On the Influence of Cosmic Radiation on the Isotopic Composition of the Elements

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Abstract

When cosmic radiation interacts with the atomic nuclei of the atmosphere, it produces a great number of stable as well as radioactive isotopes. The radioactive isotopes are fairly easy to detect but the production of stable isotopes can only be detected by the changes in isotopic composition of the elements that might be caused by this effect. The amounts of Li^6, Ne^{21}, and A^{36} that are produced in this way have been calculated. If the mean intensity of cosmic radiation in the past has been the same as at present it turns out that the production of these isotopes causes a change in isotopic composition which is barely detectable.

A study of this effect gives a possibility of obtaining information about the cosmic-ray intensity in the past. If no cosmic-ray effect is found, it shows that the mean cosmic-ray intensity in the past could not have been considerably higher than at present. If the cosmic-ray intensity during the earlier stages in the earth's evolution were higher than at present, one could expect easily detectable changes in the isotopic composition of some elements.

Introduction

It is well known that cosmic radiation interacts with the atomic nuclei of the atmosphere, producing a number of radioactive isotopes. C^{14} was the first one to be found. In recent years a number of other isotopes have been found: H^3, Be^7, P^{32}, S^{35}, and Cl^{39}. Cosmic radiation will, of course, also produce stable isotopes and this might slightly change the isotopic composition of some elements. These changes are expected to be very small, but there does not seem to be any quantitative estimate of this effect. The only isotopes for which a discernible effect can be expected are, of course, those of very small abundance.

If cosmic radiation has any influence on the isotopic composition of an element, its composition will vary from place to place. Let us assume, for example, that cosmic radiation changes the composition of an element which is a constituent of the atmosphere. In order to detect this change one must compare the composition of a sample from the atmosphere with a sample from a place which has not been influenced by cosmic radiation. This can be, for example, traces of the primordial atmosphere enclosed in igneous rock. Another case might be that the isotope produced by cosmic radiation is brought into solution in the oceans. The isotopic composition of the element in question will then be different in sea water and in minerals.

If this effect can be found, it has a very interesting application as a method of measuring the mean cosmic-ray intensity during the time which has passed since the formation of the earth. The present intensity of cosmic radiation

is well known. The various reactions which produce the isotopes of interest in this case are also well known. The present production rate of a certain isotope can, therefore, be calculated with a fairly high accuracy. If it can be shown that cosmic radiation has caused a change of the isotopic composition and if this change can be measured, it gives directly the mean intensity over the period during which this process has been going on. An investigation of this problem is of value, even if no change of the isotopic abundance due to cosmic radiation can be found. It gives, at any rate, an upper limit for the mean intensity. A knowledge of the mean intensity of cosmic radiation, or an upper limit, is definitely of great interest in connection with the problem of the origin of cosmic radiation.

Since the effect in question must be very small, it is of interest to know the accuracy in the determination of the relative isotopic abundance. The accuracy is considerably higher for measurements of small changes in abundance than for absolute determinations. UREY and his collaborators (1950) claim an accuracy of 0.02 % for a mass spectrometer specially built for this type of measurement.

Connected with this is the problem of the variations in isotopic composition which have been found recently in several elements. They are probably caused by isotopic fractionation in the chemical reactions occurring in nature. These variations are greatest—several per cent —in elements like carbon and sulphur, which take part in cyclic processes, especially processes in living matter. The smallest variations can be expected in the rare gases, which do not take part in any chemical reactions. This is fortunate, since two of the three elements to be dealt with in the present discussion are rare gases. Hence the small changes caused by the cosmic radiation might show up without interference from other effects.

In the following the elements which should show the greatest variations will be discussed.

Lithium

When cosmic-ray particles interact with the nitrogen and oxygen nuclei of the atmosphere, a great number of nuclei are produced. Among them are the two lithium isotopes with the mass numbers 6 and 7. The rate of production is roughly the same for the two isotopes. The lithium produced in the atmosphere is brought down to the surface of the earth and the greater part will eventually be dissolved in the oceans. Natural lithium contains 7.5 % Li^6 and 92.5 % Li^7. The lithium production by the cosmic radiation will, therefore, make the relative isotopic abundance of Li^6 higher in sea water than in lithium minerals. The present production rate of Li^6 is calculated in the following way. The interaction of the cosmic radiation with nitrogen has been studied by BROWN (1954). He used a cloud chamber filled with nitrogen and observed the stars produced in the gas. From the size distribution of the stars observed, one can estimate the number of interactions that lead to Li^6. The absolute rate of interaction was also measured. The altitude variation of the star production is known. (LORD 1951, BENIOFF 1956.) Hence the total production of Li^6 can be calculated. It turns out to be 0.3 atoms Li^6 per cm^2 per sec.

The production of Li^6 has been going on for a length of time which should be roughly the same as the age of the earth. It is not necessary to know the exact value—the calculation is necessarily rather approximate. The value $4 \cdot 10^9$ years has been used. The production of Li^6 during this time is $4 \cdot 10^{16}$ atoms per cm^2, assuming that the mean intensity of cosmic radiation has been the same as the present intensity. The oceans contain about $1.7 \cdot 10^{20}$ atoms Li^6 per cm^2 averaged over the entire surface of the earth. The change in isotopic abundance is then about 0.03 %. Hence even if the cosmic-ray intensity has been constant, a change is obtained which should be possible to detect. If the intensity were higher during the earlier stages of the evolution, a considerable effect could be expected.

There are, however, some complications. A certain amount of lithium has been removed from the oceans through adsorption by mud and other finely divided constituents of the sea water. In fact, it has been estimated that only 0.2 % of the lithium which has been dissolved during the weathering of rocks is in solution in the oceans (GOLDSCHMIDT 1954). This does not mean that the rest has been removed from the sea water. A great part has been absorbed by the weathering products *in situ* or has been removed from the fresh

water before reaching the oceans. In a recent investigation HORSTMAN (1957) concludes that "lithium probably does not enter the sea in solution in any appreciable quantity". Hence this effect will probably not influence the order of magnitude of the calculated change.

Another complication is the low atomic weight of lithium. The various isotopic fractionation processes working in nature are probably more effective for lithium than for the heavier elements, concealing the influence of the cosmic radiation. Still, it seems worth while to try to find the cosmic-ray effect. There does not seem to exist any measurements of the isotopic composition of lithium in sea water.

Neon

Cosmic-ray particles also interact with the argon in the atmosphere, giving a great number of isotopes, among them Ne^{21}. This isotope is the rarest of the neon isotopes, its relative abundance being 0.25 %. The abundance of neon in the atmosphere is low, and a relatively small production of neon by the cosmic radiation is enough to change the isotopic composition. The production of the three neon isotopes should be roughly the same and the relative abundance of Ne^{21} will, therefore, increase. The cosmic-ray induced reactions in argon have been studied by BROWN (1954) and it is possible to calculate the production rate of Ne^{21} in the same way as for Li^6. The result is $1.2 \cdot 10^{-3}$ atoms per cm^2 per sec. or $1.5 \cdot 10^{14}$ atoms per cm^2 in $4 \cdot 10^9$ years, if the mean intensity has been the same as the present intensity. This figure has to be compared with the atmospheric content of Ne^{21}, which is $9 \cdot 10^{17}$ atoms per cm^2. The cosmic-ray production is according to this estimate 0.02 % of the total amount of Ne^{21}. The cosmic-ray effect can be investigated by comparing the composition of atmospheric neon with the neon contained in igneous rocks.

It has been suggested bu SUESS (1949) that a certain isotopic fractionation occurs because of the escape of neon from the gravitational field of the earth. One would expect the same effect for other gases like nitrogen and gaseous carbon compounds, but no effect of this kind has been experimentally verified. It can be determined, if it exists, by comparing the relative abundance of the isotopes Ne^{20} and Ne^{22} in atmospheric neon with the neon contained in igneous rocks. The correction due to this effect for Ne^{21} can then be calculated, and it would still be possible to find the cosmic-ray effect if it were large enough.

There does not seem to be any measurements which can be used for the present purpose. WETHERILL (1954) has measured the isotopic composition of neon from some uranium minerals and finds that the abundance of Ne^{21} is much higher than normal. This must be due to some nuclear reaction, probably O^{18} (α, n) Ne^{21}.

Argon

The spallation in the atmosphere of the most abundant argon isotope A^{40}, gives among other products the two argon isotopes A^{38} and A^{36}. The production of A^{38} is predominant. It can be estimated in the same way as described above. The result is $3 \cdot 10^{14}$ atoms per cm^2 in $4 \cdot 10^9$ years. The amounts of A^{36} and A^{38} in the atmosphere are $7 \cdot 10^{20}$ and $1.4 \cdot 10^{20}$ atoms per cm^2, respectively. The production by cosmic radiation is evidently so small that it is impossible to detect unless the intensity was very much higher in the past.

Argon can, however, be produced in a quite different way. When cosmic-ray neutrons are absorbed by the earth's crust and the water of the oceans, a great number of radioactive nuclei are formed. The situation is especially simple for the oceans. The fast neutrons are rapidly slowed down by the water. Among the major constituents of sea water, chlorine has by far the largest cross-section for slow neutrons. A great number of the neutrons are, therefore, absorbed by chlorine, giving mainly Cl^{36}, which decays to A^{36}. Also the neutrons absorbed by solid material will give A^{36} to a great extent. The amount of A^{36} produced has been estimated in the following way. LATTIMORE (1951) has measured the neutron flux using emulsions loaded with boron. It can also be obtained from the investigations of cosmic-ray stars (BROWN 1954, HARDING 1949, GEORGE and EVANS 1950). The values obtained agree very well. The amount of A^{36} produced can then be obtained knowing the average composition of the earth's crust and sea water and the cross-sections of the various elements.

The result is $3 \cdot 10^{14}$ atoms per cm^2 in $4 \cdot 10^9$ years if the mean intensity is assumed to be the same as the present one. It is too small to influence the isotopic composition of argon.

There is, however, some possibility that the cosmic-ray production of A^{36} has been higher than calculated above. The production by cosmic-ray neutrons is not very efficient, since only a very small part of the total number of neutrons is produced at the surface of the earth. One might ask if the conditions in the past were more favourable for neutron absorption by chlorine. The following possibility is of interest. It is generally assumed that the present atmosphere is of secondary origin and that its constituents have been retained by the earth in the form of some suitable chemical compound during the early stages of the evolution. For nitrogen NH_4Cl has been suggested. This means that the atmosphere, during some part of its evolution, might have contained NH_4Cl (UREY 1952). Since chlorine has a large cross-section for neutron absorption, a small amount of NH_4Cl in the atmosphere would be enough to increase the production of A^{36} very much. In fact most of the neutrons would give A^{36}. The total number of neutrons is very well known from the investigations on C^{14}. If these conditions existed for, let us say $2 \cdot 10^9$ years, the amount of A^{36} produced would be $1.5 \cdot 10^{17}$ atoms per cm^2. This is 0.02 % of the atmospheric content of A^{36}.

There are some measurements by FLEMING and THODE (1953) and by WETHERILL (1954) on the isotopic composition of argon from minerals. They show an excess of A^{38} relative to A^{36} compared with atmospheric argon. Unfortunately the minerals investigated are uranium or thorium minerals. The change has therefore by these authors been attributed to nuclear reactions caused by the α-particles. The only possible reaction is Cl^{35} (α, p) A^{38}. A calculation shows, however, that this interpretation is met by some difficulties. The cross-section for this reaction is not known experimentally. The cross-section for capture of α-particles in Cl^{35} gives an upper limit for the cross-section of the (α, p) reaction. The capture cross-section has been calculated from the formula given by BLATT and WEISSKOPF (1952). It is known that such calculations give good agreement with the experiments. The yield of the (α, p) reaction in a thick target with the composition of pitchblende was then calculated for the energies of the α-particles from uranium. It then turns out that in order to get the measured excess of A^{36}, the amount of chlorine in the mineral must be 0.25 %. This is much higher than what can be expected (DOELTER 1929). Even if there were so much chlorine, it would not solve the difficulty. The neutron flux, which is known to exist in uranium minerals, gives A^{36} by the reaction Cl^{35} (n, γ) Cl^{36}. The neutron flux can be calculated from the amount of neutron induced fission in the mineral. It then turns out that the ratio of A^{36} to A^{38} produced is 20. This calculation has been made using the values reported for pitchblende from the Belgian Congo. There are, of course, some uncertainties in the calculation, but it seems unlikely that the order of magnitude is wrong. Hence it does not seem to be possible to explain the excess of A^{38} by nuclear reactions in the mineral.

ALDRICH and NIER (1948) have published mass spectra for atmospheric argon as well as argon from the mineral Langbenit. Unfortunately there is a background peak at the mass number 38 and it is therefore impossible to get the $\frac{A^{36}}{A^{38}}$ ratio directly from the published spectra. One can, however, calculate the background peak from the known composition of atmospheric argon. If one then makes the reasonable assumption, that the relative heights of the background peaks are the same in the two spectra, it is possible to get the $\frac{A^{36}}{A^{38}}$ ratio for the argon sample from Langbenit. The value obtained is 2.5 compared with 5.35 for atmospheric argon. In this case, too, there seems to be an excess of A^{38}.

From this one can perhaps draw the conclusion that there is evidence that argon in some minerals shows an anomalously high A^{38} content compared with atmospheric argon. An explanation of this could be that the A^{36} content in atmospheric argon has increased owing to cosmic radiation. Clearly the experimental material is too meagre to allow any definite conclusions. A careful investigation of the composition of argon from various minerals would be very interesting.

Conclusions

Concluding one can say that the influence of cosmic radiation on the isotopic composition of some elements is small but not quite negligible. If the mean cosmic-ray intensity in the past has been the same as at present, the changes in isotopic composition are barely detectable. They would probably be masked by other small effects, however. If no cosmic-ray effect is found, it shows that the cosmicray intensity in the past could not have been considerably higher than at present.

If the cosmic-ray intensity during the earlier stages in the earth's evolution were higher than at present, one could expect easily detectable changes in isotopic composition. In this connection it is important to note that the magnetic field of the earth reduces the average cosmic-ray intensity by almost a factor of ten. If the magnetic field in the past was weaker than at present, it would imply a higher cosmic-ray intensity during the earlier stages in the earth's evolution.

REFERENCES

ALDRICH, L. T. and NIER, A. O., 1948: Argon 40 in potassium minerals. *Phys. Rev.* **74**, pp. 876—877.

BENIOFF, P. A., 1956: Cosmic-ray production rate and mean removal time of beryllium-7 from the atmosphere. *Phys. Rev.* **104**, pp. 1122—1130.

BLATT, J. M., and WEISSKOPF, V. F., 1952: *Theoretical Nuclear Physics.* New-York: J. Wiley & Sons. 864 p.

BROWN, W. W., 1954: Cosmic-ray nuclear interactions in gases. *Phys. Rev.* **93**, pp. 528—534.

DOELTER, C., 1929: *Handbuch der Mineralchemie:* Band IV:2 Dresden und Leipzig 1929, Th. Steinkopf. 1494 p.

FLEMING, W. H., and THODE, H. G., 1953: Argon 38 in pitchblende minerals and nuclear processes in nature. *Phys. Rev.* **90**, pp. 857—858.

GEORGE, E. P., and EVANS, J., 1950: Observations of cosmic-ray events in nuclear emulsions exposed below ground. *Proc. Phys. Soc.* **63** A, pp. 1248—1264.

GOLDSCHMIDT, V. M., 1954: *Geochemistry.* Oxford: Clarendon Press.

HARDING, J. B., LATTIMORE, S., LI, T. T., and PERKINS, D. H., 1949: Absorption of star-producing radiations under ice. *Nature* **163**, pp. 319—320.

HORSTMAN, E. L., 1951: The distribution of lithium, rubidium, and cesium in igneous and sedimentary rocks. *Geochim. and Cosmochim. Acta* **12**, pp. 1—28.

LATTIMORE, S., 1951: Rate of production of neutrons in ice by cosmic-rays. *Phil. Mag.* **42**, pp. 331—337.

LORD, J. J., 1951: The altitude and latitude variation in the rate of occurrence of nuclear disintegrations produced in the stratosphere by cosmic-rays. *Phys. Rev.* **81**, pp. 901—909.

SUESS, H. E., 1949: Die Häufigkeit der Edelgase auf der Erde und im Kosmos. *J. Geol.* **57**, pp. 600—607.

WETHERILL, G. W., 1954: Variations in the isotopic abundances of neon and argon extracted from radioactive minerals. *Phys. Rev.* **96**, pp. 679—683.

UREY, H. C., MCKINNEY, C. R., MCCREA, J. M., EPSTEIN, S., and ALLEN, H. A., 1950: Improvements in mass spectrometers for the measurement of small differences in isotopic abundance ratios. *Rev. Sci. Instr.* **21**, pp. 724—730.

UREY, H. C., 1952: *The Planets.* New Haven: Yale University Press. 245 pp.