# Reports

## Secular Variations in the Abundances of Heavy Nuclei in Cosmic Rays

Abstract. The ratios of fluxes of heavy cosmic-ray nuclei  $[(30 \le Z \le 40)/(20 < Z \le 28)]$ , where Z is the atomic number], based on studies of meteorites and lunar samples, show little variation with time, distance in the solar system (1 to 3 astronomical units), or kinetic energy (30 to 2000 megaelectron volts per nucleon). Samples exposed for time intervals of  $10^3$  to  $10^8$  years show that the variations in the flux of  $30 \le Z \le 40$  nuclei relative to that of the iron group nuclei ( $20 < Z \le 28$ ) have remained within the range of ( $1.3 \pm 0.6$ ) ×  $10^{-3}$  during the last billions of years, thus indicating a remarkable similarity in the elemental composition of sources responsible for these nuclei in the cosmic radiation.

Time variations in the intensity and composition of cosmic rays are of importance in an understanding of their origin. Observations of radionuclides produced in meteorites by cosmic rays have indicated that the average intensity of the proton component of the galactic cosmic radiation integrated over the mean life of the radionuclides (1 to 109 years) has remained essentially at the current level, within a factor of 2 (1), during the last  $10^9$  years. The flux of the iron group [very heavy (VH)] nuclei integrated over the last  $12 \times 10^6$  years, as deduced from the St. Severin meteorite, also agrees fairly well with their contemporary flux (2). We now report observations on the variation of the flux of heavier nuclei  $[(30 \le Z \le 40)]$ , where Z is the atomic number] relative to that of the iron group  $(20 < Z \le 28)$  over short differential time intervals in the past. The abundances of very very heavy (VVH) nuclei  $(Z \ge 30)$  in cosmic rays are sensitive indicators of the elemental composition of the sources and hence may be used to identify the nucleosyn-

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Authors of Reports published in Science find that their results receive good attention from an interdisciplinary audience. Most contributors send us excellent papers that meet high scientific standards. We seek to publish papers on a wide range of subjects, but financial limitations restrict the number of Reports published to about 15 per week. Certain fields are overrepresented. In order to achieve better balance of content, the acceptance rate of items dealing with physical science will be greater than average.

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thetic processes operative in the source regions. Since these nuclei originate in local discrete sources in the galaxy, a secular variation in their intensity and composition may also be expected at the earth as the solar system passes through the arm and interarm regions of the galaxy.

With this objective in view, we have studied the relative abundances of VVH  $(30 \le Z \le 40)$  and VH (20 < Z $\leq 28$ ) nuclei in galactic cosmic rays, at various energy intervals, as a function of time and distance from the sun (Fig. 1). Lunar samples and meteorites, exposed at different epochs, provided appropriate samples for this study. Using the fossil track technique of charge identification (4), we have studied interior samples from two lunar rocks (3), samples 12002 and 12020, having effective surface exposure ages of 2.2  $\times$  $10^6$  and  $2.6 \times 10^6$  years, respectively, and grains selected from surface dust and well-stratified lunar cores 12025, 12028, and 15002. Depth profiles of a variety of cosmic-ray effects, for example, solar flare track densities, neutron fluences, and concentrations of cosmogenic rare gases, indicate that the bottom of core 12025, 12028 (a core with two sections), containing 11 layers in all, was first irradiated on the lunar surface about  $300 \times 10^6$  years ago and that of the Apollo 15 drill core, sample 15001 through 15006 (a core with six sections), containing 58 textural units, about  $10^9$  years ago (5, 6). Although these deposition models of lunar soil dif-

Table 1.	Observed f	lux ratios	of VVH	/VH	nuclei in	cosmic	rays.
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Sample	Expo- sure inter- val $(\times 10^6$ years)	Time before present $(\times 10^6$ years)	Energy range (Mev per nucleon)	Track density $\rho_{VH}$ (× 10 <sup>6</sup> cm <sup>-2</sup> )	Track density ratio $\left(\frac{\rho_{VVH}}{\rho_{VH}}\right)$ $(\times 10^{-3})*$	$ \begin{pmatrix} Flux \\ VVH \\ VH \\ (\times 10^{-3})^{\dagger} \end{pmatrix} $
		Apo	ollo rocks			
12002	2.2	0	60-100	6.76	$1.8 \pm 0.2$	1.49
12002	2.2	0	100-300	5.6	$1.4 \pm 0.2$	1.16
12020	2.6	0	~ 20	13	$1.90 \pm 0.2$	1.57
12020	2.6	0	20-50	7-12	$2.7 \pm 0.6$	2.22
12020	2.6	0	100-250	3.4-5.1	$1.49 \pm 0.3$	1.20
12020	2.6	0	800900	6.65	$1.94 \pm 0.2$	1.61
		Apoilo	surface dust	t		
15101.131	69	ò	$\ge 20$	20	$2.18 \pm 0.2$	1.81
15211.49	17	0	$\geq$ 30	34-48	$2.00 \pm 0.2$	1.66
15211.72	23	0	$\geq 30$	16-28	$1.98 \pm 0.4$	1.65
15231.70	35	0	$\geq$ 30	30	$1.67 \pm 0.3$	1.38
,		Apollo	o core layers			
12025.46	45	26	~ 30	11-17	$1.66 \pm 0.4$	1.38
12028.63	≤1	145	30-400	8.1	$0.9 \pm 0.2$	0.75
12028,76	10	146	≥ 30	17-60	$0.89 \pm 0.2$	0.70
12028,193	20	161	$\geq$ 30	8-30	$1.83 \pm 0.9$	1.52
12028,118	30	193	≥ 30	25-70	$1.54 \pm 0.4$	1.27
12028,133	60	223	≥ 30	26-27	$1.15 \pm 0.3$	0.96
15002,111	23	756	$\geq 30$	20-30	$1.04 \pm 0.5$	0.91
15002,79	17	814	$\geq 20$	15-30	$1.40 \pm 0.5$	1.16
15002,43	18	913	$\geq 30$	56	$1.41 \pm 0.3$	1.16
		М	eteorites			
Patwar	60	0	100	14-17	$1.7 \pm 0.5$	1.36
Patwar	60	0	1500	1.3	$1.4 \pm 0.2$	1.34
Itzwisis	50	0	2000	1.1	$1.2 \pm 0.2$	1.0
esyanoe (TRG)	0.01-0.1	<b>~</b> 4500	≥20	21-47	$1.3 \pm 0.2$	1.4

\* The errors also include the range of scatter for cases where more than one crystal from a particular sample was studied.  $\dagger$  See (10). fer in details, all of them are in support of the view that most lunar core layers build up sequentially with time as they become covered by fresh ejecta soil. A consequence of the steeply falling track production rate due to galactic nuclei,  $\dot{\rho}(X)$  as a function of depth X,

#### $\check{\rho}(X) \propto (7.5 + 0.53X)^{-6.15}$

is the fact that the contribution to the tracks recorded becomes increasingly smaller as the shielding depth increases (3). In a simplified case, it may be assumed that most of the tracks were recorded at shallow depths when the particular layer was close to the surface. A time sequence so determined for these two core strata has been used here (5). Lunar samples thus provided grains irradiated for brief spans of time up to  $10^9$  years in the past at 1 A.U.

In order to obtain similar information about the region farther from the sun, we have also examined two stony iron meteorites, Patwar (7) and Itzwisis. The oldest cosmic-ray records that we could find are those in trackrich grains (TRG) of the Pesyanoe meteorite, which were irradiated for  $10^3$  to  $10^4$  years in the formative stages of the solar system, possibly  $4.5 \times 10^9$  years ago (8). Table 1 lists the relevant details for all the samples.

In this investigation we used experimental techniques similar to those employed earlier (4, 7). Olivine grains were suitably developed for revelation of tracks, and VH and VVH tracks, as identified (9) by their surface track lengths (1 to 15  $\mu$ m and > 20  $\mu$ m, respectively), were counted (4, 10). The records of galactic cosmic rays could be easily distinguished from those of solar nuclei on the basis of the observed track density gradients. Solar nuclei, which dominate below a kinetic energy of 20 Mev per nucleon, tend to give steep track density gradients (characteristically with a slope of -1) in the near-surface regions ( $< 100 \ \mu m$ ) of the exposed grains (3). These were excluded from the present study, and so the results presented in Table 1 refer to the galactic cosmic-ray nuclei only. Most samples show values of the VVH/ VH flux ratio ranging from  $0.7 \times 10^{-3}$ to  $1.8 \times 10^{-3}$ . We estimate the overall



Time before present (× 10<sup>6</sup> years)

Fig. 1. The flux ratio of  $30 \le Z \le 40$  relative to  $20 < Z \le 28$  nuclei in cosmic rays for different epochs in the past. The bar on each point represents the time span over which the sample was irradiated with galactic cosmic rays. The typical error in each ratio is  $\pm 30$  percent. The irradiation time for lunar dust is model-dependent and could have errors of  $\pm 20$  percent, increasing cumulatively in the past (3). The open symbols represent data at high energy (E = 0.2 to 2 Gev per nucleon), and the solid symbols represent an energy range of 0.02 to 0.4 Gev per nucleon. The circled triangle shows the marginal increase around 0.05 Gev per nucleon observed in rock 12020.

errors to be  $\pm$  30 percent. It is possible that the effective exposure time of individual dust grains is much smaller than the surface exposure age of the core layers and therefore that the small variations in the VVH/VH ratio from crystal to crystal (Table 1) are genuine. However, since these variations are not significantly larger than the expected errors, it appears that the VVH/VH ratio has remained similar at (1.3  $\pm$  $(0.6) \times 10^{-3}$  over the whole time period studied (11). Systematic measurements as a function of energy (E), made in lunar rock 12020, also show no variation in the VVH/VH ratio except a possible marginal increase, up to  $2.2 \times$  $10^{-3}$ , in a narrow energy interval around 50 Mev per nucleon. As a consequence of energy loss arising from solar wind interaction and adiabatic deceleration, the nuclei with  $E \approx 100$ Mev per nucleon at 1 A.U. originate at much higher energies in the interstellar space. The spectra at  $E \approx 100$  Mev per nucleon in the interplanetary space, therefore, do not represent the interstellar spectra (12). Furthermore, solar flare nuclei at low energies (< 20 Mev per nucleon) have a VVH/VH ratio of  $13.4 \times 10^{-3}$  (4) compared to the photospheric ratio of  $1.2 \times 10^{-3}$ , indicating a charge-dependent acceleration process operative at the sun. We therefore confine the discussion that follows to energies > 100 Mev per nucleon.

The effects of solar modulation should be similar for VH and VVH nuclei because of similar charge-tomass ratios. Thus, for all energies from about 100 Mev per nucleon to 2 Gev per nucleon, where the VVH/VH ratio is constant, it may be concluded that the energy spectra of VH and VVH nuclei are parallel in the interstellar space and may reflect the source composition.

The exposure time over which the records have been averaged is comparable to or longer than the average lifetime (13) of the heavy nuclei in the galaxy (~  $10^6$  years). At any time, therefore, a mixture of cosmic rays from a large number of sources are present in the interstellar space, and it is difficult to observe cosmic rays from a single source. Samples exposed during different epochs, however, represent cosmic-ray records from different sets of cosmic-ray sources. Since the VVH/VH ratio at E > 100 Mev per nucleon may reflect the source composition, it is instructive to discuss various nucleosynthetic processes responsible for the production of these nuclei. The abundances of various nuclei up to the iron group show a good agreement with their production estimates in explosive nucleosynthesis (14). The heavier nuclei can, however, be produced in a variety of processes, s, r, or p processes, from the seed nuclei of the iron group (15). The relative abundance of VVH/VH nuclei in some s process models, for example, is higher by a factor of 4 as compared to that in r process synthesis. Under suitable conditions, nuclei beyond the iron group  $(Z \approx 30)$  could also be formed in explosive nucleosynthesis or other freeze-out processes that follow nucleosynthesis (14). The VVH/VH ratio therefore is a good indicator of the processes dominating the production of VVH nuclei since for the various processes there is a wide variation in the expected VVH/VH ratio. The absence of a wide variation in the VVH/VH ratio in cosmic rays with time therefore suggests that these nuclei are not produced in a variety of processes; rather, the nuclei are probably produced in a single process that yields a VVH/VH ratio of  $\approx 1.3 \times 10^{-3}$ . It seems remarkable to us that this ratio has remained the same in cosmic rays in the past. This observation indicates that most of the heavy cosmic rays originate in sources of similar heavy element composition and in similar nucleosynthetic processes.

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  16. We thank A. Padhye of the Tata Institute of Fundamental Research (TIFR) for carrying out some of the measurements. We are grateful to D. Lal, A. S. Tamhane (TIFR), M. N. Rao, and J. N. Goswami for discus-sions; to E. L. Krinov for Pesyanoe samples; and to the National Aeronautics and Snace and to the National Aeronautics and Space Administration for the lunar samples.
- 20 February 1974; revised 16 April 1974

### Kerogen Recycling in the Ross Sea, Antarctica

Abstract. Analyses of the stable isotopes of the organic carbon and microscopic examination of the sediment particles suggest that up to 90 percent of the organic matter in Ross Sea sediments is derived from the igneous and ancient metamorphic and sedimentary rocks that are being glacially eroded on the Antarctic continent and transported seaward.

The nondescript, insoluble, organic material found in most sedimentary rocks is defined as kerogen. Kerogen is the major reservoir of organic carbon on the earth's surface, containing over three orders of magnitude more carbon than all the other organic carbon reservoirs combined. The mode of purification used for kerogen testifies to the inertness of the material: the rock is dissolved away from the carbonaceous matter through the use of hydrochloric and hydrofluoric acids (1). This chemical and presumably biological inertness suggests that much of the kerogen in ancient sediments that are being weathered and transported to the sea may be redeposited with relatively little alteration. If this is true, then the kerogen and the various elemental and mineralogical constituents found in sediments may represent several weathering and redeposition cycles. For each cycle a new portion of kerogen formed by the maturation of contemporaneous organic matter may be added to the amount already formed in previous cycles. Because of the complex nature of kerogen, it has been difficult to determine whether an incremental accumulation with time has occurred. For Holocene sediments found in temperate and tropical environments the problem has been the masking of the recycling process by the tremendous mass of recently living land-derived organic matter. However, this is not a problem in polar regions where terrestrial ecosystems are practically nonexistent. We report here on a study of the amounts and isotopic

composition of the organic carbon in sediments of the Ross Sea, Antarctica, which shows that much of the sedimentary organic carbon is derived from the rocks being eroded by glaciers on the Antarctic continent.

Sediment cores were collected along the edge of the Ross Ice Shelf during cruise 51 of R.V. Eltanin in February 1971. At that time, in the middle of the austral summer, the shelf extended to about 78°S. Surrounding the Ross Sea to the southwest extends the Transantarctic Mountain chain composed of a complex mixture of sedimentary, metamorphic, and igneous rocks of Precambrian to Late Cenozoic age. Glaciers spread from many of the valleys of this mountain chain down into the Ross Sea and give rise to the Ross Ice Shelf, the largest feature of its type on the earth's surface.

The primary core used in this study, core 51-18, was taken about 160 km northeast of McMurdo Sound at the edge of the ice shelf. We examined the size fraction greater than 74  $\mu$ m under a microscope to estimate the relative amounts of the various types of organic and nonorganic derived particles. The widths of the bars in Fig. 1A indicate the relative amounts within one constituent category. These widths are not comparable from category to category. The amounts and stable carbon isotope compositions of the total organic carbon (~95 percent kerogen) were determined (2) on the size fractions smaller than and larger than 250  $\mu$ m. These are given in Fig. 1B and Table 1.