- 13. G. B. Udintsev, I. G. Boichenko, V. F. Kanaev, The Bottom Relief of the Bering Sea (research translation, Air Force Cambridge Research Laboratories, Bedford, Mass., bridge Research Laboratories, Bedford, Mass., 1959), pp. 1-88; K. Venkatarathnam, Univ. Wash. Spec. Rep. 41 (1969); U.S. Geol. Surv. Open-File Rep. (1971); M. S. Grim and D. A. McManus, Mar. Geol. 8, 293 (1970).
 14. M. L. Jackson, Soil Chemical Analysis (Prentice-Hall, Englewood Cliffs, N.J., 1958), pp. 210, 225.
- 219-225. 15. W. S. Broecker and J. L. Kulp, *Amer.*
- Antiquity 22, 1 (1956).
 16. E. L. Hamilton, U.S. Navy Electron. Lab. Rep. 1283 (1965).

Alpha Particles in Solar Cosmic Rays over the Last 80,000 Years

Abstract. Present-day (1967 to 1969) fluxes of alpha particles from solar cosmic rays, determined from satellite measurements, were used to calculate the production rates of cobalt-57, cobalt-58, and nickel-59 in lunar surface samples. Comparisons with the activities of nickel-59 (half-life, 8×10^4 years) measured in lunar samples indicate that the long-term and present-day fluxes of solar alpha particles are comparable within a factor of approximately 4.

The solar helium abundance is an important parameter for astrophysical considerations of stellar burning processes. Among the techniques used to investigate the helium abundance near the surface of the sun have been spectroscopic studies of prominences and the chromosphere (1) and measurements of the composition of the solar wind (2) and solar cosmic rays (3).

17. F. F. Koczy, Geochim. Cosmochim. Acta 1,

The authors thank D. A. McManus for his criticism of the manuscript, R. W. Roberts and D. Kachel for their assistance in the

laboratory analyses, and R. J. Echols for his help with the biogenic components. Much of

the interpretation was done while H.J.K. was

supported by a fellowship from Texaco, Inc. Research supported by NSF grants GA-11126

and GA-28002. Contribution No. 677, Depart-

ment of Oceanography, University of Wash-

73 (1951).

ington.

14 November 1972



Fig. 1. Fluxes of alpha particles produced from seven large solar flares in the period 28 May 1967 to 1 January 1970. The fluxes plotted are summed over the duration of the individual events. The inset contains the average alpha particle spectrum over the approximately 900-day period.

Inferences about the helium abundance deep within the sun have been drawn from the results of the ³⁷Cl solar neutrino experiment (4).

Spectroscopic measurements indicate that the number ratio of alpha particles to protons (α /p ratio) in prominences and the chromosphere is approximately 0.06 (1). Another analysis of the solar spectrum yielded $\alpha/p^{\circ} \sim 0.1$ (5). Although the helium composition of the solar wind varies a great deal, the α/p ratio infrequently rises above about 0.1 (6) and is about 0.045 from longterm measurements (2). From the rather low limits set on the solar neutrino fluxes, a value of $\alpha/p \approx 0.07$ is obtained (4).

Measurements of the fluxes of alpha particles in solar cosmic rays by rocket and balloon techniques during solar cycle 19 (April 1954 to September 1964) seemed to indicate that the fluxes of alpha particles and the α/p ratio differed from flare to flare. When particles of equal energy per nucleon are compared, the α/p ratio changed even during a single flare event (3). These results have been confirmed for lowerenergy ($\lesssim 10$ Mev/nucleon) alphas during the maximum of solar cycle 20 (which began in October 1964) by continuous measurements made from satellites during the events (7, 8). Further results have shown that frequently the α/p ratio during a single flare event is constant only when the particles are compared on the basis of equal energy per charge, but that this ratio also changes from flare to flare (7, 9). From these cosmic ray studies, the α/p ratio is generally found to be less than about 0.1 for particles of equal energy per nucleon.

It is desirable to obtain information about solar conditions, and particularly the solar helium abundance, in the past. The fluxes of energetic particles from ancient solar flares can be studied by measuring the long-lived radionuclides produced by these particles in extraterrestrial matter. No radionuclides produced by solar cosmic rays have been observed in meteorites, since their original surfaces undergo considerable ablation in the earth's atmosphere. Cosmogenic radionuclides are also produced in cosmic dust (micrometeoroids) in interplanetary space (10, 11). Since meteoric material continuously falls onto the earth and is accumulated in ocean sediments, measurements have been made in marine sediments for cosmogenic radionuclides (11, 12). There are

SCIENCE, VOL. 179

considerable uncertainties in the results for marine sediments, both in the experimental measurements (12) and in the rates of the accretion of cosmic dust (10).

The acquisition of samples of lunar material through the several Apollo missions has made it possible to investigate the solar proton flux over the past several million years. In brief, the analyses for both short-lived (half-life: ⁵⁶Co, 77 days; ²²Na and ⁵⁵Fe, both 2.6 years) and long-lived (²⁶Al, 7.4 × 10⁵ years; ⁵³Mn, 3.7 × 10⁶ years) radionuclides produced by proton-induced reactions appear to indicate that solar flares have produced cosmic ray protons with similar fluxes and spectra over the past 1 million years (13–15).

Information about the abundance of helium in solar cosmic rays in the past can be obtained by comparing presentday fluxes of solar alpha particles, determined by satellite measurements, with those inferred from an analysis of long-lived radionuclides produced by alpha-induced reactions in lunar material. The best reactions with which to observe the effects of solar alpha particles are the ones that produce the radionuclides ⁵⁹Ni (half-life, 8×10^4 years), ⁵⁸Co (72 days), and ⁵⁷Co (271 days) by alpha particle bombardment of iron. Although most of the bombardment-produced radionuclides found in lunar materials result from energetic solar protons and galactic cosmic ray particles (16), the low abundances of the elements Co and Ni in lunar samples (approximately 100 parts per million) compared to the high abundance of Fe (approximately 10 percent by weight) means that ⁵⁹Ni, ⁵⁸Co, and ⁵⁷Co are mainly produced by solar alpha particle reactions with iron. The important alpha-induced nuclear reactions are 56 Fe(α ,n) 59 Ni (threshold energy, 1.4 Mev/nucleon), 56 Fe(α ,pn)-⁵⁸Co (3.4 Mev/nucleon), and ⁵⁶Fe- $(\alpha, p2n)^{57}$ Co (5.6 Mev/nucleon) (17). Only the ⁵⁷Co is produced in significant quantities from other sources; about half of the 57Co observed in lunar samples is produced by solar protons by the ⁵⁷Fe(p,n)⁵⁷Co reaction.

Cobalt-57 has been measured in the top surface layers of lunar rocks 10017 (13) and 12002 (14). Cobalt-58 has not yet been measured in lunar samples. The detection of 5^{8} Co is made difficult by its short half-life and by the similarity of its decay scheme to that of 77-day 5^{6} Co, which is produced in large quantities by solar protons (13, 23 MARCH 1973



14). Nickel-59 was measured in the Apollo 11 bulk soil sample 10084 (13), and a preliminary value has been reported for the top millimeter of rock 12002 (18). Nickel-59 is a useful nuclide to study because its long half-life goes well beyond present-day alpha measurements. In addition, its half-life is only one-tenth that of 2^{6} Al, which was used in the solar proton comparisons (13-15) and thus provides an intermediate reference point for studies of possible variability in the production of solar cosmic rays.

Information on the present-day (1967 to 1969) production and spectra of low-energy alpha particles in solar flares has been obtained from measurements made with instruments flown by Bell Laboratories on the Explorer 34 and Explorer 41 satellites. Alpha particles were distinguished from electrons and protons by energy loss and range as well as by the use of an on-board particle identifier (19). Plotted in Fig. 1 are spectra of the total alpha particle fluxes measured after seven of the largest solar events that occurred in the interval 28 May 1967 to 1 January 1970. These seven spectra have predominantly power-law dependences on energy, with the exponent in the power law ranging in value from -1.5 to -3.3. The horizontal "error bars" indicate the widths of the differential energy channels.

The inset in Fig. 1 is the timeaveraged spectrum of alpha particles measured over the approximately 900 days that include the seven events. This time-averaged alpha spectrum, obFig. 2. The differential flux of solar alpha particles at several depths in the mocn, calculated for an isotropic bombardment of a semi-infinite plane. The incident flux (0 g cm^{-2}) is the time-averaged flux shown in the inset of Fig. 1, integrated over 2π . The production rates for the formation of the radionuclides ⁵⁶Ni, ⁵⁸Co, and ⁵⁷Co from iron for these alpha particle fluxes are shown as a function of the depth in the moon. (Note the break in the depth scale at 0.02 g cm⁻².)

tained near the peak of solar cycle 20, has an $\vec{E}^{-1.9}$ energy dependence over the range of approximately 1 to 12 Mev/nucleon. Over one-half of the alphas included in this averaged spectrum were produced in the three largest events, 12 April 1969 (day 102), 18 November 1968 (day 323), and 6 July 1968 (day 188). There were additional fluxes of alphas from numerous flares in the time interval covered by the spectra of Fig. 1, but their inclusion would not increase the fluxes in the inset spectrum by more than a factor of approximately 2. This in part compensates for the fact that the approximately 3-year period sampled here occurred during a period of solar maximum and thus does not include any data for a period of solar minimum, when the solar alpha particle fluxes are expected to be much smaller. Thus, it is reasonable to compare the average flux shown in the inset of Fig. 1 with the long-term flux inferred from the ⁵⁹Ni measurements (20). An examination of the time-averaged proton fluxes over the 900-day interval indicates that the α/p ratio was about 0.02 to 0.04 for particles of equal energy per nucleon (21).

The rates of production of the radionuclides ⁵⁹Ni, ⁵⁸Co, and ⁵⁷Co in lunar samples by solar alpha particles can be calculated by using the model of Reedy and Arnold (16). If we assume only ionization energy losses and an isotropic flux for the incident alpha particles, the differential flux of alpha particles can be calculated for various depths in the moon for any energy spectrum of incident alpha particles. For the power-law energy spectrum shown in the inset of Fig. 1, the calculated fluxes of alpha particles for several depths in the moon are shown in Fig. 2. Because of the short range of the alpha particles in lunar material (an alpha particle with an energy of 2 Mev/nucleon has a range of only about 10^{-2} g cm⁻²), the flux of low-energy alpha particles decreases rapidly with depth.

The production rate for a particular radionuclide can be obtained by an integration over energy of the product of these calculated differential fluxes and the cross sections for the specific reaction (16). The cross sections used for the 56 Fe(α ,n) 59 Ni reaction were the experimental values up to an incident energy of 3.8 Mev/nucleon (22) and values estimated from nuclear systematics above that energy. The peak cross section was 600 millibarns at 3.5 Mev/ nucleon. The cross sections used for the ⁵⁶Fe(*a*,pn)⁵⁸Co and ⁵⁶Fe(*a*,p2n)⁵⁷Co reactions were the experimental values up to an incident energy of 17 Mev/ nucleon (23). The calculated production rates for these three radionuclides as a function of depth are shown in the inset of Fig. 2.

The production rate for ⁵⁷Co in the top 0.33 g cm⁻² of the surface [corresponding to the sample OP-1 of rock 12002 (14)] is 43 atoms per minute per kilogram of iron [atom min-1 $(kg Fe)^{-1}$]. This calculated value is consistent with the activity of 95 ± 36 disintegrations per minute (dpm) per kilogram of iron measured for ⁵⁷Co in OP-1 (16.8 percent Fe) (14). In the sample, the solar proton contribution was independently estimated (from ⁵⁶Co activities) to be 54 dpm (kg Fe) $^{-1}$, leaving 41 (\pm about 100 percent) for the solar alpha contribution.

The lowest energy product, ⁵⁹Ni, has the highest production rate at the surface, 661 atoms min⁻¹ (kg Fe)⁻¹, and its production rate decreases most rapidly with depth, half the production being in the top 7×10^{-2} g cm⁻² (approximately 0.2 mm) of the surface. The total production of ⁵⁹Ni from iron by alpha particles is 0.019 atom min⁻¹ per square centimeter of surface. From the production rate profile of Fig. 2, the production rate for ⁵⁹Ni in the OP-1 sample (0 to 0.33 g cm⁻²) is 40 atoms min^{-1} (kg Fe)⁻¹. The production rates from other sources are small, the rates from ${}^{58}Ni(n,\gamma){}^{59}Ni$ and ${}^{60}Ni(n,2n){}^{59}Ni$ (secondary neutrons from galactic cosmic rays), and ⁶⁰Ni(p,pn)⁵⁹Ni and ⁵⁹Co(p,n)⁵⁹Ni (solar protons), being estimated as 0.5, 0.1, 1.2, and 1.4 atoms \min^{-1} (kg Fe)⁻¹, respectively, for this sample. Because of the steepness of the ⁵⁹Ni production profile with depth, steady removal (erosion) of surface material can affect the activity of ⁵⁹Ni. The erosion rate for rock 12002 was estimated to be 0.5 mm (150 mg cm⁻²) per 10⁶ years. The present-day

activities of ⁵⁹Ni, calculated from the production profile of Fig. 2 by using erosion rates of 75, 150, and 300 mg cm⁻² per 10⁶ years, are 32, 28, and 23 atoms min⁻¹ (kg Fe)⁻¹. The activity of ⁵⁹Ni expected in OP-1, when the other sources of production and the estimated erosion rate are corrected for, is 25 dpm (kg Fe) $^{-1}$, within a factor of 3 of a preliminary value of 54 ± 18 measured for 59Ni in this layer of rock 12002 (18).

The alpha particle flux inferred by SHRELLDALFF (13) from their Apollo 11 ⁵⁹Ni data was not much better than an order of magnitude estimate because of uncertainties in the data. The activity of ⁵⁹Ni measured by shRellDALFF in soil sample 10084 (12.3 percent Fe) was 3.3 ± 1.0 dpm kg⁻¹, but since the depth of the sample was not known, the alpha particle flux could not be determined from the activity alone. SHRELLDALFF also measured a limit of less than 3 dpm kg⁻¹ for ⁵⁹Ni in the top 12 mm of rock 10017 (15.3 percent Fe). Using both these results, SHRELLDALFF estimated the omnidirectional (4π) flux of alpha particles with energies above 2.5 Mev/nucleon to be approximately 10¹ cm⁻² sec⁻¹ over the mean life of ⁵⁹Ni. The corresponding value of the omnidirectional integral flux for the average spectrum of Fig. 1 is 8 cm $^{-2}$ sec $^{-1}$.

In summary, measurements of radionuclides in the top surfaces of lunar samples can be used to infer the flux of solar alpha particles with energies of a few million electron volts over time intervals of about 1 year (from measurements of ⁵⁸Co and, with less accuracy, ⁵⁷Co) and about 10⁵ years (from measurements of ⁵⁹Ni). The measurements of ⁵⁷Co in lunar samples are consistent with the alpha particle fluxes measured by satellites. The Apollo 11 results and the Apollo 12 results for ⁵⁹Ni in lunar samples indicate that the solar alpha particle flux over the last 10⁵ years has been comparable, within a factor of approximately 4, with that measured by satellites during 1967 to 1969. If the large flare of August 1972 is included, the agreement would tend to be somewhat better (20). These results, together with the result that the average solar proton flux during the last 10⁶ years is similar to the presentday fluxes (14), indicate that the longterm $(10^5$ to 10^6 years) emission of both energetic protons and alpha particles by solar flare processes has not been very different from that observed today. Hence, it can be inferred that the solar surface material that is accelerated by flare processes has had approximately the same composition ratios between hydrogen and helium for approximately 10⁵ years.

L. J. LANZEROTTI

Bell Laboratories,

Murray Hill, New Jersey 07974

R. C. REEDY*, J. R. ARNOLD Department of Chemistry,

University of California, San Diego,

La Jolla 92037

References and Notes

- 1. T. Hirayama, Solar Phys. 19, 384 (1971). 2. A. Hundhausen, Rev. Geophys. Space Phys.
- 8, 729 (1970). 3. S. Biswas and C. E. Fichtel, Space Sci. Rev.
- S. BISWas and C. E. Fichtel, Space Sci. Rev. 4, 709 (1965).
 I. Iben, Jr., Ann. Phys. 54, 164 (1969); R. Davis, Jr., Bull. Amer. Soc. 17, 527 (1972).
 G. M. Nikolsky, Solar Phys. 6, 399 (1969).
 J. Geiss, P. Hirt, H. Leutwyler, *ibid.* 12, 458 (1970).
- (1970).J. Lanzerotti and M. F. Robbins, ibid. 10, 7. I
- 212 (1969). T. P. Armstrong and S. M. Krimigis, J. Geophys. Res. 76, 4230 (1971). 8. T. P.
- L. J. Lanzerotti, Report UAG-8 (World Data Center A, U.S. Department of Commerce, Boulder, Colo., April 1970); NASA Tech. Memo. X-2440 (1972), p. 193; M. Scholer, D. Hovestadt, B. Hausler, Solar Phys. 24, 475 9. L (1972).
- 10. D. Lal and V. S. Venkatavaradan, Earth Planet. Sci. Lett. 3, 299 (1967).
- 11. H. Hasegawa, K. Yamakoshi, M. Noma, T. Maihara, Can. J. Phys. 46, S930 (1968).
- Maihara, Can. J. Phys. 46, S930 (1968).
 12. S. Tanaka, K. Sakamoto, J. Takagi, M. Tsuchimoto, Science 160, 1348 (1968).
 13. SHRELLDALFF, "Proceedings of the Apollo 11 lunar science conference," Geochim. Cosmochim. Acta 2 (Suppl. 1), 1503 (1970).
 14. R. C. Finkel et al., in Proceedings of the Second Lunar Science Conference, A. A. Levinson, Ed. (M.I.T. Press, Cambridge, Mass., 1971), vol. 2, p. 1773.
 15. A. K. Lavrukina and G. K. Ustinova, Nature 232, 463 (1971).
 16. R. C. Beedy and J. R. Arnold, J. Geophys.
- R. C. Reedy and J. R. Arnold, J. Geophys. Res. 77, 537 (1972). 16.
- 17. For example, the shorthand ${}^{56}Fe(\alpha,pn){}^{58}Co$ is used for the nuclear reaction ${}^{56}\text{Fe} + \alpha \rightarrow p +$ $n + {}^{58}$ Co, where α denotes an alpha particle, p a proton, and n a neutron.
- Parioton, and n'a neuron.
 M. Wahlen, personal communication.
 L. J. Lanzerotti, H. P. Lie, G. L. Miller, Inst. Elec. Electron. Eng. Trans. Nucl. Sci. NS-16, 343 (1969).
- 20. The occurrence of the large solar event in August 1972 indicates the dangers of assuming that any one 3-year period is typical. Preliminary examination of the alpha particles (3.4 Mev/nucleon) measured by Bell Labora-tories during this event indicates that the fluxes of particles of this energy (from 4 larger than in the April 1969 event (Fig. 1). L. J. Lanzerotti and C. G. Maclennan, J.
- Geophys. Res., in press. 22. M. Wahlen, thesis, University of Bern, Bern,
- Switzerland (1969). A. Ewart, C. Valentine, M. Blann, 23. Nucl
- *Phys.* **69**, 625 (1965); A. Ewart and M. Blann, *J. Inorg. Nucl. Chem.* **26**, 967 (1965).
- 24. This work was completed while L.J.L. was a This work was completed while L.J.L. was a visiting scientist in the physics department, University of Calgary, Calgary, Alberta, Canada. We thank R. Finkel and M. Wahlen of the University of California, San Diego, for their excellent assistance and comments. The work at the University of California was supported by NASA grant NGL 05-009-148.
- Present address: Group CNC-11, Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544.

10 October 1972; revised 11 December 1972