioned above but not the second and third, and it introduces many problems of its own (such as an expected cutoff in the proton spectrum at $10^{15}$ ev). But in any event it is challenging to realize that in studying the cosmic ray energy and mass spectra we are gaining valuable information about element synthesis and stellar evolution not only from our own Galaxy, but from other galaxies as well.

Cameron: It would be worthwhile to seek γ-rays from M31. Our Galaxy would not interact with these γ's; M31 is massive enough and subtends a large enough angle in the sky to favor detection as a discrete γ-ray source.

## REFERENCES

Aller, L. H. (1960) Chemical Composition (Interscience Press, New York).

Cocconi, G. (1956) Il Nuovo Cim. 3, 1433.

Fermi, E. (1954) Astrophys. J. 119, 1.

Ginzburg, V. L. (1958) Progr. Elem. Particle and Cosmic Ray Physics IV, 339.

Singer, S. F. (1958) Progr. Elem. Particle and Cosmic Ray Physics IV, 205.

van Woerden, H., Rougoor, G. W., and Oort, J. H. (1957) Compte Rendus Acad. Sci. Paris 244, 1691.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# LOWER ENERGY COSMIC RAYS AND THE SOLAR CYCLE

P. Meyer
Enrico Fermi Institute for Nuclear Studies
University of Chicago

The low energy portion of cosmic radiation (from 100 Mev up to several Bev), which may produce spallation nuclides in meteorites, is strongly influenced by solar phenomena. On the one hand one does observe striking changes in the flux and energy spectrum of the galactic cosmic radiation which are correlated with the 11-year solar activity cycle or with single solar flares (Forbush decreases), and on the other hand one observed occasional solar emission of high energy particles with an energy spectrum quite different from that of the galactic component. We wish to discuss briefly these two phenomena, namely (1) solar modulation of galactic cosmic radiation, and (2) solar emission of high energy particles.

Let us consider first the general changes occurring during the 11-year solar cycle. During years of solar minimum the differential spectrum of the isotropic galactic proton flux closely follows a power law, with

$$N(E)dE = \text{const. } E^{-2.7}\, dE. \tag{1}$$

The range of validity of this relation extends from about 1 Bev up to 20 to 30 Bev at least; no low-energy cutoff seems to be present but the exact shape of the spectrum is not known for the lower energies. During the active phase of the Sun, however, the galactic proton flux is reduced by a factor of about two for energies above 1.5 Bev, and follows a flatter power law distribution, namely

$$N(E)dE = \text{const. } E^{-2.0}\, dE. \tag{2}$$

At solar maximum there is a distinct low-energy cutoff: the number density of protons drops rapidly below 1 Bev. The two cases are illustrated in Figure 1, necessarily somewhat approximately since the information at hand really applies only to the present solar cycle.

It is widely believed today that during the active phase of the solar cycle a steady stream of ionized particles of density near 100 particles/ $cm^3$ travels outward from the Sun at velocities the order of 500-1000 km/ sec and is responsible for this cosmic ray modulation. The kinetic energy of the particles in this "solar wind" would be only the order of $10^3$ ev, far



Copyright © National Academy of Sciences. All rights reserved.

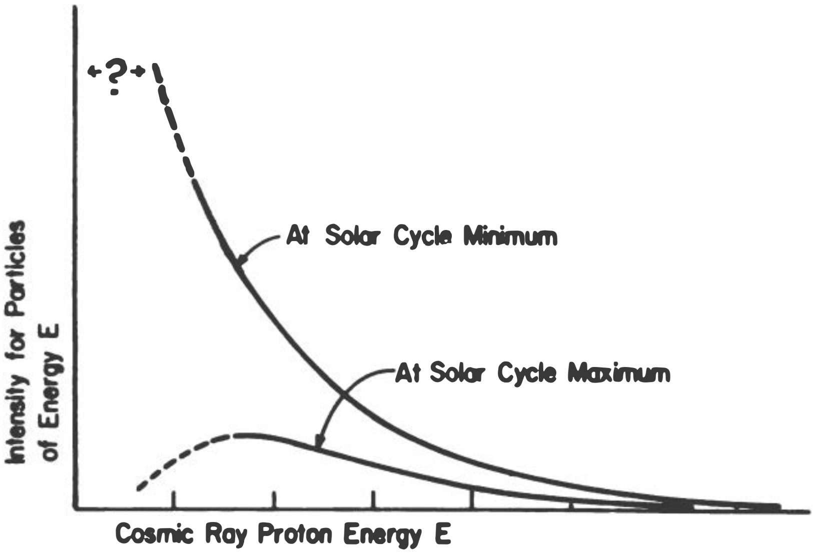


Figure 1.

too small to produce nuclear effects, but nonetheless capable of producing important modulation effects on the interplanetary magnetic fields, and through this agent affecting the trajectories of all ionized particles (including cosmic rays) in the solar system. We shall return to this point.

The cosmic ray phenomena associated with solar flares are of two different types, production and modulation. The Sun emits clouds or bursts of particles at the time of a flare. Most small flares (Class 1), which may appear at the rate of several per day at peak activity, probably confine themselves to ejecting relatively low energy streams of particles. Somewhat larger flares (Classes 2 and 2+) may rarely emit particles with energies extending up to perhaps 300-400 Mev. The energy spectrum of these particles appears to follow approximately an $E^{-4}$ law. This spectrum was observed by balloon experiments. The much rarer flares of intensity Classes 3 and 3+ are known to occasionally emit particles up to about 20 Bev. During one such event (February 1956) the energy spectrum of the high energy particles was measured to go roughly as $E^{-6}$.

On the basis of radionoise absorption data and several months of satellite and space probe observations one may estimate the frequency of the occurrence of solar particle emission during the past maximum of solar activity. It appears that, in this phase of the solar cycle, the Sun emits particles with energies up to 400 Mev once or twice a month. The emission of relativistic protons seems to occur about once every four years and no obvious correlation with the phase of the solar cycle has been found for such events.



Copyright © National Academy of Sciences. All rights reserved.

The bulk of low energy particles emitted in a solar flare usually has a velocity of 1000-2000 km/sec, which results in a transit time from Sun to the Earth of about a day or two. This solar plasma is assumed to be the cause of the Forbush decreases in galactic cosmic ray intensity and of geomagnetic effects which often follow an intense flare. The low-energy galactic cosmic ray flux during a Forbush decrease will show a sharp drop which in some cases may amount to as much as 25-50 percent. The recovery is much slower, and appears to take place approximately exponentially over the next few days or weeks.

A number of theories have been proposed to account for various of these effects which we have noted. One which is unusually comprehensive in scope is that of Eugene Parker at the University of Chicago. Parker notes that the high temperature of the solar corona may extend several solar radii into space, with the result that the corona expands hydrodynamically with supersonic velocity into space. The radially-directed gas can therefore extend the magnetic lines-of-force of the general solar field in the radial direction. These radially-directed fields can be expected to extend for many astronomical units outward from the Sun, and with the rotating motion of the Sun imposed on them they should appear as spirals. Parker shows that this hydromagnetic system leads to the well known nose instability beyond the orbit of Earth. In the instability region the lines-of-force are highly disordered and form a diffuse barrier that shields the inner portions of the solar system from some of the lower energy galactic cosmic radiation. In other words, Parker's theory suggests a reason for the reduction in the galactic cosmic ray flux during the active phase of the 11-year solar cycle, and it explains the return of the galactic flux to normal during the quiet phase by the absence (or very great reduction) of the solar wind in the inactive part of the solar cycle.

The role of a solar flare in producing the Forbush decrease, in Parker's view, is to eject a stream of ionized particles into the spiral lines-of-force which wind outwards from the Sun. Since the ejection is purely radial, the ions impart some transverse energy to the spiral magnetic field which is propagated along the lines-of-force as a shock wave. This shock wave has a radial velocity of about 2000 km/sec. It reaches the Earth about a day after the onset of the flare and continues beyond. As it passes, a sharp decrease in galactic cosmic ray intensity occurs (the Forbush decrease), due to the fact that incoming charged particles in the Mev to several Bev range cannot penetrate the shock barrier, and the galactic cosmic rays continue to be excluded until the shock waves have finally been damped out by particle collisions over a period of days or weeks. A slight increase in galactic cosmic radiation prior to the onset of the Forbush decrease is expected in this model and has been noted several times. Note that the stationary barrier which affects the galactic cosmic radiation throughout the active phase of the solar cycle acts



Copyright © National Academy of Sciences. All rights reserved.

efficiently only on particles with less than ~5 Bev energy. The shock barrier in Parker's model has quite different characteristics which may explain the energy range affected by the Forbush decreases.

There are a few points in Parker's theory which are of special importance in relating the cosmic ray flux to nucleogenesis in the meteorites. First, the meteorites would be expected to receive a full flux of galactic cosmic radiation during whatever time their orbits were outside the instability barrier, regardless of the phase of the solar cycle. When the meteorites are inside this barrier they receive a normal flux of galactic radiation during the quiet phase of the Sun and a reduced flux of radiation below 3-4 Bev during the active phase. The more energetic galactic flux is not affected by the solar cycle (see Olbert's paper). Second, the "brightness" of the solar particle emission after flares does not depend on the distance of the meteorite from the Sun, so long as it is within the instability zone (the solid angle which the Sun subtends does depend on the distance, of course). The instability barrier probably reflects flare particles and accounts for the observed storage in the inner solar system which may last several days.

It is probably premature to do too much speculation about the exact nature of these effects. It will be very nice, however, to try to correlate the theory with observations when we have a space probe travelling in or beyond the zone of instability.

Arnold: In estimating the effects of the solar modulation of the cosmic ray primaries on the production of nuclides in a meteorite, it is important to know how much material has been removed from the surface. If very much ablation has occurred, then perhaps none of the cosmogenic nuclides we see were produced by primaries below about 1 Bev.

Olbert: There is no doubt about the existence of the sort of instability which Parker postulates, provided a stream of ionized particles does flow from the Sun during the active phase. Any time we have a dynamical motion of ionized gases with magnetic fields frozen in, the pressure is anisotropic and instabilities can arise. Yet the analysis is essentially dimensional in character, and the position of the instability (predicted to be near the orbit of Mars) is probably uncertain by a factor of two.

Kohman: Where does the acceleration of the "solar wind" arise?

Meyer: This is based on the experimental temperature distribution of the corona--essentially the solar wind is a steady-state dynamical expansion of a hot gas. The problem is hydrodynamic in nature and does not depend on magnetic forces. Nonetheless, there are some ambiguities



Copyright © National Academy of Sciences. All rights reserved.

in the solutions of the hydrodynamic equations, so that the solar wind concept is by no means universally accepted.

Fish: Are the lines-of-force which extend radially in Parker's model somehow connected back to the Sun in order to satisfy div **B** = 0? If so, any shock waves propagating outward would also eventually present a shock front directed inward, toward the Sun, and should result in anisotropies in the galactic cosmic ray flux some days after a flare.

Van Allen: It is not strictly necessary that the lines-of-force be closed for the divergence of **B** to vanish, provided the lines-of-force constitute a sufficiently complicated network.

Fish: It might be possible to apply a critical test to Parker's model for cosmic-ray modulation by studies of the occasional large solar flares which occur during the quiet period in the solar cycle. If Parker's theory is correct there should be much smaller Forbush decreases following these flares than would be the case for flares during the active period.



Copyright © National Academy of Sciences. All rights reserved.

# ON THE GEOPHYSICAL AND GEONUCLEAR SIGNIFICANCE OF THE EARTH'S RADIATION BELTS

James A. Van Allen
State University of Iowa

## Introduction

An immense region around the earth has been found to be populated by energetic protons and electrons, temporarily trapped in periodic orbits in the earth's magnetic field [Van Allen, McIlwain and Ludwig (1959), Vernov, Chudakov, Vakulov and Logachev (1959), Van Allen and Frank (1959a), (1959b), Van Allen (1959)]. The region is found to be subdivided into two distinctly different belts or zones--the inner zone and the outer zone. An annular "slot" or region of minimum intensity of charged particles separates the two regions of high intensity. (Figure 1.)

During the some two and a half years (February 1958 to present) since discovery of this phenomenon with the Explorer I satellite, a large variety of experimental and theoretical work has been conducted by workers in the United States and Russia. Salient features of present knowledge are as follows:

1. The intensity-structure of the inner zone is stable in time to a known accuracy of some 20 percent.

2. There are enormous temporal fluctuations of the intensity-structure of the outer zone. The intensity of radiation at a specified point in space has been observed to vary by as much as three orders of magnitude (1000/1) as measured with a given detector. Also the outer zone often exhibits a complex and time-varying detailed spatial structure. These fluctuations are, broadly speaking, correlated with geomagnetic storm activity and hence with solar corpuscular radiation.

3. A typical time history of the outer zone on the occasion of a magnetic storm comprises a marked "dumping" or depletion of detectable radiation accompanied by aurorae, and enhanced atmospheric airglow, recovery of the intensity with a characteristic time of the order of a day to a value exceeding (perhaps by manyfold) its pre-storm value, then gradual relaxation toward a quasi-steady value characterizing the general period of time of the observations.



Copyright © National Academy of Sciences. All rights reserved.

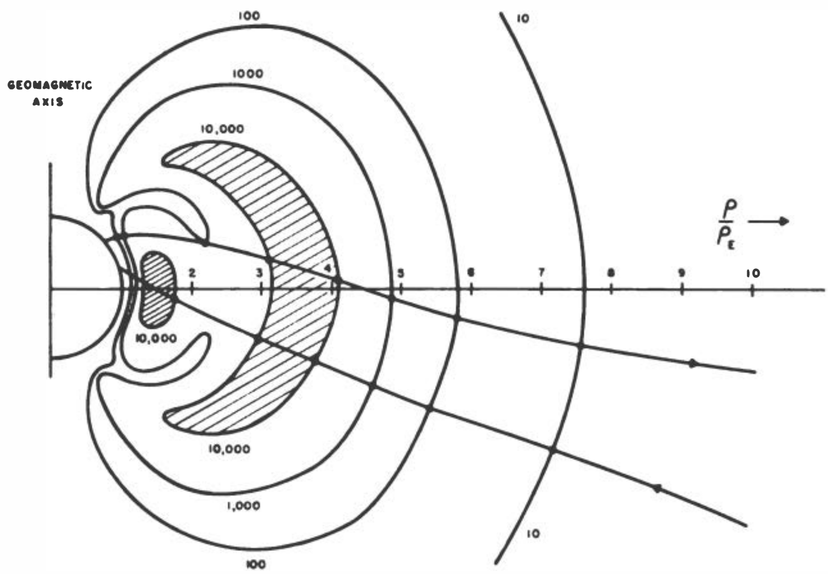


Figure 1. Intensity-structure of the trapped radiation around the earth. The diagram is a section in a geomagnetic meridian plane of a three-dimensional figure of revolution around the geomagnetic axis. Contours of constant intensity are labeled with numbers 10, 100, 1000 and 10, 000. These numbers are the true counting rates of an Anton Type 302 Geiger tube carried by Explorer IV and Pioneer III. The linear scale of the diagram is relative to the radius of the earth--6371 km. The outbound and inbound legs of the trajectory of Pioneer III are shown by the slanting, undulating lines. See references for further discussion.

4. There seems to be very little doubt that the outer zone is the result of intrusion of solar plasma into the geomagnetic field, of its temporary trapping there and of "local acceleration" of the charged particles therein.

5. The composition at the heart of the outer zone on an occasion of high intensity [ Van Allen and Frank (1959b)] was as follows:

Electrons of energy greater than 20 kev:
Omnidirectional intensity $\sim 1 \times 10^{11}/cm^2$ sec.
Electrons of energy greater than 200 kev:
Omnidirectional intensity $\leq 1 \times 10^{8}/cm^2$ sec.
Protons of energy greater than 60 Mev:
Omnidirectional intensity $\leq 10^{2}/cm^2$ sec.



Copyright © National Academy of Sciences. All rights reserved.

The typical level of intensities in the outer zone is at least an order of magnitude less than given in this example. There remains an especially important lack of observational knowledge of the intensity of charged particles of lesser energies than those specified.

6. The composition at the heart of the inner zone (more-or-less time-stationary during the past 30 months) has been estimated by the author as follows:

Electrons of energy greater than 20 kev:
Maximum unidirectional intensity $\sim 2 \times 10^9/cm^2$ sec sterad.
Electrons of energy greater than 600 kev:
Maximum unidirectional intensity $\sim 1 \times 10^7/cm^2$ sec sterad.
Protons of energy greater than 40 Mev:
Omnidirectional intensity $\sim 2 \times 10^4/cm^2$ sec.

7. It now appears very probable that the high energy proton component of the inner zone is due to the decay of a small fraction of the neutron albedo which emerges from the top of the Earth's atmosphere as a result of cosmic ray interactions with atmospheric constituents. The origin of the electron component in the inner zone is not as clear but is likely the same.

## Geonuclear Effects of the Trapped Radiation

In estimating quantitative geonuclear effects of the trapped radiation, it is appropriate to take the effects of the cosmic radiation as a standard of comparison.

Present evidence indicates that the intensity of trapped electrons (in either the inner or the outer zone) of sufficiently high energy ($\gtrsim$10 Mev) to produce nuclear disintegrations in the atmosphere is many orders of magnitude too small to be of interest in geonuclear processes.

Only the protons in the inner zone are of potential interest in this connection. But if the neutron albedo hypothesis of their origin is indeed correct, it follows that their contribution to geonuclear effects is only a trivial fraction of the total of cosmic ray effects. The argument is as follows: The fraction of cosmic ray energy carried away from the top of the atmosphere by energetic neutron albedo does not exceed $10^{-1}$. Furthermore, only some $10^{-3}$ of neutrons of energy greater than 0.5 Mev decay within the region of the geomagnetic field in which they are effectively trapped. Thus the total source strength for trapped protons of possible geonuclear interest is energetically much less than $10^{-6}$ of the total source strength for all cosmic ray processes. Trapped particles enjoy a vastly greater geometric path length than do cosmic ray secondaries



Copyright © National Academy of Sciences. All rights reserved.

which move downward into the atmosphere. But their physical path length (i. e., amount of material penetrated before coming to rest) is unchanged. Hence, it seems safe to conclude, without more detailed discussion, that trapped protons from cosmic-ray neutron albedo are of little or no geonuclear interest.

Nonetheless, an object exposed to the bombardment of trapped protons in the inner zone may be expected to become radioactive [Evans (1958)]. Indeed, observational evidence was obtained [Vernov and Chudakov (1960)] that the NaI crystal and the material surrounding it in Sputnik III developed a significant level of radioactivity characterized by a half-life of the order of one hour and by other half-lives of the order of weeks or more. A quantitative study of the production of nuclear stars in emulsion by protons in the lower fringe of the inner zone [Yagoda (1960)] has been made.

The reader is reminded that the underlying assumption of the preceding discussion is that the trapped protons in the inner zone are the decay products of neutron albedo generated in the atmosphere by cosmic ray bombardment. It is conceivable, though perhaps unlikely, that they arise from geomagnetic capture of solar protons. But even in this case the geonuclear effects of trapped protons are found to be negligible. (The direct effects of incoming solar protons require separate consideration.) The approximate agreement of the World's inventory of $C^{14}$ with that expected from cosmic ray production alone [Libby (1955)] provides independent, over-all confirmation of the more detailed estimates given above.

## Geophysical Effects of the Trapped Radiation

In contrast to the negative assessment of geonuclear effects, it is reasonably certain that the trapped radiation is intimately associated with, and in some cases essential to, a wealth of geophysical (i. e., atomic, molecular and electronic) effects. The inner zone is of secondary importance in this connection. But the outer zone participates to an important degree in many local and worldwide geophysical phenomena. Indeed its existence provides an important new element in the understanding of a variety of problems of long standing. A comprehensive discussion of such matters is not attempted here but the following listing of pertinent phenomena indicates the scope of current investigations:

1. high latitude aurorae
2. low latitude aurorae
3. airglow
4. geomagnetic storms and quiescent geomagnetic effects
5. atmospheric heating, particularly in the auroral zone
6. ionospheric effects
7. generation of radio noise and whistlers



Copyright © National Academy of Sciences. All rights reserved.

Arnold: The time required for six passages of Sputnik III through the inner zone should be a day or so, and consequently we might expect nuclides to have been produced with lifetimes of the order of a few hours to a few days. None of the aluminum isotopes falls in this range--they all have half-lives of a few seconds or minutes, except for $Al^{26}$ which is $10^6$ years. $I^{126}$ has a half-life of 13.3 days and could result from a (p, pn) reaction on the iodine in the scintillation crystal. [There are no obvious candidates among the light isotopes which might reasonably be formed from $Al^{27}$, $Mg^{24-26}$, $O^{16}$, or $N^{14}$, having the right half-life and emitting either positrons or 0.5 Mev gammas.]

Cameron: In view of the large radiation belts which we infer for Jupiter from the radio noise and polarization observations, we might expect Saturn to have substantial radiation belts as well. The fact that it apparently does not could be attributed to the presence of rings--the dust or ice particles would simply absorb the protons and electrons. It seems doubtful that Saturn could maintain its rings for 4.5 billion years under these conditions; if not, then the rings must be steadily regenerated.

## REFERENCES

Evans, T. C. (1958) private communication to the author.

Libby, W. F. (1955) Radiocarbon Dating, 2nd edition, 175 pp., University of Chicago Press.

Van Allen, J. A. (1959) "The Geomagnetically-Trapped Corpuscular Radiation," J. Geophys. Res. 64, 1683-1689.

Van Allen, J. A., and Frank, L. A. (1959a) "Radiation Around the Earth to a Radial Distance of 107,400 Km." Nature (London), 183, 430-434.

Van Allen, J. A., and Frank, L. A. (1959b) "Radiation Measurements to 658,300 Km with Pioneer IV," Nature (London), 184, 219-224.

Van Allen, J. A., McIlwain, C. E., and Ludwig, G. H. (1959) "Radiation Observations with Satellite 1958ε," J. Geophys. Res. 64, 271-286.

Vernov, S. N., and Chudakov, A. E. (Moscow 1960) "Investigations of Radiation In Outer Space," in Proceedings of the Moscow Cosmic Ray Conference, pp. 19-29, Vol. III, ed. S. I. Syrovatsky, International Union of Pure and Applied Physics.



Copyright © National Academy of Sciences. All rights reserved.

Vernov, S. N., Chudakov, A. E., Vakulov, P. V., and Logachev, Yu. I. (1959) "Study of Terrestrial Corpuscular Radiation and Cosmic Rays during Flight of the Cosmic Rocket," Doklady Akad. Nauk. SSSR 125, 304-307.

Yagoda, H. (1960) "Star Production by Trapped Protons in the Inner Radiation Belt," Phys. Rev. Letters 5, 17-18.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# MICROMETEORITE STUDIES FROM EARTH SATELLITES

W. M. Alexander
National Aeronautics and Space Administration
Goddard Space Flight Center

At the present time, solid particles of less than $1 \times 10^{-1}$ millimeters or so in radius cannot be detected by means of radar or photographic images, and thus can only be recorded through the dynamical effects of an impact. Both the mass spectrum and velocity spectrum of the impacting particles are initially of major interest, and these quantities may be resolved by means of a pair of complementary detectors, one of which measures momentum and the other energy.

Of the sensors now in use, the momentum detector is in the highest state of development and has been flown in a number of space vehicles. It consists of a crystal microphone mounted against a sounding board. The output of the crystal-microphone-amplifier system has been calibrated for particles of various masses at velocities from 20 cm/sec to 2 km/sec and has been found to be a linear function of momentum (Kells 1959). Variation of velocity over six orders of magnitude with momentum held constant produced a change in output of less than a factor of two. The calibration will be extended to velocities above 10 km/sec to place the microphone sensor on a perfectly sound basis, and this may be achieved this year.

The energy detector consists of a phototube made opaque by means of a 1000 Å coating of aluminum over the surface. When particles hit the surface some of their energy is converted into a light flash. The integrated emission in the flash appears to be a single-valued function of energy, but the light-flash calibration is not as advanced as that of the crystal microphone detector. The mechanisms of energy conversion and conversion efficiencies are still inadequately understood. In any event the device is limited to high velocity particles; no energy detector has yet been fully developed for low velocity particles.

Actually, it has not been possible to use the maximum capacity of the microphone sensors on space vehicle flights up to this time. The momentum detector has recently flown in three such vehicles, $\alpha$ '58, $\eta$ '59, and Pioneer I, and has obtained information principally on impact frequencies of particles with momenta larger than a given threshold:



Copyright © National Academy of Sciences. All rights reserved.

| Space Craft | No. of Events | Impact Frequency ($m^{-2}$ $sec^{-1}$) | Momentum Threshold (dyne sec) |
|---|---|---|---|
| α '58 (Dubin 1960) | 153 | $1 \times 10^{-2}$ | $2.5 \times 10^{-3}$ |
| η '59 (LaGow 1960) (Alexander 1960) | 3725 ± 25 | $2 \times 10^{-3}$ | $1 \times 10^{-2}$ |
| Pioneer I (Dubin 1960) | 17 | $4 \times 10^{-3}$ | $2 \times 10^{-4}$ |

The data for η '59 has been corrected for spurious events caused by interference with the magnetometer interrogation system, and the correction is believed to be reliable within the error quoted.

Rocket flights of the energy detector in 1955 (Berg 1956), and 1960 (Berg 1960), have shown that impacts on the leeward side of the Earth, with respect to the Earth's orbital motion are ≈50 to 100 times less frequent than on the windward side. This is about the magnitude of the effect which would be expected under the assumption that the micrometeorites converge toward the Earth's orbit from regions farther away from the Sun, and possess no less than Earth's escape velocity (11 km/sec).

On the basis of the flights of the energy and momentum detectors a crude mass spectrum can be derived from the micrometeorites. The spectrum is, at this time, indefinite and warrants only a few conclusions:

1. The line relating the logarithm of the number of impacts above threshold to the mass of the impacting particles has a slope near -1.

2. The line relating the logarithm of the number of impacts to the logarithm of the momentum of the impacting particles has a slope near -1.

3. On the basis of extrapolations of the size distributions of cm- and mm-sized meteors detected by means of photography and radar (Watson 1952; Manning 1959, Whipple 1958), it would appear that 1 - 10 μ radius micrometeorites are between one and three orders of magnitude less abundant than the satellites indicate. Whipple (1959) however points out that the luminous efficiencies of meteors depends highly on particle mass and density, and are probably not known to better than within two orders of magnitude. Whipple's own estimate of the luminous efficiency of meteors, based on an assumed density of $\rho = 0.05$ gm/cm$^3$, leaves less than one order of magnitude between the frequency value extrapolated from meteors down to micron size and the observed frequency of



Copyright © National Academy of Sciences. All rights reserved.

micrometeorites. The discrepancy is probably not to be regarded as a serious problem in view of the large extrapolations involved--at least until the composition of meteors can be established much more definitely.

4. According to satellite measurements to date, the amount of material falling to Earth in the micrometeorite range is of the order of $5 \times 10^{-12}$ g/m$^2$/sec, which is equivalent to about $10^4$ tons per day.

Pure impact detectors have been flown on several occasions, but are thus far capable of giving only semi-quantitative data. Wire grids may have been broken on Explorer III but not on Explorer I (Manning 1959). A 6μ film, made opaque with 1000 Å of Al, has apparently received an impact opening 15μ in diameter, as measured by a CdS photocell on Explorer VII (LaGow 1960). A package ejection system is being developed which will be tried this year. The package will carry cosmic ray emulsions, and attempts will also be made to recover micrometeorite material by stopping them in a lucite trap.

Van Allen: How sure are you that micrometeorites exist at all?

Alexander: We have tested the momentum detector for several possible sources of spurious signals: thermal shock, vibration, interference from interrogation of other instruments. On $\eta$ '59, magnetometer interrogations have produced spurious signals, and corrections have been made to this date--in fact, the lowest false count rate was in a month of high magnetometer interrogation. An in-flight calibration system which produces a diaphragm mechanical shock, is being placed in operation to check the functioning of the microphone sensor.

Anders: From his studies of deep-sea sediments Pettersson (1950 and 1960) has concluded that the volume of nickel-rich spherules falling to Earth each day must amount to a few thousand tons. This material is in a limited size range and constitutes only iron which has been melted. Naturally there is some unmelted iron falling too, and much silaceous and cometary material. It is interesting that the total micrometeoritic material that has been found in the satellite studies is only about ten times the amount of material in the form of spherules.

## REFERENCES

Kells, M. C., et al. (1959) in Proceedings of the Third Symposium on Hypervelocity, Vol. 1, 361-384.

Dubin, M. (1960) Planetary and Space Science, 2, 121-129.



Copyright © National Academy of Sciences. All rights reser

LaGow, H. E., and Alexander, W. M. (1960) in Proceedings of First International Space Science Symposium, Vol. 1.

Alexander, W. M., LaGow, H. E., and Secretan L. (1960) NASA Technical Notes (in press).

Dubin, M. (1960) in Proceedings of the First International Space Science Symposium, Vol. 1

Berg, O. E., and Meredith, L. H. (1956) J. Geophys. Res. 61, 751.

Berg, O. E., and Alexander, W. M. (1960) NASA Technical Notes (in press).

Watson, F. G. (1952) Between the Planets.

Whipple, F. L. (1958) Am. Rocket Soc., Paper No. 499-57.

Manning, L. A. and Eshleman, U. R. (1959) Proc. IRE 47 (2), 186.

Whipple, F. L. (1959) SNAP sponsored conference on Meteoroid Hazard to Space Power Plants.

Manring, E. R. (1959) Planet Space Sci. 1, 27-31.

LaGow, H. E., and Secretan, L. (1960) NASA-MSFC Report on Scientific Results on Explorer VII.

Pettersson, H., and Rotschi, H. (1950) Nature 166, 302.

Pettersson, H. (1950) Sci. Ann. (August)

Ibid. (1960), Sci. Ann. 202 (2), 123.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# EXPLORATION OF THE MOON AND PLANETS

A. R. Hibbs
Jet Propulsion Laboratory
California Institute of Technology

## Planned Program of the California Institute of Technology Jet Propulsion Laboratory

The program for exploration of the Moon and planets is fairly well mapped out, for the next few years at least. In about a year's time there will be two test flights of Earth satellites very similar to each other. (These will be "space craft," defined as the instrument package, or "payload," plus structure, power supply, antennae, etc.) These satellites will be powered by solar batteries, and will have useful lifetimes of about two to three months. Their apogee will be between one and two million kilometers, so that they will have periods of approximately two months.

Beginning in the second half of 1961, three space craft will be aimed at the Moon; payloads will be in a capsule equipped with retrorocket. The instruments, including a single-axis seismometer, are being designed to withstand impact of 70-100 meters/sec on a surface similar to concrete. The seismometer will report for several weeks. Before the release of the capsule, while approaching the lunar surface, a γ-ray spectrometer, with 32-channel analyzer, will look for $K^{40}$ activity. If the lunar surface has as much or more $K^{40}$ as chondritic meteorites, it will be detectable. Vidicon photographs of the lunar surface will also be transmitted. Impact will be south and a little west of the crater Kepler. The exact location will be determined by radio-tracking.

Now to somewhat more speculative plans. Venus will be in a favorable position in August 1962, Mars in November of the same year. It is possible that attempts will be made to launch space craft at either or both of these targets. Their payloads will be designed for two types of experiments. In-flight experiments will examine properties of interplanetary space, while spectrograms, of the planetary atmosphere in the case of Venus and of the Martian surface, will be made on arrival at the planets.

In 1963 or 1964, when it is hoped that the Centaur rocket will be ready, better-controlled soft landings on the Moon will be attempted. In 1965, if the Saturn rocket is available, it is very likely that devices can



Copyright © National Academy of Sciences. All rights reserved.

be landed on the Moon capable of returning samples to the Earth. Further plans are quite nebulous.

In terms of planetary exploration, it will be 1964 or later before a payload is landed on the surface of Mars or Venus, and not until 1966 or 1967 will remote-controlled experiments, of the type expected for the Moon in 1963, be possible for our planetary neighbors. The return of samples from Mars or Venus is much more difficult than from the Moon, so that planetary surface exploration by roving vehicles, etc., will be considerably in advance of actual delivery of samples.

## Mixing of the Lunar Surface Material

In designing a program of lunar exploration, it is important to have some idea of the properties of the surface. One of the most significant of these properties is the amount of dust on the surface. Measurements of the flux of micrometeorites in regions of space near the Earth have been made (giving $8 \times 10^{-3}$ particles $cm^{-2}$ $sec^{-1}$); coupled with a reasonable estimate of the size of these particles, it seems that the order of one meter thickness of micrometeorites should be deposited on the Moon's surface in a billion years. Also, there is good evidence for some long-term erosion processes on the Moon. [Hibbs engaged in an outline of the controversy between the lava and dust hypotheses of the origin of the lunar maria, essentially following the line of argument published by T. Gold (1955). Olbert pointed out that he had made some calculations of the space charge and saltation distances to be expected for dust grains charged by Gold's postulated photoelectric effect, which indicate that extensive movement of dust might be expected, although the system represents a very complex problem in electro-hydrodynamics and the conclusions are somewhat uncertain.] In any case, one would expect that deposition of micrometeorites and erosion and transport of dust would obliterate the initial surface markings. Yet the ray systems, which may be billions of years old, and certain markings on the floors of the maria remain visible. The problem is then: to what extent can localized mixing of material from the original surface with the overlying dust layer, by means of meteoritic pitting, preserve the initial surface characteristics? Assign, for coding purposes, a "whiteness" to the initial surface, and a "blackness" to the material added. Then the fraction of white material on the present surface is $W(x)$, where $x$ is the depth of the dust layer. Two rates are involved: $r_m$ is the rate at which meteorites redeposit material from depth onto the present surface; $r_e$ is the rate of deposit of "black" material. It may be assumed that the color at a distance $x$ from the original surface obeys the equation:

$$W(x) = \frac{r_m}{r_e + r_m} \cdot \frac{1}{V} \int_0^x A(x - \xi)\, W(\xi)\, d\xi \qquad (1)$$



Copyright © National Academy of Sciences. All rights reserved.

where V is the volume of the "average" meteorite pit (averaging done in some suitable but undefined way) and $A(x - \xi)$ is the horizontal cross-sectional area at depth $x - \xi$. The solution of equation (1) is particularly simple for a square-well type crater of average depth $l_o$:

$$W(x) = e^{-\lambda x} \qquad (2)$$

where the characteristic distance $\lambda$ is implicitly defined as:

$$\lambda \ell_o = - \frac{r_m}{r_e + r_m} (1 - e^{+\lambda \ell_o}) \qquad (3)$$

Assuming $r_m = r_e$, $\lambda = 1.26 \ell_o^{-1}$. Clearly, material could only be mixed through a characteristic distance the order of the average crater depth under these conditions. Therefore, the preservation of the color contrasts in the maria by this mechanism would require craters large enough to be observable, if the postulated 100-meter depth of dust is indeed present. If one attempts to avoid this dilemma by increasing the efficiency of mixing material from depth, then it is necessary to assume that $r_m \gg r_e$. In this case, an approximation may be made: $r_e/r_m \cong 1/2\lambda\ell_o$. Thus, for mixing "white" material from depths ten times the average crater depth, $r_e$ must be about 5 percent of $r_m$. This could only be valid if the material brought in by the meteorites were negligible compared to the material stirred up, and if the rate at which dust is transported were small. [Considering that the "average" depth is probably very nearly that of micrometeorite craters, either $r_e/r_m$ must be still smaller, or the depth of dust on those regions of the maria where apparently "original" surface marks remain must be much less than the order of 100 meters.]

Regardless of the proper parameters to fit into these equations, such mixing processes must have occurred to some extent. Apparently, the lunar surface has had a complex history which should be explicitly considered when analyzing the data which will be available in the next few years.

Kohman: I wonder if there is really a net deposition of material on the Moon? Much of the incoming material must be simply vaporized.

Hibbs: In any case, there is some redeposition. This may be seen in the rays associated with some of the prominent craters. Also, erosion from the "highlands" and mixing should occur in any case, although the rates are quite uncertain.

Cameron: The rate of transport of eroded material, which is an important component of $r_e$, is especially sensitive to sintering. It may be that, considering a steady state balance between erosion and sintering, conditions strongly favor sintering of these fine particles, so that $r_e$ is quite low.



Copyright © National Academy of Sciences. All rights reserved.

Arnold: It is a difficult problem, because there are effects acting in both directions. For example, sintering should be slow in the absence of water, but accelerated by the hard vacuum and by the unsaturated bonds due to solar radiation.

## REFERENCE

Gold, T. (1955) "The Lunar Surface," *Mon. Nat. of Roy. Astro. Soc.* 115, 585.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# DIAMONDS IN METEORITES

Edward Anders
Enrico Fermi Institute for Nuclear Studies
University of Chicago

Diamonds have been reported in five meteorites; in one case, Carcote, the identification is uncertain and has never been confirmed. The remaining diamond-bearing meteorites comprise two ureilites (a rare sub-class of achondrites), Novo Urei and Goalpara, and two coarse octahedrites, Magura and Canyon Diablo.

The orthodox interpretation of the origin of meteoritic diamonds is that they were formed from graphite under high pressures. The pressures required are about $1.6 \times 10^4$ atm. at $298^\circ$ K and about $3.2 \times 10^4$ atm. at $1000^\circ$ K. Assuming that the source of pressure was the gravitational potential of a meteoritic parent body of reasonable density ($\sim 3.3$ g $cm^{-3}$), the minimum radius of such an object must have been $\sim 1000$ km. In order for one-half of the interior of the parent body to be at or above the minimum pressure, it must have been of approximately lunar size (radius $\sim 1700$ km). These considerations impelled Harold Urey to postulate a generation of lunar-sized parent objects as a principal feature of his proposed history of the meteorites.

Parent objects of lunar size, however, are hard to reconcile with evidence discussed in two papers by members of our group (Goles, Fish and Anders, 1960; Fish, Goles and Anders, 1960) and with the model of meteoritic synthesis developed in the latter paper. Accordingly, we felt it was necessary to re-examine critically the orthodox interpretation of meteoritic diamonds to determine whether sustained high pressures were required by the evidence. This investigation, which was carried out mainly by Mr. M. E. Lipschutz, took two forms: a study of the thermodynamics of diamond synthesis in the presence of metallic iron, and an empirical study of the pressure and temperature conditions under which Canyon Diablo diamonds were formed.

Since excess Fe is always present in diamond-bearing meteorites, and since diamonds in Canyon Diablo at least are not found associated with graphite nodules but with other minor phases, it appears that the Fe-C-$Fe_3C$ system is the appropriate one to consider in investigating diamond formation in meteorites. Cohenite ($Fe_3C$) can decompose to



Copyright © National Academy of Sciences. All rights reserved.

either diamond or graphite over a wide range of temperatures, but the reactions are quite sensitive to pressure. When the loci of $\Delta F = 0$ for these decomposition reactions are plotted in the P-T plane, a very informative diagram results (Figure 1; see Lipschutz and Anders, 1960, for discussion of the derivation and significance of this diagram). In the absence of a solvent, this graph may be interpreted as a phase diagram indicating the stable and metastable forms of carbon at a given temperature and pressure. Note that in the presence of excess iron, both graphite and diamond are unstable with respect to cohenite in the area to the

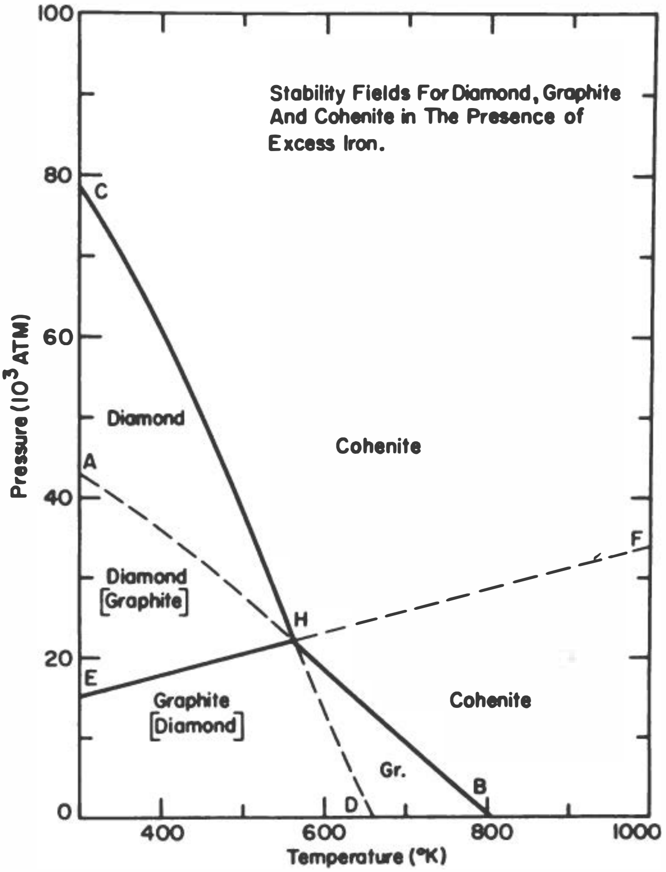


Figure 1. Stability fields for diamond, graphite and cohenite in the presence of excess iron. Metastable phases are indicated by brackets. In the absence of a solvent, this diagram is equivalent to a phase diagram, whereas in the presence of a solvent, it may be used to infer the shift in equilibria upon pressure or temperature change.



Copyright © National Academy of Sciences. All rights reserved.

right of lines CH and HB. From this, it can be shown that it is virtually impossible to construct a plausible history that would permit the preterrestrial formation of the alleged "graphite pseudomorphs after diamond" and the Widmanstätten pattern.

In the presence of a solvent, the decomposition reactions of cohenite may be treated as ordinary chemical equilibria at small values of $\Delta F$. All stable reactants and products will be present in appreciable amounts at any temperature and pressure, their proportions being governed only by the value of the equilibrium constant. One can still use Figure 1 to infer qualitatively the shift in equilibrium for any temperature or pressure change. Of course, diamond may form metastably below the line EH, and graphite, above this line.

Our study of diamond-bearing specimens of Canyon Diablo has strongly supported the suggestion by Nininger that the diamonds in this meteorite were synthesized during impact, rather than pre-terrestrially. This conclusion is based on the following line of reasoning: It has long been noted that diamonds in Canyon Diablo: 1) are not associated with graphite; 2) are always associated with troilite; 3) are imbedded in areas which contain cohenite, though not necessarily in contact with the diamonds; 4) are submicrocrystalline, with an ultimate particle size of a few hundred angstroms, as determined by x-ray diffraction; and 5) are irregularly distributed, so that one specimen may be rich in diamonds while other large pieces are completely free of them. In addition, Nininger has pointed out that the diamond-bearing specimens are usually small (<5 kg), are found only on the rim of the crater, and display evidence of reheating. We may infer that there are two distinct types of Canyon Diablo specimens, that the distribution and other properties of the diamond-bearing ones indicate a correlation between conditions during impact and the presence of diamonds, and that the precursor of the diamonds was, as argued above, cohenite rather than graphite. Since there seems to have been a causal relationship between reheating and diamond synthesis (or, at least, they occurred simultaneously), study of the conditions of reheating should provide information on the conditions of diamond formation.

Microscopic examination of diamond-bearing Canyon Diablo specimens reveals that, in contrast to the diamond-free type, the kamacite ($\alpha$-phase) is polycrystalline, the Neumann lines have been annealed out, and there are borders of martensite (metastable iron-carbon alloy, formed only by very rapid cooling of carbon-rich $\gamma$-phase) around cohenite lamellae. These features may be reproduced quite accurately by heating meteorite specimens to 850 - 950 °C for a few seconds and quenching in cold water, as determined by experiments on the structurally very similar Odessa octahedrite. Trial-and-error experiments with Odessa, using various minor phases ($Fe_3P$, $Fe_3C$, etc.) and their eutectics as



Copyright © National Academy of Sciences. All rights reserved.

temperature indicators, show that diamond-bearing Canyon Diablo specimens were reheated to temperatures between 925 and 1000 °C. Rapid cooling, on a time scale of <2 min., is indicated by the formation of martensite rather than pearlite around cohenite inclusions. Since no one would seriously propose that the original mass of the Canyon Diablo meteoroid (~2 x $10^6$ tons, according to Öpik) could be quenched so rapidly, it is clear that these structures were developed after the fragments attained their present small size, i.e., during impact.

Thus, we would hypothesize that the Canyon Diablo diamonds formed from cohenite during impact. The fact that diamond rather than graphite was formed may be due to the high pressures prevailing in the decelerating meteoroid during its penetration into the ground, to localized stresses due to differential thermal expansion, or to metastable formation of diamond due to preferential nucleation. We may estimate a lower limit to the time during which the meteoroid was under compression by equating this with the minimum time for penetration to the floor of the crater. For an initial velocity of 15 km/sec, one obtains $\geq 1.7 \times 10^{-2}$ sec for this "compressive deformation" time. At growth rates of ~0.1 mm/min (Bovenkerk et al., 1959), diamonds of ~300 Å size could have been formed in this time. For the other processes suggested, similar time scales should apply.

What of the other diamond-bearing meteorites? In the case of Magura, the mode of diamond synthesis may be similar to that proposed for Canyon Diablo. Many fragments of Magura have been recovered, and they display clear evidences of different thermal histories. We cannot say whether or not there is a one-to-one correlation between the presence of diamonds and intense reheating in specimens of this meteorite, but it is at least possible that these features were produced during impact. For the ureilites, on the other hand, it may easily be established that they arrived at the Earth's surface with relatively low terminal velocities, so that this explanation would not apply. Nevertheless, at least once in their preterrestrial history the ureilites must have been involved in a violent collison (upon the occasion of the break-up of their parent body). It is tempting to conjecture that the diamonds found in the ureilites were formed in that event. Unfortunately, nothing is known regarding the mode of occurrence of these diamonds, so that it is not possible to decide whether they could have been synthesized in such an impact.

Cameron: Can you set a lower limit to the size of the crater produced by, for example, Magura, if this meteorite contains diamonds which were formed on pact?

Anders: It would be possible to do this, using the diamond growth-rate mentioned and an estimate of initial velocity, but I would expect that



Copyright © National Academy of Sciences. All rights reserved.

the value obtained for the crater depth could be uncertain by a factor of one hundred and therefore of little use.

## REFERENCES

Bovenkerk, H. P., Bundy, F. P., Hall, H. T., Strong, H. M., and Wentorf, R. H., Jr. (1959) Nature (London) 184, 1094.

Fish, R. A., Goles, G. G., and Anders, E. (1960) Astrophys. J. 132, 243.

Goles, G. G., Fish, R. A., and Anders, E. (1960) Geochim. et Cosmochim. Acta. 19, 177.

Lipschutz, M. E., and Anders, E. (1960) submitted to Geochim. et Cosmochim. Acta.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# HEAVY-ELEMENT ACTIVATION ANALYSES OF METEORITES

George W. Reed, Jr.
Argonne National Laboratory

During the past few years, a comparatively large number of neutron activation analyses for various heavy elements have been done on meteorites by our group (Hamaguchi, Kigoshi, Turkevich and Reed). The results of this work in general have been published already (Hamaguchi *et al.*, 1957) or are in press (Reed *et al.*, 1960), but it seems worthwhile to review briefly some of the data, indicating certain of the principal implications they may have.

Analyses have been done for Hg, Tl, Pb, Bi and U; Ba has been determined as a by-product of the uranium work. In some cases, information on isotopic abundances has been obtained. These results have significance for the investigation of the so-called "cosmic" (really meteoritic, in this case) abundance curve, and for the history of the meteorites.

In particular, it has been possible to show the identity of meteoritic and terrestrial ratios of isotopes of Ba, Hg and U (to within 10 percent or better), and to confirm independently some of the mass spectrometric work on meteoritic Pb-isotope ratios. The accuracy of our work is of necessity not as good as can be obtained by mass-spectrometry, but problems of terrestrial contamination are generally less prominent in our methods. A significant point is that Indarch (enstatite chondrite), Mighei, and Orguiel (carbonaceous chondrites) not only contain much more Pb, by a factor of about ten, than the "ordinary" (hypersthene-bronzite) chondrites studied so far, but their $Pb^{208}/Pb^{204}$ ratios are the same, within experimental errors, as the ratio for primordial Pb in troilite from iron meteorites, as determined both by Patterson (1956) and by ourselves. Since these chondrites are very different from iron meteorites, it would seem that Patterson's identification of primordial Pb is strongly supported by these results.

A very interesting pattern can be inferred from the data on elemental abundances in the meteorites studied. "Ordinary" chondrites have rather low and reasonably constant abundances of Tl, Pb, Bi and U, while the first three elements are very much more abundant in enstatite and carbonaceous chondrites (see Table 1). U (and Ba) are quite constant in all chondrites. The elemental abundances of the carbonaceous and



Copyright © National Academy of Sciences. All rights reserved.

TABLE 1

Abundances of Heavy Elements in Chondrites

| | Tl ($10^{-9}$ g/g) | Pb ($10^{-6}$ g/g) | Bi ($10^{-9}$ g/g) |
|---|---|---|---|
| "Ordinary" Chondrites | ~0.4 | 0.01 - 0.4 | ~3 |
| Abee (Enstatite Chondrite) | ~87 | ~3.5 | ~80 |
| Indarch (Enstatite Chondrite) | 125 | ~2.0 | prob. > 100 |
| Mighei (Carbonaceous Chondrite) | 97 | ~1.5 | 180 |
| Orguiel (Carbonaceous Chondrite) | 141 | ~3.0 | 130 |

enstatite chondrites agree more nearly with the predicted values of Suess and Urey and of Cameron than those observed in "ordinary" chondrites. The small amount of data on Hg abundances appears to fit this pattern, except that the carbonaceous chondrite Orguiel seems to have a remarkably high content of Hg. It may be quite possible to correlate Hg contents with the thermal history, as indicated by compactness, hardness, content of volatiles, etc., of the meteorites. Further studies of Hg in stone meteorites are in progress.

While the variation in Hg abundances is probably at least in part a reflection of different thermal histories, no such simple explanation seems applicable to the observations for the other elements. A variation in sulfur content among the several classes of chondrites exists, but is too small to account for the differences in elemental abundances without postulating large fractionations of the trace elements relative to sulfur.

Analyses of U and Bi in Nuevo Laredo, which is an achondrite strongly enriched in the former element, allow us to infer the time elapsed between the nucleosynthesis of $Np^{237}$ and the solidification of this meteorite. Assuming that $2.2 \times 10^6$ year $Np^{237}$ (ancestor of $Bi^{209}$) was formed in a single-event nucleosynthesis with abundance equal to that of $U^{235}$, the time elapsed until the solidification of Nuevo Laredo was $\geq 2 \times 10^7$ years, given that there is $\sim 10^{-7}$ g U/g and $< 2 \times 10^{-9}$ g Bi/g in Nuevo Laredo. [This lower limit concurs with the I-Xe ages reported elsewhere by Reynolds and with the models proposed by Cameron and Kohman in this volume, in the sense that it is about an order of magnitude less than the time spans indicated.]

Anders: The abundance anomalies pointed out by Reed may well be of great significance for considerations of the histories of meteorites. These large fractionations of certain trace elements may be explained



Copyright © National Academy of Sciences. All rights reserved.

by a model recently proposed by Fish, Goles and Anders (1960). The important conclusions reached are that such a fractionation is not only possible but that Hg, Tl, Pb, Bi and In should be affected by it and Se, Zn and Cd should not be fractionated. It is suggested that the rare meteorites which are troilite-rich (e.g., Soroti) may represent a reservoir of certain trace elements which are chalcophile at high temperatures, including those elements subject to the abundance anomalies discussed by Reed.

Weatherill: From your work on Nuevo Laredo, could you state whether the Pb-Pb and U-Pb ages are or are not concordant?

Reed: The ages are very close to agreeing with each other. We feel, however, that we do not know the Pb isotopic composition accurately enough to answer this question. Certainly, the way to go at it is to measure, with high sensitivity, U, Th and Pb in the same sample. We are trying to develop a procedure for this now.

Turkevich: The discrepancies in the Nuevo Laredo ages are still somewhat outside what we believe to be our experimental error, but it is possible that with further work they could be brought into agreement. For every other meteorite on which we have enough information, the ages are definitely discordant, and therefore, in our view, suspect.

## REFERENCES

Fish, R. A., Goles, G. G., and Anders, E. (1960) Astrophys. J. 132, 243.

Hamaguchi, H., Reed, G. W., and Turkevich, A. (1957) Geochim. et Cosmochim. Acta 12, 337.

Reed, G. W., Kigoshi, K., and Turkevich, A. (1960) Geochim. et Cosmochim. Acta (in press).

Patterson, C. C. (1956) Geochim. et Cosmochim. Acta 10, 230.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# STRONTIUM AND RUBIDIUM IN STONE METEORITES

Paul W. Gast
Department of Geology and Mineralogy
University of Minnesota

The contents of Rb, Sr, in some instances K, and the $Sr^{87}/Sr^{86}$ ratio have been measured for a number of calcium-rich achondrites and hypersthene-bronzite chondrites. Most of the measurements reported here were done on the same mass spectrometer, under closely similar conditions, in order to minimize any errors arising from varying mass discrimination effects. Much of this data has been reported previously and requires little comment.

TABLE 1

Rb and Sr in Ca-rich Achondrites

| Achondrite | Rb (ppm) | Sr (ppm) | $Sr^{87}/Sr^{86}$ |
|---|---|---|---|
| Pasamonte | 0.21 | 79.3 | 0.7012; 0.7005; 0.6997; 0.702 |
| Sioux City | 0.25 | 65.7 | 0.7011; 0.7018 |
| Nuevo Laredo | 0.37 | 72.1 | 0.7027 |
| Moore County | 0.16 | 78.6 | |

Note that the enrichment of Sr relative to Rb in these achondrites is so great that there would be a negligible increment added to $Sr^{87}$ even in 5 A. E. The presumed primordial ratios of $Sr^{87}/Sr^{86}$ are in good agreement with one another; a value of 0.7015 is suggested as the best estimate of the primordial ratio. (Note that the results for Pasamonte are in serious disagreement with that reported by Schumacher (1956) on the same sample, 0.686 ± 0.006.)

Analyses have been made on four chondrites, three of which give results that appear to be quite reliable. The last four entries in Table 2,



Copyright © National Academy of Sciences. All rights reserved.

TABLE 2

K, Rb, and Sr in Chondrites(1)

| K | Rb | Sr | 87/86 | Age(2) |
|---|---|---|---|---|
| | | Forest City | | |
| 820 | 2.75 | 10.3 | 0.755 | 4.8 A.E. |
| | | Modoc | | |
| --- | 3.45 | 13.1 | 0.757 | 5.0 A.E. |
| 862 | 3.03(3) | 10.8 | --- | |
| 872 | 3.04 | -- | 0.7555 | 4.6 A.E.(4) |
| | | Richardton(5) | | |
| --- | --- | 9.8 | 0.756 | |
| 818 | 2.95 | 11.4 | --- | |
| 844 | 3.17(3) | 9.9 | --- | |
| --- | 2.73 | -- | n.d. | |
| 821 | 3.02(3) | 10.0 | --- | |
| 803 | 2.67 | -- | n.d. | |
| <10 | <0.4 | n.d. | --- | |
| 884 | 3.10 | n.d. | --- | |
| 838 | 2.93 | n.d. | --- | |
| --- | 2.74 | 9.3 | --- | |

(1) Absolute values in parts per million.

(2) The ages given are model ages in the sense that they date the hypothetical differentiation of calcium-rich achondrites from chondritic material. They are really dates for the isolation of Sr with an 87/86 ratio of 0.7015 from material of the mean composition of the chondrites, calculated using a $Rb^{87}$ half-life of 50 A.E., rather than the recent value of 47 A.E. (Flynn and Glendenin 1960).

(3) These numbers represent duplicate determinations on two aliquots of the same solution of the sample.

(4) The 4.6 A.E. age is probably more reliable.

(5) Note that the variance in Rb and Sr contents is very large, both for replicate analyses on the same samples and between samples. This variance appears to be a property of the meteorite itself, and is not associated with experimental error.



Copyright © National Academy of Sciences. All rights reserved.

on Richardton, represent analyses of four 0.7 g samples, using different chemical procedures. It is of great interest that digestion with perchloric acid alone, for 15 hours, did not succeed in bringing into solution the meteoritic K and Rb (10 ppm K by this procedure compared with ~830 by others). The small amount of residue from this digestion appeared to be a single mineral, by optical criteria. Powder photographs of the residues afforded several weak lines corresponding to plagioclase feldspar, as might be expected from the inferred high K and Rb content. Perhaps the sampling problem is due to the presence of large grains of plagioclase, inhomogeneously distributed, in which most of the K and Rb is situated.

Reynolds: We have done some density separations on Richardton samples, and find that the K is not associated with material of density similar to feldspars.

Gast: The identification is somewhat uncertain, since the index of refraction of the residue in question was too high for plagioclase. (Subsequent to the conference, a density separation was made on the insoluble residue. The density of the mineral under consideration was found to be less than 2.7. Thus plagioclase cannot be ruled out on the basis of density.)

Anders: Could the material be maskelynite, with a small amount of included plagioclase to give the weak x-ray lines?

Gast: Perhaps, although the grains displayed a definite birefringence, which would not be expected of a glassy material.

Anders: It could be strained maskelynite; we have observed a number of of strained silicate glasses in meteorites.

If one takes a grand average of the data on Richardton, the model age obtained is 4.5 A. E.

The problem of sample variability, noted in the Richardton results and present to some extent for Modoc as well, becomes acute when studying Beardsley. (Table 3.) The first four lines refer to analyses on Beardsley I, a sample obtained from Nininger which was collected two years after infall. Beardsley II (data below the dashed line) was obtained from Anders, ultimately from the Perry collection of the University of Michigan, and was collected the day after infall. The ages 5.8 and 5.5 A. E. have been previously reported.

Beardsley II is very different. It has the highest Rb content known for any meteorite, and the highest K content measured in a chondrite. While the total Sr is not changed from that of Beardsley I, the radiogenic



Copyright © National Academy of Sciences. All rights reserved.

TABLE 3

K, Rb and Sr in the Beardsley Chondrite

| K (ppm) | Rb (ppm) | Sr (ppm) | 87/86 | Age (A. E.) |
|---|---|---|---|---|
| | | Sample I | | |
| 917 | 4.90 | 11.2 | 0.812 | 5.8 |
| 895 | 4.83 | 10.4 | 0.812 | 5.5 |
| 905 | 4.80 | 10.8 | --- | |
| --- | 4.67 | -- | n.d. | |
| | | Sample II | | |
| 1227 | 14.5 | 10.6 | --- | |
| 1247 | 14.6 | -- | 0.960 | 4.25 |

$Sr^{87}$ clearly follows the Rb. The age derived (which is known to high accuracy because of the very high $Sr^{87}/Sr^{86}$ ratio) is much lower than the ages derived from the data on Beardsley I.

In considering these ages, it is very important to keep in mind that there can be only one primordial $Sr^{87}/Sr^{86}$ ratio, by the definitions used in constructing the model. If this is not 0.7015, as assumed, but rather some higher value in order to bring the data on the two Beardsley samples into agreement (for example, 0.73), then it is necessary to explain the $Sr^{87}/Sr^{86}$ ratio in the achondrites in some special way. Any such explanation must lead to the conclusion that Beardsley is significantly different from all the other meteorites studied.

A more promising explanation seems to be that of leaching. [A discussion of this topic followed, with Gast, Anders, Hurley, Goles, Turkevich, Broecker, and Arnold participating, among others. It was pointed out that the leaching would have to be such that it would remove Rb and radiogenic Sr but not primordial Sr. The results on perchloric acid digestion were cited as an objection to a large effect by leaching. Gast indicated that he intended to test this hypothesis experimentally, by attempting to leach out Rb and $Sr^{87}$ artificially.]

Addendum: Several experiments attempting to leach the alkalies out of the second Beardsley sample were performed subsequent to the conference. In the first attempt (A), two grams of the powdered



Copyright © National Academy of Sciences. All rights reserved.

meteorite were washed in warm water (~70° C) for 24 hours. The alkalies in the water were then determined in the usual way. The procedure was repeated in boiling water for 4 hours on the same powder (B). The results of these experiments are shown in Table 4. Eighteen and 55 per cent of

TABLE 4

Per Cent Extractable Alkalies in Beardsley II

| | K | Rb | Cs |
|---|---|---|---|
| A | 3.5% | 12.6% | 35% |
| B | 1.8% | 5.7% | 20% |

the Rb and Cs respectively were removed in these experiments. The results support the hypothesis that rubidium and radiogenic strontium were removed from the first Beardsley sample while it was in the ground. The separation of Rb and Cs from K during this experiment further suggests that all the alkali metals are not in the same phase in this meteorite.

## REFERENCES

Flynn, K. F., and Glendenin, L. E. (1959) Phys. Rev. 116, 744.

Schumacher, E. (1956) Helv. Chim. Acta. 39, 538-547.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# SECULAR CHANGES IN THE CONCENTRATION OF ATMOSPHERIC RADIOCARBON

Hans E. Suess
School of Science and Engineering
University of California at La Jolla

The technique of radiocarbon dating has proved to be an extremely valuable tool of research. The method is based on the following premises: (1) constancy of the cosmic ray flux; (2) constancy of the various carbon exchange reservoirs.

It was found empirically that these assumptions were correct within certain limits of error of the measurements, but refinements in the technique of $C^{14}$ determination have now shown the existence of deviations which demonstrate that one or both of the above assumptions are not precisely correct. The first observations were made by deVries (1958) who believed that his measurements indicated a correlation between the $C^{14}$ activity of wood dated from tree rings, and glacial advances and retreats. Broecker, Olson, and Bird (1959) have confirmed the deviations observed by deVries for the past two or three centuries. They also found that a specific $C^{14}$ activity one to two percent above that of the 19th century level must have prevailed during the 16th and 17th centuries. Other observers have stated that using Libby's half-life, radiocarbon dates of material from Roman times are accurate within less than 100 years. Further observations by European workers were discussed during the last radiocarbon conference in Groningen in 1959. These observations were made on tree rings of the sequoia gigantea from the British Museum dating back about 1500 years. Deviations in radiocarbon ages have also been observed by Elizabeth Ralph (1959) on samples of historically known age from early Egyptian dynasties. The samples give too high a $C^{14}$ concentration by about five to ten percent, or in other words look too young by about 400 to 800 years.

Through the courtesy of Dr. Terah L. Smiley of the Tree Ring Laboratory, University of Arizona, Tucson, we have obtained wood samples from sequoia gigantea dating back to 1100 B. C. The first thousand years in this tree produced wide rings, and ample sample material was available so that it was possible to make one determination for each century. For the younger periods (from 100 to 1700 A.D.) the wood was scarce and only one sample every 200 years could be measured. Some of



Copyright © National Academy of Sciences. All rights reserved.

the runs had to be made at a reduced pressure. As this period will be covered by measurements in Europe, we have concentrated on measurements of material older than 200 A. D.

In addition to the sequoia gigantea wood samples, two samples of two different kinds of woods (acacia and sycamore) from the tomb of King Zoser (Egypt) have been measured. According to Professor Wilson of the Oriental Institute in Chicago, who originally made this wood available to Dr. Willard Libby, King Zoser's tomb should date from about 2700 B. C., but could be as young as 2500 B. C. The wood samples of 10 to 15 grams each were first split into small fragments, then boiled with dilute NaOH and then with 3 molar HCl and washed and dried. About one-third of the material, presumably gum and cellulose, was lost by the treatment. The counting was done in an Oeschger counter (Houtermans and Oeschger, 1958) that had a background of 1.63 cpm and a counting rate of 22.44 cpm when filled with $C_2H_2$ of 940 mm Hg pressure prepared from wood grown around 1880. $C^{13}$ determinations were made in the laboratories of Dr. S. Epstein of the California Institute of Technology and Dr. Harmon Craig of the University of California, La Jolla. The results of our measurements and the deviations from the theoretical values, calculated with a half-life of 5568 years, are shown in Figures 1 and 2. The results of the measurements by Broecker, et al. (1959) are also shown in the figures. Other laboratories have obtained results that fit ours in a similar manner. There seems to be fluctuations in the $C^{14}$ concentration through time of the order of one percent, presumably outside the limits of error of the measurements, that are superimposed on a more general trend. A maximum of radiocarbon concentration existed around 1500 A. D. During the first millenium A. D. radiocarbon concentrations fluctuated around the 19th century values, as observed by other workers. During the first millenium B.C., however, the $C^{14}$ concentration seemed to have dropped almost linearly. The deviations found through measurements of historically known samples by Elizabeth Ralph, as well as the results from our wood from the tomb of King Zoser seem to follow the trend indicated by our measurements on sequoia wood for the time from 1100 B.C. to 100 A.D.

Obviously at this time no conclusive explanation can be given for the cause of $C^{14}$ fluctuations with time. As noted by deVries (1958) the maximum in the 16th century coincides with a general advance of glaciers and cold winters. Theoretically one can expect that the average climate of the Earth will affect the $C^{14}$ activity in two different ways which will tend to cancel each other. One is the increased downward mixing of surface ocean water because of a weaker thermocline in higher latitudes during periods of cold weather. The second is lower $CO_2$ concentration in air due to a lower partial $CO_2$ pressure at lower temperatures, which would tend to increase the specific $C^{14}$ activity of the $CO_2$ in air. Without a detailed analysis it is impossible to predict which effect would predominate.



Copyright © National Academy of Sciences. All rights reserved.

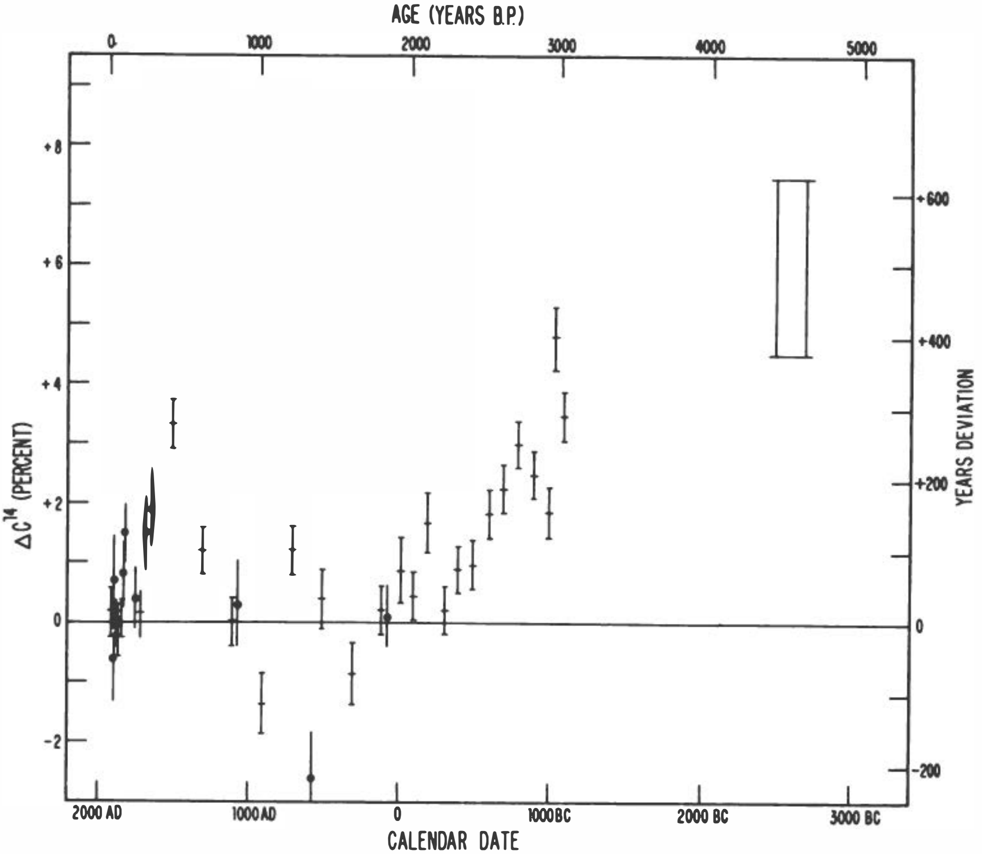


Figure 1. Logarithm of $C^{14}$ activity of wood samples (mostly sequoia gigantica) versus time of growths. Crosses represent LJ measurements. Solid circles represent values by Broecker et al. Straight line represents decay line of $C^{14}$ assuming 5568 years half-life.

The changes in the cosmic ray flux can be due to changes in (1) the galactic component, (2) the solar activity, and (3) the Earth's magnetic field. Cosmic ray induced radioactivities in meteorites seem to indicate no major change in cosmic ray activity during geologic time (Arnold, 1960). There is, however, some evidence for secular changes in the Earth's magnetic field. According to E. and O. Thellier (1959; see Elsasser et al., 1956 for other references) the Earth's magnetic field was about 65 percent stronger in Roman times than it is now. Elsasser, Ney, and Winkler (1956) have computed the change of cosmic ray intensity which might have reached the Earth's surface, using Störmer's theory,



Copyright © National Academy of Sciences. All rights reserved.

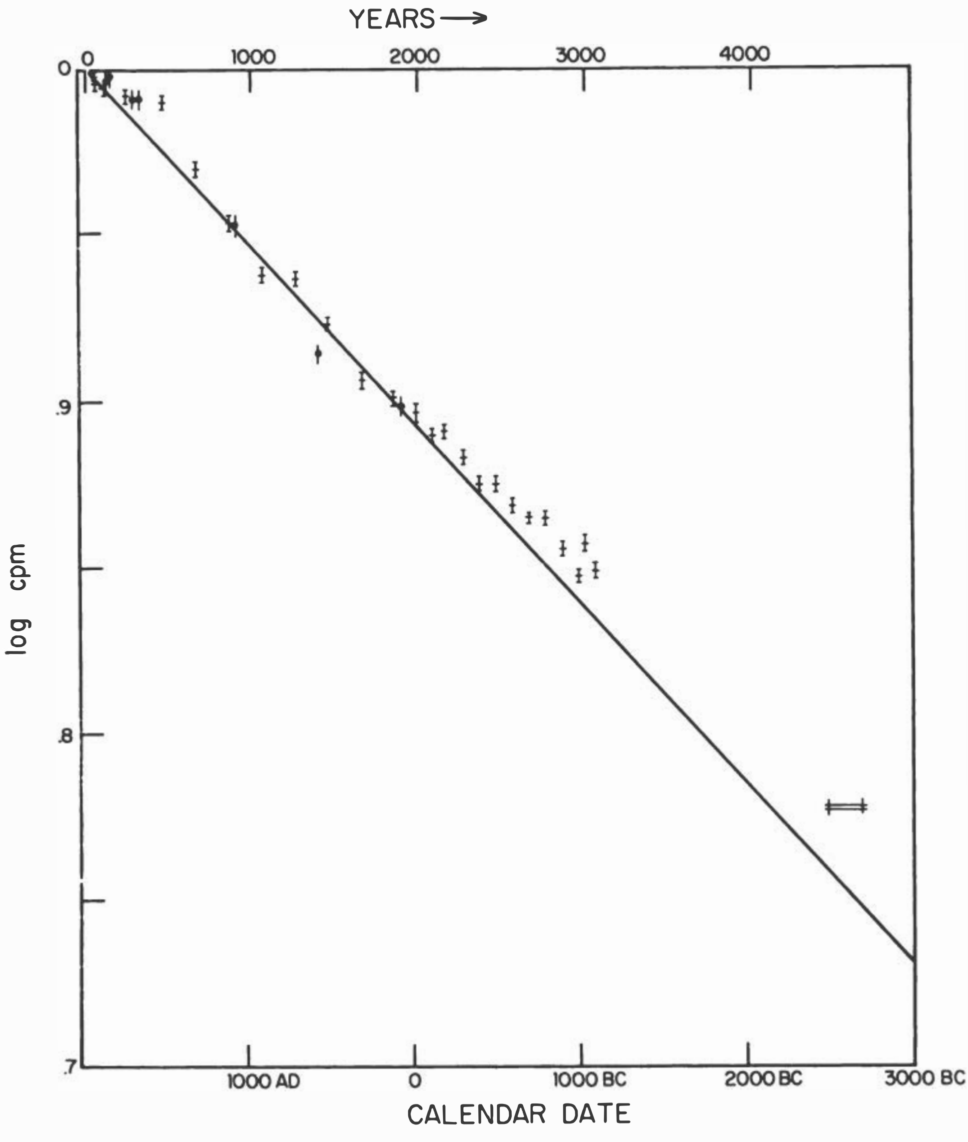


**Figure 2.** **Deviation of empirical results from theoretical line as given in Figure 1. Solid circles show values by Broecker et al. for comparison.**



Copyright © National Academy of Sciences. All rights reserved.

and have concluded that the radiocarbon concentration in Roman times should have been about three percent lower than in the 19th century, if the Earth's magnetic field only began to decline 2000 years ago, which would mean that an object ~2000 years old would be dated about 240 years too old. If the decay in the magnetic field has been an exponential one, and has persisted for the last 4000 years, then the $C^{14}$ inventory two thousand years ago would have been so much lower that the samples of this age would give an age about 1000 years too old. The authors have not considered the possibility of more rapid fluctuations of the Earth's magnetic field and effects of such fluctuations on the $C^{14}$ activity in atmospheric $CO_2$ due to the slowness of isotopic exchange with the bicarbonate in the oceans. Such effects might possibly explain some of the observed data.

The results of the measurements indicate that for samples older than 2000 years the conventional radiocarbon age may appear perhaps as much as ten percent younger than the actual age of the material. For example, the famous Two Creeks wood, dated by Libby (1955) and by other investigators as 11, 350 years old, may actually be some 12, 500 years old; or the maximum extent of the North American ice sheet during the last glaciation measured by the author (Suess 1954) to have occurred between eighteen and nineteen thousand radiocarbon years ago may actually have been reached twenty to twenty-one thousand years ago. However, it is also possible that the observed deviations represent fluctuations from a mean which on the average deviates to a much lesser degree from the true ages.

I am grateful to the following people for their assistance: Dr. G. Bien and Mrs. P. Sandoval for laboratory work; Dr. S. Epstein and Dr. H. Craig for $C^{13}$ determinations; Dr. Terah Smiley for supplying the wood samples. This work was carried out under contract with the Division of Biology and Medicine of the U.S. Atomic Energy Commission.

Broecker: deVries attempted to detect the effects of the eleven-year sunspot cycle in his samples. It is uncertain whether solar activity should enhance or reduce $C^{14}$ production (see papers by Olbert and by Meyer in this volume), but in any event there was certainly too long an integration time in the over-all terrestrial $C^{14}$ inventory, due to the two thousand year average lifetime of a $C^{14}$ nucleus, for deVries to have noticed an effect. There should be a much larger effect on the $C^{14}$ level due to bomb testing--the $C^{14}$ level in this year's rings should be about 20 percent higher than in the 1953 rings. We have already compared 1953 rings with somewhat older rings and have found a one and a half percent contamination, which is not beyond possible experimental error.



Copyright © National Academy of Sciences. All rights reserved.

## REFERENCES

Arnold, J. R. (1960) private communication.

Broecker, W. S., Olson, E. A., and Bird, J. (1959) Nature 183, 1582.

deVries, H. (1958) Proc. Amsterdam Acad. Sci. B61, 1.

Elsasser, W. M., Ney, E. P., and Winkler, J. R. (1956) Nature 178, 1226.

Libby, W. F. (1955) Radiocarbon Dating (University of Chicago Press, Chicago).

Ralph, E. K. (1959) Am. J. of Science, Radiocarbon Suppl. 1, 45.

Suess, H. E. (1954) Science 120, 467.

Thellier, E., and Thellier, O. (1959) Annales de Geophysique 15, 285.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# RADIUM-URANIUM AGE DETERMINATIONS ON MARINE SHELLS

Wallace S. Broecker
Lamont Geological Observatory

The unexplained deviations in $C^{14}$ ages, discussed by Suess, make it even more important to develop an independent check of the $C^{14}$ method for ages greater than 5000 years, beyond which point historical dating and dendrochronology fail. A method originally conceived by Potratz based on inequilibrium in the U series may be suitable for this purpose. This method depends upon the observation that the ratio of $A_{Th^{230}}/A_{U^{238}}$ in recently formed marine carbonates generally seems to be quite low (<0.05). With time, secular equilibrium will be reestablished in accordance with the 80,000 year half-life of $Th^{230}$. A similar systematic relationship exists for $Pa^{231}$ ($3.5 \times 10^4$ year $t_{1/2}$), a member of the $U^{235}$ series.

In order to investigate and, if possible, exploit these methods of determining ages, a program has been begun at Lamont Observatory, measuring $C^{14}$, $U^{235}$ and $U^{238}$, $Ra^{226}$, $Pa^{231}$, $Th^{232}$ and total Ba in samples of marine carbonates. $Ra^{226}$ (1600 year $t_{1/2}$) is expected to come into equilibrium with its parent, $Th^{230}$, in times the order of a few thousand years, and can be measured with high sensitivity by looking at the decay rate of its gaseous daughter $Rn^{222}$. $Th^{232}$ and Ba are measured in order to estimate the initial levels of $Th^{230}$ and $Ra^{226}$ respectively. This allows the reliability of the assumption that $Th^{230}$ and $Ra^{226}$ are initially absent to be checked in each sample.

The calculation of an age from these measurements involves two basic assumptions: that the concentration of $Th^{230}$, $Ra^{226}$ or $Pa^{231}$ at the time of formation is known, and that the shells have been a closed system since their formation. It seems well established, from the work of Sackett and Potratz, that $Th^{230}$ and $Pa^{231}$ are essentially absent in many recently formed marine carbonates. $Ra^{226}$ is present initially and, although it decays to the level at which it is supported by $Th^{230}$ within five to ten thousand years, spuriously long ages could result from excess $Ra^{226}$ in very young shells. This ambiguity can be resolved by measuring the $C^{14}$ age of the sample. Since Ra is known to migrate in marine clays, measurements of $Th^{230}$ as well as $Ra^{226}$ should be made whenever possible. The cross-check provided by the $Pa^{231}$-$U^{235}$ age will also be



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

helpful, although the low isotopic abundance of $U^{235}$ and the relatively short half-life of $Pa^{231}$ indicate that this method has a limited range of applicability.

Measurements have been made on three types of marine carbonates: contemporary shells; shells and corals of known radiocarbon age; carbonates sufficiently old so that radioactive equilibrium should have been attained. A check of the analytical methods on uranium and radium standard samples indicates that standard deviations are ≤10 percent (U) and ≤5 percent ($Ra^{226}$), with sensitivities of 10 ppb U and $2 \times 10^{-6}$ ppb $Ra^{226}$ for analyses of 30 gram carbonate samples.

Table 1 displays the results on contemporary gastropods. Note that the uranium concentration is very low; if this level of U had proved to be characteristic of all shells measured, dating by this method would be very difficult. The shells of non-zero age all have U contents >0.3 ppm. There are two plausible explanations for this discrepancy. That a species effect may be operating is suggested by data of Totsomoto and Goldberg and of Sackett on U in marine carbonates. Shells of other types will be analyzed to check on this. Alternatively, the uranium may be secondary, and

TABLE 1

Results on Contemporary Gastropods

| | | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $Ra^{226}$ / $Ra^{226}$ equil. |
|---|---|---|---|---|
| Jamaica | 1884 | 0.7±0.2 | <0.01 | >2.0 |
| | 1930 | 0.6±0.2 | <0.02 | >1.0 |
| Bahamas | 1880 | 2.0±0.2 | <0.01 | >3.0 |
| | 1950 | 1.1±0.2 | <0.01 | >6.0 |
| Iceland | 1840 | <0.2 | ---- | --- |
| | 1910 | 0.2±0.1 | ---- | --- |
| | 1946 | 0.2±0.1 | ---- | --- |
| Hawaii | 1840 | 0.5±0.2 | 0.04±.02 | 0.36±.20 |
| | 1936(?) | 1.0±0.2 | 0.13±.03 | 0.21±.06 |
| Tahiti | 1880 | 0.3±0.1 | ---- | --- |
| | 1957 | 0.4±0.2 | ---- | --- |



Copyright © National Academy of Sciences. All rights reserved.

might build up with increasing age. A series of identical shells of varying ages is being analyzed to check this hypothesis.

From the levels of $Ra^{226}$ given in Table 1, it seems that ~1 x $10^{-14}$ g/g is a typical value for the initial $Ra^{226}$. It is unlikely that more than ~3 x $10^{-14}$ g/g could have been present in any of the shells studied, since processes of shell-formation almost certainly result in a lower Ra/Ca ratio in shells than in sea-water, judging from the behavior of Sr and Ba in this system. Assuming ~1 x $10^{-14}$ g/g $Ra^{226}$ initially present, errors in the U-Ra ages due to initial Ra should be negligible in shells greater than 5000 years in age.

Results of measurements on very old samples are given in Table 2. A progressive acid leach was used on the Eniwetok coral. Clearly, more data is needed to decide whether the positive deviations from equilibrium ratios are significant.

TABLE 2

U-Ra Measurements on Samples of Very Great Age

| Sample | Material | Fraction % | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $Ra^{226}$ / $Ra^{226}$ equil. |
|---|---|---|---|---|---|
| Potratz Ls. Standard | Mississippian Limestone | 0-100 | 36±1 | 1.00±.03* | 1.00±.05 |
| 584-B | Miocene Coral, Eniwetok | 0-50 | 73±3 | 1.6±.1 | 1.26±.11 |
| | | 50-100 | 58±2 | 1.4±.1 | 1.18±.11 |

*Results by other laboratories using four different techniques average 1.05.

The data presented in Tables 3, 4, and 5 are quite encouraging, although some of the second-order effects are not understood. The agreement between Ra-U and $C^{14}$ ages for Eniwetok Aragonitic Corals (Table 3) indicates that errors due to initial $Ra^{226}$ are probably not important. The data on British Columbia and Spitsbergen shells generally show reasonably good agreement, when the first 10 - 30 percent of the sample is neglected. A puzzling feature is the systematic decrease, often by large factors, in the $Ra^{226}$ content during the first stages of the progressive acid leach.



Copyright © National Academy of Sciences. All rights reserved.

TABLE 3

Aragonitic Coral from Eniwetok Atoll

| Sample # Fraction % | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $\frac{Ra^{226}}{Ra^{226}_{equil.}}$ | Ra-U Age ($10^3$ y) | $C^{14}$ Age ($10^3$ y) |
|---|---|---|---|---|---|
| 482-A | | | | | |
| 0-100 | 3.5±.3 | 2.4±.2 | 0.040±.004 | 4.8±0.5 | 3.8±0.2 |
| 482-B | | | | | |
| 0-100 | 6.4±.8 | 2.8±.2 | 0.062±.008 | 7.3±0.9 | 5.6±0.2 |
| 482-C | | | | | |
| 0-100 | 5.0±.3 | 3.0±.2 | 0.047±.004 | 5.7±0.5 | 5.9±0.2 |

TABLE 4

Results on British Columbia Shells

| Sample # Fraction % | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $\frac{Ra^{226}}{Ra^{226}_{equil.}}$ | Ra-U Age ($10^3$ y) | $C^{14}$ Age ($10^3$ y) |
|---|---|---|---|---|---|
| 391-C | | | | | |
| 0-30 | 8.7±.4 | 0.33±.03 | 0.74±.08 | 155±35 | |
| 30-43 | 1.5±.2 | 0.43±.04 | 0.10±.01 | 12±2 | 12-0.3 |
| 43-100 | 0.47±.10 | 0.18±.03 | 0.072±.014 | 8.6±1.6 | |
| 475-A | | | | | |
| 0-30 | 42±2 | 1.15±.11 | 1.01±.12 | > 200 | >39 based |
| 0-30 | 40±3 | 1.14±.10 | 0.97±.11 | > 200 | on other |
| | | | | | $C^{14}$ dates |
| 30-43 | 10±.5 | 1.07±.11 | 0.27±.03 | 36±4 | and strati- |
| 30-43 | 8.4±1.0 | 0.92±.10 | 0.25±.03 | 32±4 | graphic |
| | | | | | evidence; |
| 43-100 | 3.5±.3 | 0.29±.02 | 0.34±.04 | 48±5 | should be |
| 43-100 | 2.5±.3 | 0.20±.02 | 0.35±.04 | 49±6 | <50 |
| 475-B | | | | | |
| 0-30 | 7.6±.6 | 0.28±.03 | 0.75±.09 | 160±50 | >39 |
| 30-42 | 4.7±.4 | 0.46±.04 | 0.28±.03 | 38±5 | same age |
| 42-100 | 4.0±.3 | 0.36±.04 | 0.32±.03 | 44±5 | as 475-A |



Copyright © National Academy of Sciences. All rights reserved.

TABLE 5

Results on Spitsbergen Shells

| Sample # Fraction % | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $\frac{Ra^{226}}{Ra^{226}_{equil.}}$ | Ra-U Age ($10^3$ y) | $C^{14}$ Age* ($10^3$ y) |
|---|---|---|---|---|---|
| 572-A | | | | | |
| 0-10 | 7.1±.5 | 0.62±.05 | 0.32±.03 | 44±5 | --- |
| 10-55 | 4.9±.2 | 1.29±.10 | 0.11±.01 | 13±1 | 9.4±.2 |
| 55-100 | 3.7±.2 | 1.64±.13 | 0.064±.006 | 7.8±.8 | 9.7±.2 |
| 572-B | | | | | |
| 0-10 | 6.8±.3 | 0.44±.03 | 0.43±.03 | 65±6 | --- |
| 10-55 | 3.3±.2 | 0.69±.04 | 0.13±.01 | 16±1 | 9.1±.2 |
| 55-100 | 2.1±.2 | 0.54±.04 | 0.10±.01 | 11±1 | 9.5±.2 |
| 572-C | | | | | |
| 0-10 | 68±3 | 1.2±.1 | 1.54±.13 | -- | |
| 10-45 | 65±3 | 1.2±.1 | 1.44±.13 | -- | 34±1 |
| 45-100 | 50±3 | 1.7±.1 | 0.83±.07 | 208±60 | 34±1 |

* Measurements by Ingrid Olsson of the Uppsala, Sweden, Radiocarbon Laboratory.

This pattern is very common and does not seem to be reflected in the uranium contents. However, simple surficial contamination by $Ra^{226}$ may not be the answer, since in only one case (572-C) is there more $Ra^{226}$ than would be expected at equilibrium, so that some correlation with $U^{238}$ is apparent. The origin of this $Ra^{226}$ distribution must be considered an open, and perhaps important, question.

The discrepancy between Ra-U and $C^{14}$ ages for sample 572-C (Table 5) is probably due to inseparable contamination with modern $C^{14}$. Stratigraphically, 572-C is certainly considerably older than 572-A or 572-B and may be from a previous interglacial period, which would agree with the Ra-U ages.

Table 6 gives data on miscellaneous shell samples in which the two ages do not agree at all. The $C^{14}$ ages are well-established. The most likely cause for this disagreement seems to be initial $Th^{230}$; comparisons of $Th^{232}$ contents are in progress to determine whether this could be the case. Another interesting feature of this data is that acid leaches of



Copyright © National Academy of Sciences. All rights reserved.

TABLE 6

Results on Miscellaneous Shells

| Sample # Fraction % | $Ra^{226}$ $10^{-14}$ g/g | $U^{238}$ $10^{-6}$ g/g | $\frac{Ra^{226}}{Ra^{226}_{equil.}}$ | Ra-U Age ($10^3$ y) | $C^{14}$ Age ($10^3$ y) |
|---|---|---|---|---|---|
| 521 (Borneo) | | | | | |
| 0-10 | 2.7±.3 | 0.39±.03 | 0.19±.02 | 23±2 | |
| 10-55 | 1.7±.2 | 0.34±.03 | 0.14±.02 | 17±2 | 5.6±0.2 |
| 55-100 | 1.3±.1 | 0.41±.03 | 0.087±.01 | 11±1 | |
| 248A (Ellsmere Island) | | | | | |
| 0-10 | 8.3±.4 | 0.71±.05 | 0.32±.03 | 45±5 | |
| 10-55 | 2.8±.2 | 0.54±.04 | 0.15±.02 | 18±2 | 7.2±0.2 |
| 55-100 | 2.8±.2 | 0.57±.04 | 0.14±.01 | 17±2 | |
| 581 (Argentine Shelf) | | | | | |
| 0-15 | 22±1 | 1.8±.15 | 0.33±.03 | 43±5 | |
| 15-100 | 9.6±5 | 0.68±.05 | 0.38±.03 | 56±6 | 16±3 |

sample 581 do not evidence a systematic, uncompensated decrease in $Ra^{226}$. If this can be correlated with the fact that this sample has probably never been above sea level, it may indicate that deposition of $Ra^{226}$ from ground-water is crucial in developing the pattern of $Ra^{226}$ concentration noted in the other samples.

The Ra-U age method has also been applied to materials believed, on various grounds, to be of last interglacial age. Ages obtained range from 100,000 years to 134,000 years, which is in reasonable agreement with dates for the last interglacial age estimated from extrapolations of sedimentation rates for ocean cores.

In conclusion, although the Ra-U method may not be capable of providing a precise independent check of the $C^{14}$ ages, it is promising as a means of extending the range of age measurements in marine carbonates to perhaps 200,000 years. Much accessory data (e.g., $Th^{230}$, $Th^{232}$, $Pa^{231}$ contents) is needed before the method can be applied with confidence.



Copyright © National Academy of Sciences. All rights reserved.

Problems Related to Interplanetary Matter
http://www.nap.edu/catalog.php?record_id=18683

# OXYGEN ISOTOPE MEASUREMENTS IN GLACIAL ICE

S. Epstein
California Institute of Technology

Due to the fact that the vapor pressure of $H_2O^{18}$ in equatorial waters is about 0.8 percent less than that of $H_2O^{16}$, the water vapor in tropical air masses is about eight mils depleted in $O^{18}$ relative to mean water. [The depletion or enrichment in permils is computed from the following formula:

$$\delta = \left| \frac{(O^{18}/O^{16})_{sample}}{(O^{18}/O^{16})_{ocean}} - 1 \right| \times 1000 \qquad (1)]$$

As the water vapor in the air masses moves to higher latitudes, it undergoes further isotopic fractionation, since $H_2O^{18}$ precipitates preferentially to $H_2O^{16}$. The degree of fractionation depends upon the thermal gradient through which the air mass moves; the thermal gradient produces a separation equivalent to a multistage distillation. The path followed by the air mass should not greatly affect the fractionation. Isotopic measurements performed on rainwater or snow obtained at different locations--at Hawaii, where $\delta = -1$ to $-3$; at Pasadena ($\delta = -2$ to $-16$); Saskatchewan ($\delta = -10$ to $-26$); Greenland ($\delta = -21$ to $-47$), and the South Pole ($\delta = -44$ to $-51$)--reveal a striking latitude dependence. Hundreds of measurements were performed, and the $\delta$ values quoted represent extremes for that location; experimental errors were just a few tenths of a mil. The large South Pole negative $\delta$ values are probably a result not only of a steep thermal gradient between equator and pole but also reflects the lack of reevaporation from the cool oceanic waters over which the southward-bound air masses pass.

Because the steepness of the thermal gradient determines the extent of isotope fractionation and is seasonal, it was decided to measure isotope ratios at various depths in a glacier. Figure 1 shows the $\delta$ values found in the Greenland glacier at depths from one to five meters. These depths represent the years 1957, 1956, 1955, 1954, and 1953. A pronounced summer-winter effect is easily seen in the results, together with considerable fine structure representing changing isotope ratios during the course of a single storm. During the years 1956 and 1955 some surface melting is known to have taken place for a few weeks during the summer.



Copyright © National Academy of Sciences. All rights reserved.