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Effects of Cosmic Rays on Meteorites

Meteorites are more suitable than terrestrial targets for studying the fossil record of cosmic radiation.

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Cosmic rays and meteorites are two classes of material bodies that reach the earth from outer space. The study of their interactions has helped to increase our understanding of both.

A great many things are known about the cosmic radiation. In the region of space near the earth it consists mainly of high-energy protons, with an important component of alpha particles, and a smaller fraction of heavier nuclei. Typical energies are in the region of 10^9 electron volts (Gev) per nucleon, although particles of far higher energy occur. Between the discovery of the neutron in the early 1930's and the advent of very-high-energy machines a few years ago, most of the information available on fundamental particles came from cosmic-ray studies. Still, despite abundant data on composition, energy, and direction of motion, the place and mode of origin of cosmic rays is only partially understood.

Cosmic-ray bombardment affects terrestrial materials. The best-known example of such an effect is the production of carbon-14 by the $N^{14}(n,p)C^{14}$ reaction. The half-life of C^{14}, 5600 years, and the biological importance of carbon, make this nuclide most important for chronological studies. Carbon-14 is not the only radioactive nuclide produced by cosmic rays; other short- and long-lived nuclides are formed by interaction between the high-energy particles and the nitrogen, oxygen, and argon of the air. Cosmic-ray-produced nuclides occur in very small amounts in the solid surface materials of the earth because of the absorption by the mass of air (1 kg/cm^2) above us. However, they can be detected by modern methods of low-level counting. Extensive reviews covering recent advances in this field are available (*1*).

The reactions which produce the observed radioactive species on earth occur mainly in the stratosphere and upper troposphere. What happens to each nuclide after formation depends on its chemical and physical properties. If it is long-lived or stable, it will be transported away from the point of origin, and usually it undergoes various chemical reactions. In general, these changes are only partly understood. As a result, it is difficult to decipher the fossil record of cosmic radiation which the data undoubtedly contain. The one exception is, again, C^{14}. The general success of the C^{14} dating method is very good evidence for the approximate constancy of the intensity of the cosmic ray striking the atmosphere over the last few half-lives of this nuclide. Apparent small variations in the C^{14} concentration may be due to past changes in intensity. However, other geochemical or geophysical phenomena (such as changes in the rate of CO_2 transport across the air-sea interface and magnetic field variations) may be involved.

Unlike the earth's atmosphere, the meteorites are physically and chemically stable targets. While their history presents problems of its own, they are much more suitable than terrestrial targets for an extended study of the fossil record of cosmic radiation. Until samples of the surface of the moon and of other bodies without atmosphere (including, for some purposes, artificial targets) become available, meteorites will be the most suitable bodies for such studies.

Course of Radiation in a Meteorite

Let us follow the radiation into a meteorite. High-energy protons and other charged nuclei enter its surface from every direction. After passing through matter of density about 100 grams per square centimeter, each particle undergoes an interaction, giving rise to new active particles of various energies and types. These secondary particles undergo further interactions, and this cascade process continues until the energy is dissipated. Among the nuclear particles in which we are interested are high-energy protons of primary and secondary origin, neutrons of all energies, and, in the case of condensed bodies such as meteorites, important numbers of pi mesons also. The energy dependence of the primary flux is shown in Fig. 1 (*2*). Above 1 Gev it is well represented by a law of the form $(1 + E)^{-2.5}$, where E is the energy in Gev. The mean kinetic energy per nucleon is in the neighborhood of 4 Gev. In space, the total flux over all angles amounts to about five nucleons per square centimeter per second. Inside the meteorite body the intensity of the high-energy particles decreases with depth in

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accordance with a mean absorption thickness (for a collimated beam) of about 150 g/cm^2. The low-energy secondary flux increases in the first few centimeters below the surface of the body. After that it passes through a maximum and then decreases, somewhat more slowly than the primary intensity decreases. The total particle flux increases rapidly at first, as the low-energy particles become much more numerous than the remaining high-energy ones. Below a depth of 150 g/cm^2 or so, the total flux decreases, the energy spectrum changing (steepening) only slowly.

A special effect arises in quite large bodies. Here an appreciable flux of low-energy, or thermal, neutrons makes an appearance. There are very few experimental data showing the effects of thermal neutrons in meteorites (*3*). Products with which we are concerned here are produced by spallation—a generic term for nuclear reactions at energies above about 10 Mev. In these reactions the emission of one or more nucleons from the target nucleus is to be expected, and the products will range in mass from that of the target nucleus down to single neutrons or protons.

There are a number of possible sources of time variation in the intensity of the cosmic radiation reaching us on earth. Even in the last few decades, a cyclic change of the flux has been detected, with a period corresponding to the 11-year solar cycle (*2*). The lowest-energy portion of the primary cosmic radiation is much affected by the sun. There is a general decrease in this radiation at times of high solar activity, punctuated by occasional high-intensity bursts from the sun. The sources of primary cosmic radiation in the Gev region are generally believed to be in the galaxy, far from the solar system. Very energetic particles, carrying 10^{20} electron volts and even more, may be of extragalactic origin. However, the total flux appears to be mainly galactic.

It must be kept in mind that primary cosmic radiation in the energy region of 1 to 10 Gev contains most of the flux and is responsible for most of the transmutations observed. The data give information mainly about this energy region.

A number of sources for cosmic rays within the galaxy have been suggested (*4*). A supernova occurs in our galaxy every few centuries, and these catastrophes may well give rise to most of the particles observed. Other suggested sources—much more numerous, although less dramatic—are the contact binary stars and the flare stars among the red dwarfs. Whatever their origin, it is thought that the particles, after acceleration, are trapped in the magnetic fields found in the spiral-arm structure of the galaxy. The intensity of the radiation in interplanetary space will be affected by (i) variations in source intensity; (ii) variations in the general magnetic field in the region of the galaxy through which the sun is passing; and (iii) variations, like those observed during the sun-spot cycle, in the intensity and the extension into space of the sun's magnetic field. It seems highly probable that over a span of many aeons (1 AE = 10^9 years) the population of sources and the magnetic field of the galaxy in the region of the sun have changed considerably. Have there been important variations over shorter periods? The record of cosmic radiation in meteorites seems to offer the best hope of attacking this question. However, this requires that we know something about the history of the meteorites themselves (*5*).

Table 1. Measured and estimated production rates of nuclides in iron meteorites. The production cross section is given for the center of a 20-cm sphere (93 percent Fe, 7 percent Ni) from 3-Gev proton bombardment data. The estimated relative production rate in iron (6.5 percent Ni) is given for a depth of 100 g/cm^2, radius 200 g/cm^2, relative to $Cl^{36} = 1$. Measurements of the Aroos meteorite are corrected to the time of fall; dpm, disintegrations per minute.

Nuclide	Production cross section[a] (mbarn)	Estimated relative production rate[b]	Aroos[c] (dpm/kg)
H^3		12	
Be^7	6.5	0.69	
Be^{10}		.31	4.1 ± 0.4
C^{14} [d]		.13	1.8 ± 0.3[d]
Na^{22}	1.3	.11	2.1 ± 0.3
Al^{26}		.10	3.6 ± 0.4
P^{32}	5	.41	
Si^{32}		.05	0.8 ± 0.3
Cl^{36}		1	16 ± 1.6
Ar^{37}		0.65	20[e]; 15[f]
Ar^{39}		.9	16 ± 1[e]
Ca^{45}		.28	5 ± 1
Sc^{44m}	9		
Sc^{46}	18	1.5	30 ± 3
Sc^{47}	11		
Ti^{44}		0.38	4.4 ± 0.4
V^{48}	46	7.3	90 ± 45
V^{49}	107	9.6	164 ± 16
Cr^{51}	120	19	260 ± 120
Mn^{52}		6.6	
Mn^{53}		33	515 ± 52
Mn^{54}	240	38	470 ± 47
Fe^{55}	550	220	1600 ± 600
Fe^{60}		0.1	
Co^{56}	20	4.5 }	120 ± 34
Co^{58}	77	17 }	
Co^{57}	55	5.5	89 ± 9
Co^{60}		0.6	17 ± 2
Ni^{59}		1.4	60 ± 15

[a] See *26*. [b] See *24*. [c] See *11*. [d] See *37*. [e] See *30*. [f] See *36*.

History of Meteorites

The meteorites reach the earth from interplanetary space, and it seems to be firmly established that they originate inside the solar system. The one meteorite whose orbit has been well determined, Pribram (Luhy) (*6*), indeed had an orbit passing out into the asteroid belt. However, major perturbations caused by passages close to the inner planets profoundly alter the original orbits of these bodies (*7*). The moon, the planets Mars and Mercury, and the comets are all considered possible sources of meteoritic bodies. Because of the very large size and mass of Jupiter, bodies whose orbits extend out to its distance from the sun or beyond are likely to be captured by it or ejected from the solar system. On the basis of this and other arguments, it seems highly probable that meteorites have spent their careers in space closer than about 5 astronomical units to the sun.

The great majority of meteorites observed to fall are stones. In fact, about 80 percent of all observed "falls" are stones of one class, called chondrites. Chondrites contain several silicate minerals, chiefly pyroxene and olivine; iron-nickel metal (of the order of 10 percent); and troilite, ferrous sulfide (about 5 percent by weight). The commonest elements (targets) are therefore oxygen, silicon, magnesium, and iron, with important amounts of sulfur, aluminum, calcium, and sodium. Chondrites get their name from characteristic spherical inclusions called chondrules. A variety of different objects are lumped under the generic name "achondrite." Some are similar in chemical composition to the chondrites. Another group is much richer in calcium. About 10 percent of the observed falls are iron meteorites, some of which have been very large. Iron meteorites are composed of metallic iron, containing usually 5 to 10 percent of nickel. Inclusions of ferrous sulfide, iron-nickel phosphide, and graphite are usually present. Many of the common terrestrial elements are extremely rare in iron meteorites. These include not only obviously incompatible elements, such as potassium and calcium, but also some, such as manganese, silicon, titanium, and vanadium, which occur commonly in artificial iron alloys. An intermediate group, the stony irons, contain comparable quantities of metal and stone. A group of these, called pallasites, contain large and easily separated crystals of the two phases (metal and olivine).

Secular Equilibrium

Before discussing the radioactive products of cosmic-ray bombardment, let us review the notion of secular equilibrium. If a radioactive substance is produced at a steady rate for a time that is long compared to its half-life, the number of nuclei decaying will come to equal the number being formed, so that no further changes of concentration occur. The observation that the rate of decay of the radioactive species in a meteorite immediately after fall is equal, within the limits of experimental error, to the present rate of production will lead us to conclude, conversely, that the cosmic-ray intensity has been constant over periods of the order of the mean life of this species. This converse is not rigorous, since a coincidence of production and decay rates might be an accidental consequence of wide temporal variations in rate of production. However, the probability that the cosmic-ray intensity has been constant is high, especially when the same result is observed for many radioactive species of different half-lives.

How can one calculate the absolute production rate for each species at each point in the target? This rate is dependent on the absolute intensity and the energy spectrum of the bombarding flux. This in turn is a function of the depth of the sample in a particular meteorite, and of the size and shape of the meteorite body in space. It is not easy to deduce these values quantitatively, but it is possible to make rough estimates. For instance, Na^{22} and Be^{10} are produced only by high-energy particles. The primary flux of five nucleons per square centimeter per second may be reduced by 75 percent at an effective depth of 25 centimeters in iron, or 200 g/cm^2. The cross sections for these species are about 3 and 5 millibarns, respectively (1 mbarn equals 10^{-27} cm^2), at 4 Gev, the mean energy of the primary beam. The cross section is rather independent of energy in the Gev region. Therefore, the production rate of Na^{22}, for example, is

$$5 \cdot (1/4) \cdot 3 \cdot 10^{-27} \cdot 6 \cdot 10^{23}(1000/56) \cdot 60 = 2.4 \text{ atom/kg min}$$

This number is in rough agreement with the observed activity of these species in the freshly fallen meteorite Aroos (Table 1). On the other hand, Mn^{54} is a typical "low-energy product." Its rate of production is greatly increased by the large number of secondary neutrons produced in the iron block. Above about 50 Mev the cross section is in the vicinity of 30 millibarns, nearly 10 times that for Na^{22} production. But the observed activity is more than 100 times as high (Table 1). This indicates that more flux—about one order of magnitude more—is available for production of Mn^{54} at moderate depths, a result in agreement with what is known about the development of cascades in solid bodies. Between the two extremes there are many nuclides whose energy thresholds for spallation are between 0.1 and 1 Gev. Unless the production cross section is especially low, we expect the concentrations of these nuclides in iron meteorites to lie between those of Na^{22} and Mn^{54}. In a stone meteorite it is necessary to take into account the complex chemistry of the target. For example, Na^{22} in this target is produced mainly from magnesium and silicon. Here it is a typical "low-energy product," like Mn^{54}.

Time Variations

Some conclusions about time variations of the cosmic radiation can be reached by making simple comparisons among pairs and groups of products, without a detailed laboratory or theoretical study of relative production rates. In each meteorite specimen we can compare, for example, (i) short-lived and long-lived nuclides produced in very similar nuclear reactions; (ii) radioactive and stable products; and (iii) products of widely different mass numbers. If the excitation function (cross section as a function of the energy of the bombarding particle) is closely similar in shape for two species, the relative production of the two is given by the ratio of the cross sections at any energy, independent of the detailed nature of the bombarding flux. Examples are given in Table 2. The half-lives cover the range from 2 weeks to millions of years, and the differences in excitation functions (the energy dependence of cross section) in iron meteorite targets are small. As will be shown in other tables the ratios, as actually observed in iron meteorites, seem to be essentially constant from one sample to another, and they are in agreement with the production rates deduced from observed (or sometimes estimated) cross-section data. These findings indicate that the intensity of the cosmic radiation has, on the average, been preserved within an uncertainty of the order of a factor of 2.

Table 2. Short- and long-lived species produced in similar nuclear reactions in iron meteorites.

Nuclide	Half-life	Radiation
Mn^{54}	300 days	Gamma, x-ray
Mn^{53}	$\sim 2 \times 10^6$ yr	X-ray
V^{49}	330 days	X-ray
V^{48}	16 days	Beta (+)
Ti^{44}	~ 200 yr	Active daughter, beta (+)
Sc^{46}	84 days	Beta (−)
Ar^{39}	270 yr	Beta (−)
Ar^{37}	35 days	Electron capture
Cl^{36}	3×10^5 yr	Beta (−)
Al^{26}	7×10^5 yr	Beta (+), gamma
Na^{22}	2.6 yr	Beta (+), gamma
C^{14}	5.6×10^3 yr	Beta (−)
Be^{10}	2.5×10^6 yr	Beta (−)

If the cosmic-ray intensity has been constant, stable products will have accumulated at a steady rate. If relative cross-section data are available, we can calculate a "bombardment age" by considering a suitable pair of nuclides, one radioactive and one stable. An ideal case is Cl^{36} – Ar^{36}. About 80 percent of all the Ar^{36} atoms formed in an iron meteorite result from the decay of radioactive Cl^{36} (*8*). By correcting for the independently produced Ar^{36}, we can arrive directly at a "current" production rate for Ar^{36}. The ratio of the number of atoms now present to the production rate gives us the age. In Tables 3 to 6 (*9*) we present the observed data on several iron and stone meteorites, covering a wide range of radioactive and stable, volatile and nonvolatile, nuclides.

Meteorite Samples

This field of study could not exist were it not for the availability of a wide variety of meteorite samples. From

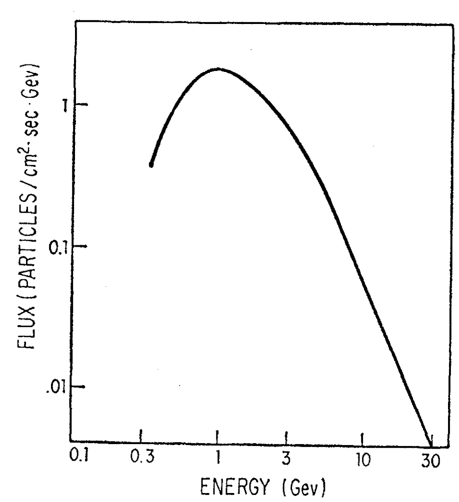

Fig. 1. Differential flux of primary nucleons near the earth (*24*). [From data of MacDonald and Webber (*2*)]

Table 3. Long-lived radioactivities produced by cosmic rays in iron meteorites.

Meteorite	Location	Date of fall[b]	Weight at recovery (kg)	Sample No.	Radioactivity (disintegrations/min kg[a])										Ar^{36} [c] (10^{-8} cm^3/g)
					Be^{10} [2.5×10^6]	Al^{26} [7.4×10^5]	Cl^{36} [3.1×10^5]	Ti^{44} [~200]	Mn^{53} [$\sim 2 \times 10^6$]	Ni^{59} [8×10^4]	Co^{60} [5.26]	H^3 [12.3]	C^{14} [5600]	Ar^{39} [325]	
Aroos (syn., Jardymlynsky)	Azerbaijan, U.S.S.R.	1959	150		4.1±0.4	3.6±0.4	16±1.6	4.4±0.4	515±52	60±15	17±2	35±7[d]	1.8±0.3[e]	16±2[d] 16±0.9[f] 17.2±0.5[g]	28
Deep Springs (nickel-rich ataxite)	Rockingham County, N.C.	(Find)	11.5				6.0±0.6		570±60	0±34					58
Mt. Ayliff	Cape Province, South Africa	(Find)	13.6				17±2			40±20					28
Grant	New Mexico	(Find)	500		4.0±0.3	3.6±0.2	12.4±1.2[h]		360±40	56±14				0.02±0.09[i]	18
Williamstown	Grant County, Ky.	(Find)	31		3.5±0.4	2.9±0.4	3.8±0.7		360±40	0±14					17
Clark County	Kentucky	(Find)	11				4±1		320±35					.9±1.3[j]	46
Treysa	Hesse, Germany	1916	63				22±2	3.0±0.7	270±30	60±14		80±12[i] 100±30[i]		20±0.4[g,j] 13±0.6[i]	21
Admire (metal phase, pallasite)	Lyons County, Kan.	(Find)	>50		1.7±0.3	1.5±0.5	7.4±0.9		200±20	300±30					2.5
Odessa	Ector County, Tex.	(Find)	>1,000	(I) (II)	2.1±0.1 1.0±0.7	1.1±0.1	5.1±0.5 6.5±1.3		240±24 200±25	70±25	(<10)[k]		0.29±0.11[e]	0±0.14[j]	(I)1.6 (II)3.5
Sikhot-Alin (syn., Ussuri)	Eastern Siberia	1947	80,000		2.4±0.3		8.8±0.9	1.8±0.4	250±25	45±17	95±10[l]	0±1[i,m]	1.7±0.4[e]	7.1±0.2[j] 4.5±0.3[i]	6.4
Carbo	Sonora, Mex.	(Find)	454		1.6±0.3		5.5±0.6	0.3±0.7	280±50	74±16	(0±2)[k]		0.71±0.12[e]	0±0.14[g]	12.6
Canyon Diablo	Coconino County, Ariz.	(Find)	>30,000		0.8±0.2	0.8±0.3	1.1±0.2		120±14	60±15	(<10)[k]			0±0.15[i] 0±0.7[j]	
Brenham (metal phase, pallasiderite)	Kiowa County, Kan.	(Find)	4,320		.2±0.4	.1±0.5	0.1±0.2		<15	<20					0.0

[a] Half-lives (in years) are given in brackets. [b] "Find" indicates not an observed fall. [c] See *38*. [d] See *30*. [e] See *37*. [f] See *36*. [g] See *39*. [h] See *40*. [i] See *41*. [j] See *10, 23*. [k] See *14*. [l] At time of fall. [m] See *42*.

most of the great collections—particularly those of the Academy of Sciences of the U.S.S.R., of the Smithsonian Institution, of the American Museum of Natural History in New York, of the Harvard University Museum, and of the Nininger Collection at Arizona State University—samples have been generously made available to qualified investigators. Private collectors have also been most helpful. Freshly fallen meteorites are especially important, because of the possibility of measuring many short-lived radioactive species. News of new falls spreads rapidly across national boundaries. Samples of the Aroos, Bruderheim, and Ehole meteorites have been supplied from the countries in which they fell—the U.S.S.R., Canada, and South-West Africa. R. E. Folinsbee of the University of Alberta has collected and distributed samples of two freshly fallen meteorites, Bruderheim and (in the spring of 1963) Peace River. The utility of this international cooperation is obvious. The meteorite Harleton, recovered near Marshall, Texas, reached our laboratories in record time, arriving about 10 days after recovery. In this sample it was easily possible to measure P^{33}, which has a half-life of 14 days.

Measurement

With activities in the range of several disintegrations per minute per kilogram, the use of massive samples and extremely sensitive counting techniques is a necessity. Snedlovsky, Honda and Arnold have processed as much as 3.5 kilograms of the Odessa iron meteorite and 1 kilogram of the Harleton stone. Even so, in many cases the counting rate for the sample is less than 0.1 count per minute. Low-level techniques require heavy iron shields, anticoincidence and coincidence counting methods, and, where possible, pulse-height discrimination. In the case of some high-yield nuclides, a 50-gram sample may be sufficient. For the measurement of stable nuclides, a much smaller sample, 1 to 10 grams, is usually enough, because of the high sensitivity of the mass spectrometer. Neutron activation analyses have also been used in He^3, Sc^{45}, and Ca^{46} determinations, and this technique may be useful in making other measurements, even for radioactive nuclides such as Mn^{53}.

It is often desirable to measure the production separately in different

Table 4. Cosmic-ray-produced radioactive nuclides in stone meteorites.

Nuclide	Half-life	Radioactivity (disintegrations/min kg)				
		Bruderheim[a] (Alberta, Canada)	Harleton[b] (Texas)	Ehole[c] (Angola, Africa)	Achilles[d] (Kansas)	Admire (stone)[e] (Kansas)
Be^{10}	2.5×10^6 yr	19 ± 2	21 ± 2	19 ± 2	19 ± 2	14 ± 2
Na^{22}	2.6 yr	90 ± 10	64 ± 7	84 ± 17		
Al^{26}	7.4×10^5 yr	60 ± 6	45 ± 5	70 ± 7	50 ± 5	43 ± 4
P^{32}	14.3 days		14 ± 2			
Cl^{36}	3.1×10^5 yr	7.5 ± 0.8	7.0 ± 0.7	7.8 ± 1.0	6.0 ± 0.6	
Sc^{46}	74 days	6.2 ± 0.6	5.4 ± 0.7			
Ti^{44}	~200 yr	2.0 ± 0.2	1.4 ± 0.2			
V^{48}	16 days	34 ± 7	17 ± 2			
V^{49}	330 days	34 ± 2	20 ± 6			
Cr^{51}	28 days	110 ± 27	60 ± 20			
Mn^{53}	$\sim 2 \times 10^6$ yr	85 ± 17	44 ± 8	110 ± 20	60 ± 12	
Mn^{54}	300 days	100 ± 13	38 ± 5	90 ± 20		
Fe^{55}	2.6 yr	340 ± 80	≤180	500 ± 100		
$Co^{56} + Co^{58}$	~84 days	14 ± 4	4 ± 1			
Co^{57}	240 days	11 ± 1	6.5 ± 0.7	14 ± 6		
Co^{60}	5.3 yr	9 ± 1	1.5 ± 0.5	4.8 ± 1.2		
Ni^{59}	8×10^4 yr	12 ± 3	6 ± 6			
H^3	12.3 yr	260 ± 30[f]	{310 ± 20[g] {265 ± 15[h]	295 ± 10[g] 302 ± 10[h]		
C^{14}	5600 yr	{56 ± 3.0[i] {63 ± 5[j]	38 ± 2.4[j] 57 ± 5[i]		56 ± 7.6[j]	17 ± 1.5[j]
Ar^{39}	270 yr	{10.5 ± 1.0[f] {11.5 ± 0.3[h]	9.1 ± 0.4[h] 7.5 ± 0.5[g]	7.8 ± 0.3[g] 8.2 ± 0.3[h]		
Ar^{37}/Ar^{39}		{1.33 ± 0.13[h] {2.2 ± 0.4[g]	1.02 ± 0.13[h]	1.65 ± 0.29[h] 2.1 ± 0.3[g]		

[a] Date of fall, 1960; weight at recovery, 300 kg; iron content, 22 percent. [b] Date of fall, 1961; weight at recovery, 8 kg; iron content 22 percent. [c] Date of fall, 1961; weight at recovery, 2.4 kg; iron content ~ 30 percent. [d] Find; weight at recovery, 16 kg; iron content, 22 percent. [e] Find; weight at recovery, > 50 kg; iron content of stone phase, 8 percent. [f] See *43*. [g] See *42*. [h] See *36*. [i] See *44*. [j] See *37*.

mineral phases. A rough separation of the magnetic and nonmagnetic stone phases is quite easy. Still, the metal fraction separated from a chondrite by this method may contain a small percentage of silicates after separation. In the case of pallasites, a very clean separation is possible. A "wet" method is also useful. For example, the olivine phase in a chondrite can be dissolved in cold strong acid much more easily than the pyroxene phase can be. The metal phase can also be dissolved with $CuCl_2$ or $Hg(NO_3)_2$ solution, other phases being left behind (*10*). The real difficulties begin with the chemistry. Inconveniently large samples must be dissolved under very clean conditions, and chemical procedures must be designed for separating more than 14 elements from one sample (*11, 12*).

The chemical processing must be carried out quickly because of the decay of the short-lived species. In a number of cases the chemical procedures themselves have had to be devised in the few days between the first notice of the meteorite's fall and the arrival of the sample in the laboratory. After separation of groups and of individual elements, each counting sample must be "recycled to constant activity." That is, the element must be repurified repeatedly until the ratio of radioactivity to mass becomes constant. This is the only sure means of determining the chemical identity of the radioactive source. It is best to carry out several different chemical procedures, in order to remove unknown impurities. For the nonvolatile elements, solvent extraction, ion exchange, the use of complexing agents, electrolysis, and co-precipitation are all useful.

In some cases it is possible to measure the radioactivity of particular nuclides in a nondestructive way (*13, 14*). The gamma-ray-emitting species Mn^{54}, Al^{26}, Co^{60}, and Na^{22} can be measured in 1-kilogram samples by means of a large sodium iodide crystal and a multichannel analyzer. It is possible to make many measurements of these species, at least in meteorites where their concentration exceeds about 10 disintegrations per minute per kilogram. Extraction, purification, and measurement of the volatile rare gases require still another set of techniques. Many nonvolatile stable nuclides have been measured in iron meteorites, because of the fortunate circumstance that they are nearly free of many common elements. On the other hand, no such nuclides have been measured in stones. Even in the iron phase of stones, measurement of such nuclides is very difficult because of the inevitable silicate contamination. The avoidance of laboratory contamination is particularly difficult with such elements as potassium and manganese, which are commoner in laboratory materials than in the samples. Finally, as in other critical work, a rigid discipline of blanks and standards must be maintained, along with repeated interlaboratory comparisons. Each laboratory maintains a "library" of solutions and fractions from each meteorite processed. Frequently it is possible to return to these solutions when new ideas occur.

The data of Tables 3 through 6 present, to the experienced eye, a beautiful and orderly pattern. Apart from a few anomalies (some of them doubtless caused by experimental errors) and

Table 5. Cosmic-ray-produced stable nuclides and potassium-40 in some iron meteorites.

Meteorite	Date of fall[a]	Weight at recovery (kg)	Amt of nuclide (10^{-9} g/g)[b]					Amt of nuclide (10^{-8} cm^3/g)[c,d]			
			V^{50}	Sc^{45} [e]	Ca^{43}	Ca^{46}	K^{40}	Ar^{38}	Ne^{21}	He^3	k'_2
Clark County	(Find)	11	7.2 ± 0.8	4.4 ± 0.3	2.9 ± 0.5	0.074 ± 0.020	0.82 ± 0.03	69	15.2	1095	2.36
Mt. Ayliff	(Find)	13.6	5.17 ± 0.34	3.4 ± 0.3	2.0 ± 0.2	.046 ± 0.006	.60 ± 0.02		8.9[f]	868[f]	2.39
Aroos	1959	150	4.79 ± 0.40		1.7 ± 0.15	.039 ± 0.004	.49 ± 0.03	43	8.15	655	2.45
Williamstown	(Find)	31	3.48 ± 0.30		1.18 ± 0.10	.034 ± 0.004	.37 ± 0.02	29	5.3	465	2.53
Treysa	1916	63	2.47 ± 0.20	1.8 ± 0.2	1.15 ± 0.06	.030 ± 0.002	.38 ± 0.02	33	8.0	580	2.13
Carbo	(Find)	454	2.43 ± 0.26	1.4 ± 0.2	1.05 ± 0.15		.22 ± 0.01	18.5	3.2	315	2.64
Sikhot-Alin	1947	80,000	1.01 ± 0.08		0.49 ± 0.05	.0114 ± 0.0015	.13 ± 0.01	10.5	2.1	165	2.36
Bruderheim (metal phase)	1960	300	0.38 ± 0.17								

[a] "Find" indicates not an observed fall. [b] See *45*. [c] Standard temperature and pressure. [d] See *38*. [e] See *46*. [f] See *47*.

Table 6. Cosmic-ray-produced stable nuclides in some stone meteorites (*48*).

Meteorite	Weight at recovery (kg)	Amount of nuclide (10^{-8}cm^3/g[a])			
		He^3	Ne^{21}	Ar^{38}	Fe (%)
		Chondrites			
Bruderheim	300	47	10.1	1.41	22.5
Ehole[b]	2.4		4.78		~30
Harleton[b]	8		12.6		22
Richardton	90	32	9.5	0.96	30.6
		Achondrites			
Norton County (Calcium-poor)	1050	220	63	2.6	1.6
Johnstown[b] (Calcium-poor)	40	40.7	6.74	0.7	13.1
Nuevo Laredo (Calcium-rich)	0.5	3.8	0.45	1.79	
		Pallasite			
Admire (stone phase)	>50	134	41.2	0.87	8

[a] Standard temperature and pressure. [b] See *49*.

slight but significant variations in relative abundances, the data seem to be essentially consistent. The ratios of the neon isotopes Ne^{20}, Ne^{21}, and Ne^{22} are approximately 1 : 1 : 1, with systematic variations of a few percentage points. This is true for both iron and stone meteorites—a remarkable fact in view of the difference in the nuclear reactions involved. In the case of the argon isotopes Ar^{36} and Ar^{38}, the ratio is always about 2 : 3. The He^3, He^4 ratio is about 1 : 3. The yield of helium nuclei in spallation is comparatively high; the helium content of meteorites is generally of the order of 10^{-6} cm^3/g. This, and the very high relative abundance of He^3, made possible the first discovery of cosmic-ray products in meteorites by Paneth, Reasbeck, and Mayne, in 1952 (*15*).

These examples, and others, show that the relative yields of the common spallation products, those falling in or close to the stability "valley" on the chart of nuclides, are rather uniform. On the other hand the natural abundances of these isotopes show a very irregular pattern, having been produced in an entirely different way. The smooth distribution of products with charge and mass makes it much easier to predict the production rate of a new species.

Some of the anomalies in observed concentrations have led to other discoveries. For example, in the Williamstown iron meteorite (Table 3), the Cl^{36} content is too low, by a factor of about 4, as compared to the Cl^{36} content of the other long-lived radioactive species. This discrepant result, first obtained by Sprenkel (*16*), led to the conclusion that the Williamstown iron meteorite, which is a find rather than an observed fall, had been lying on the ground in Kentucky for about 600,000 years before its discovery. Many other meteorites of considerable terrestrial age have since been found. Two iron meteorites, Tamarugal and Ider, were found (*10, 17*) to contain almost no Cl^{36}—a result which indicates an age greater than a million years. Some stone meteorites are observed to contain little or no C^{14}; this lack of C^{14} indicates that even stone meteorites can be preserved for tens of thousands of years on the earth. For the measurements of terrestrial ages, Si^{32}, Ar^{39}, Ti^{44}, and Ni^{59} are also very useful.

Cosmic-Ray Age

Let us return to the question of the cosmic-ray age. As already described, this number is based simply on a ratio of concentrations of stable and radioactive species. If a production rate can be estimated accurately, the stable-nuclide concentration is sufficient. If this age is to be interpreted as a real time period, we must assume not only that the cosmic-ray intensity in space and time has been effectively constant but that the bombardment conditions in the meteorite, in particular the depth of a particular specimen below the surface, have remained essentially unaltered since its formation. The data shed light on these questions. Typical bombardment ages for iron meteorites are in the range of 10^8–10^9 years, while those for chondrites are in the range of 10^7 years. Both these ages are quite short as compared to the ages determined by uranium-lead, potassium-argon, and rubidium-strontium methods for the minerals contained in the meteorites. All these methods agree well on ages close to 4.5×10^9 years. The simplest explanation of these findings is that meteorites were formed as small bodies by violent collisions between larger objects a relatively short time ago. It has been thought that chondrites with ages of about 20 million years occur with special frequency (*18*). This might imply the occurrence of particular events which produced meteorites of these types in large numbers. However, these clusters cannot be considered to be definitely established. Another possibility is that the reduction in size of meteoritic bodies has occurred rather gradually through many small collisions—for example, with cosmic dust (the erosion hypothesis) (*19*). A younger age for the stones might be related to the softness of these bodies relative to iron. A number of strong objections have been raised to this erosion hypothesis in its extreme form (*20*), but the effect must undoubtedly occur to some degree. The effect of gradual erosion, or of a succession of collisions that remove small parts of the meteorite, would be a gradual increase in the effective intensity of the bombardment received by an originally deep portion of the body. At present the suspicion is that both large and small collisions may be important. It seems safe to adopt a model in which a single big collision first exposes the meteorite to an appreciable cosmic-ray flux. After this, erosion (and multiple small collisions) continue to act on the body, while occasional big collisions may reduce its size again drastically. Finally, the meteorite is either captured by the earth or one of the other planets, ejected from the solar system, or broken into pieces so small that they cannot reach the earth.

In a large meteorite the high-energy products, such as Ne* and Al^{26} from iron, occur in highest concentration on the outside. Their concentration decreases rather rapidly with depth. Specimens that were once buried deep in a larger body show much higher relative concentrations of low-energy products than specimens from nearer the surface. For example, in Table 3, Treysa shows smaller amounts of low-energy products than Carbo. The former must be from near the surface and the latter from near the center of their respective original bodies. The radioactive isotopes that we find whose half-lives are short as compared to the bombardment age must have been produced while the body had more or less its present size. Hence, their distribution with depth need not be the same as that of the stable nuclides. One would expect the low-energy products to be more prominent among the stable species. The data presented in our tables do not show any such effect. An especially favorable case for such study is the iron meteorite Grant, which has been studied extensively by Nier and his co-workers (*21*). They have presented contour maps of the rare-gas concentrations in this nearly spherical body. Goel and Kohman (*17*) have compared the depth dependence of Cl^{36} concentration with that for the stable Ar^{36}. The curve for depth dependence of Cl^{36} seems steeper, but at present the data do not fall into a consistent pattern. In other well-studied meteorites, such as Carbo and Casas Grandes, the contour lines for rare-gas concentration are not closed (*22*). Apparently these meteorites broke up in passing through the earth's atmosphere. Because of the great forces encountered by an irregular body in the atmosphere, this breaking-up process probably takes place in the great majority of cases, but often only one fragment is recovered. It would be very valuable to have more meteorites like Grant. The evidence for more than one major collision is particularly clear in the case of some very large iron meteorites, as was first pointed out by Vilcsek and Wänke (*23*). For example, in Table 3, in two specimens (I and II) of Odessa we find Cl^{36}/Ar^{36} ratios leading to two quite different ages, 100 and 300 million years. The same situation occurs in the large iron meteorite Sikhot-Alin. These second collisions exposed fresh or nearly fresh surfaces, and the high concentration of the radioactive species which built up thereafter led to an effective age intermediate between the times of the two collisions (if there were only two). In view of all these observations there is reason to believe that most iron meteoritic bodies in space have been substantially altered in size, on a time scale of 10^8 years or so.

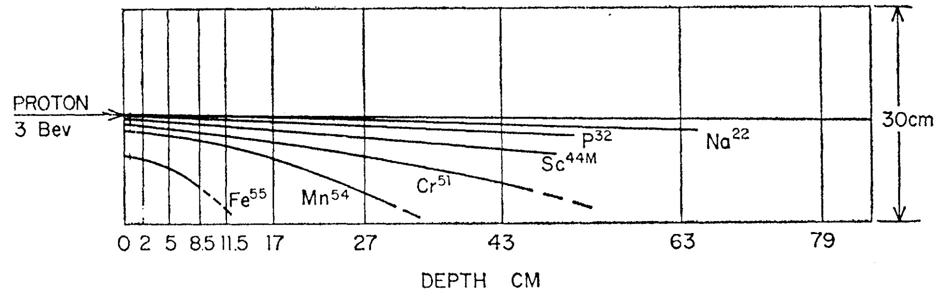

Fig. 2. Contours showing spread of the internal beam for different products (*26*). Half of the production of each species at any depth takes place inside the circle generated by rotating the corresponding line. Lower-energy products show wider spread.

Production Rates

A more detailed and quantitative treatment of relative and absolute production rates yields more results. These have been obtained in two ways. First, excitation functions in iron and other important elements have been measured for a good many species. By methods of nuclear systematics, a number of other excitation functions can be estimated with a good deal of confidence. Using the very extensive information available on primary cosmic rays and their interactions, one can estimate the flux of nuclear active particles as a function of depth in spherical meteorites, and in the limiting case of a very large body. The production rates estimated in this way are reliable to within perhaps a factor of 2, and fortunately it has been possible to verify the predictions in a number of cases where these really did precede the measurements. Unfortunately, very few cross-section data are available for magnesium, silicon, and calcium targets. It must be assumed that the substitution of neutrons and pions for protons as bombarding particles at moderate or high energies does not affect the cross sections (*24*). However, the smoothness of variation of cross section with product charge and mass, as studied by Rudstam (*25*) and others, saves us from any really serious errors. One is on particularly safe ground in comparing isobars —that is, nuclei of the same mass number. The shape of excitation functions is generally very closely the same for all isobars, and a single cross-section ratio is sufficient, except for a few cases very near the target mass.

A more direct experimental method is to simulate the conditions in space and to bombard a thick target of iron or stony material with a beam of high-energy protons, available at one of our larger accelerators. In such bombardment, large blocks of iron (*26, 27*) and also blocks of glass and metal (*27*) have been used to simulate the composition of chondrites. The products in the target were measured as a function of depth and lateral distance from the bombarding beam. The spread of the beam for different products may be seen in Fig. 2. Figure 3 shows the radius dependence of production rates. Many short-lived radioactive species have been measured. The resulting data can be integrated over angle and depth to obtain the effect of bombardment by a uniform isotropic flux.

What can be learned from this more exhaustive analysis? First, the conditions of bombardment can be better understood. A useful empirical formula, already inferred from the meteorite data themselves (*28*), is placed on a firm basis. This equation, for the total production rate of all species of mass number A, is

$$Q(A) = k(\Delta A)^{-k_2}$$

The net number of nucleons emitted, $A_{target} - A_{product}$, is ΔA. The constant k is proportional to the flux intensity, and k_2 is found to be a constant for $\Delta A > 5$. It decreases for smaller values of ΔA. The total isobaric yield can be estimated rather well from any convenient product of given A. For bombardment by a soft or low-energy flux, k_2 is large, and the yield falls off rapidly with ΔA. This constant, then, tends to increase with depth and size. A semilogarithmic plot of the total isobaric yields versus ΔA, for several nuclides, gives a comparatively accurate measure of k_2. This can be compared with the value for k_2 obtained by integrating the laboratory-bombardment data over all angles (Figs. 4, 5). Figure 4 shows how k_2 at the center of an iron sphere varies with the radius of the sphere. Figure 5 gives the variation in k_2 with depth for four different radii. The curve of Fig. 4 is the lower envelope of these curves.

Similarly, for the stable products, if the flux has been constant,

$$C(A) = k'T(\Delta A)^{-k'_2}$$

where T is the bombardment age and the constants are marked with primes to indicate that they refer to stable species. Now it is apparent that agreement between k_2 and k'_2 is a refined test of the constancy of bombardment conditions, while k and k' should agree if the flux has been constant. This is equivalent to the statement that the same T can be derived from each suitable pair of nuclides.

How much does k_2 vary? Its total observed range is from about 2.1, near the surface of small bodies (such as

Treysa) to somewhat less than 2.7 for the interior of large ones (such as Carbo). In large bodies the variation with depth is rapid at first, as the low-energy secondaries build up; then it slows down. As for the total production, it rises at first, has a broad maximum, and then falls steadily. By using both parameters, we can hope to get some measure of both the depth and the size of the original body. All this is illustrated in Figs. 2–5. Is erosion important in iron meteorites? If so, the curves have been traversed to the left, and k_2 and k'_2 must differ, the former being lower. If erosion is dominant, k'_2 should be an average of values, varying continuously from ~ 2.8 down to k_2. This would mean that k'_2 would be somewhat greater than 2.4 for Treysa, as compared to k_2 at 2.1, a difference well outside the limit of experimental error. No such difference is observed, the two values being the same as far as we can see. For a big object like Carbo, the two values would differ by less than 0.1, because of the flatter slope in this mass region. A difference of this degree cannot be excluded by the available data, and these can be used only to confirm the general correctness of the model. Fragmentary data on another small iron meteorite, Charlotte, appear to show a low value of k'_2, and this may be another good case to study.

Using the bombardment data (Table 1; Figs. 2 and 3), we can now demonstrate the constancy of the internal flux over millions of years to within 40 percent or so. Comparison of k_2 and k'_2 in Treysa shows that its size has been more or less constant over hundreds of millions of years. In this case, then, the internal flux can be directly tied back to the primary flux. Furthermore, the energy distribution of the primary flux cannot have changed enough, over the whole time of bombardment, to produce by itself a measurable change in k_2. For big meteorites, this says very little, but for small ones like Treysa, a large change in the "hardness" of the primary spectrum should show visibly.

The nuclide K^{40} is an entirely special case, with its half-life of 1.3×10^9 years. It is the only species available, even in principle, for checking the constancy of the cosmic radiation on a billion-year time scale. Unfortunately, there are difficulties. First, in the usual iron meteorites, whose cosmic-ray age is about 500 million years, only about 15 percent of the K^{40} will have decayed if the cosmic-ray intensity has been constant. This is not a very big change as compared with the differences already discussed. A very accurate estimate of the production rate relative to the stable species near it is required. Only one meteorite, Deep Springs, appears definitely to have a cosmic-ray age in excess of 1 billion years. This meteorite is a find, and apparently it has a considerable terrestrial age, so that one must make important corrections in comparing data for radioactive and stable species. Voshage (*29*) has shown that the cosmic-ray intensity in these old iron meteorites was (at least in most cases) somewhat less in the past than it is at present. It is possible that this is due to the progressive reduction in size that we have discussed, or even that the primary flux was higher long ago. We can say only that the cosmic-ray intensity many hundreds of millions of years ago was not very different from that at present. More work, including detailed comparisons of k_2 and k'_2, is needed.

At the other end of our time scale, variations in the concentration of the very-short-lived species such as P^{32}, Ar^{37}, V^{48}, Cr^{51}, and the still unobserved Mn^{52}, of 6-day half-life, and others, can tell us something about the very recent history of meteorites. In the case of orbits extending deep into the asteroid belt, one might expect effects of the variation of cosmic-ray intensity in space, especially at the periods of maximum in the solar cycle. The shielding effect of the

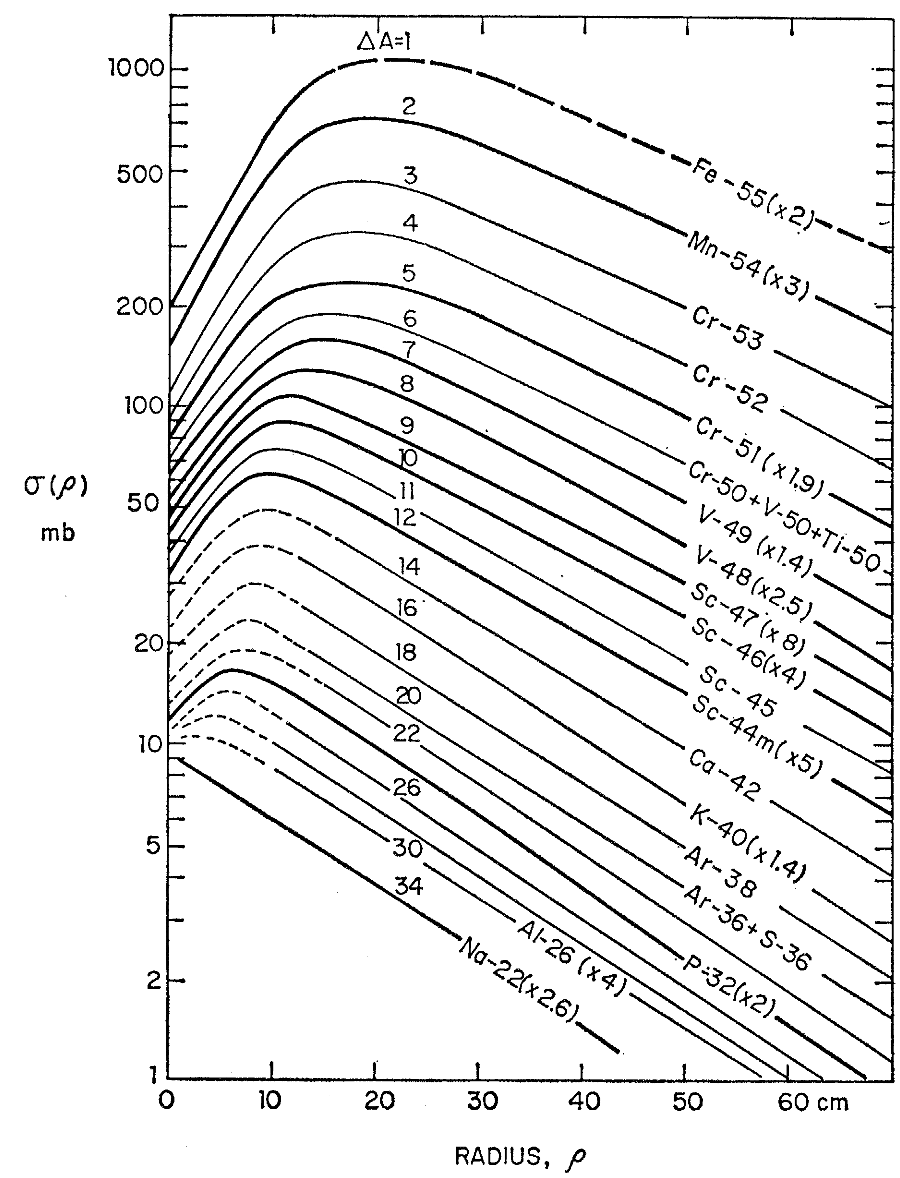

Fig. 3. Total isobaric yield from iron for each mass number as a function of radius. The isobars correspond to the production at the center of a spherical body. Light curves are interpolated between heavy curves derived directly from 3-Gev bombardment data (*26*).

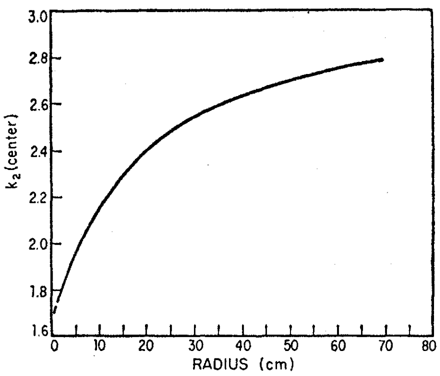

Fig. 4. The constant k_2 at the center of a spherical meteorite, as a function of radius.

sun's magnetic field ought to be less marked at these great distances than at the earth. The evidence is ambiguous (*30, 31*). Our own data do not show this effect. It may well be possible to see the activation produced in meteorites by the occasionally very intense bursts of particles from the sun.

The experience gained in this field may be utilized in two other branches of cosmochemistry. One of these is concerned with the much older record of high-energy nuclear reactions, currently believed to have occurred at the end of the process of general synthesis of the elements. In the earliest stages of evolution of our solar system, there is reason to believe, the acceleration of particles to high energies may have occurred at a much greater rate than at present. There is evidence for this (for instance, the presence of technetium) in some very young stars. According to Fowler *et al.* (*32*), the light elements deuterium, lithium, beryllium, and boron found in terrestrial materials and meteorites were produced at this stage. The total bombardment dose was perhaps 10^3 to 10^4 times higher than the "present" cosmic-ray bombardment in usual iron meteorites. Whatever the detailed mechanism may have been, these elements can only have been produced by some such nonthermal high-energy process. They must thereafter have remained at temperatures low on the million-degree scale of stellar interiors. The production process must have resembled, in important respects, present-day cosmic-ray production. In fact, the elements lithium, beryllium, and boron are very much enriched in the primary cosmic-ray beam itself, as compared to any general sample of galactic material available to us. This results from the bombardment of heavier cosmic-ray nuclei by the stationary interstellar hydrogen. If the conditions of high-energy bombardment differed to any considerable degree between the region of the earth and the region where meteorites originate, we might expect measurable differences, between the earth and meteorites, in the abundance pattern of elements and isotopes. Such differences have recently been observed (*33*).

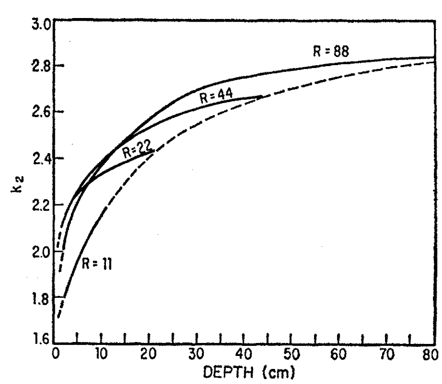

Fig. 5. The constant k_2 as a function of depth for meteorites of different radii. The dotted line represents the envelope (see Fig. 4).

Recovered Artificial Satellites

The analysis of recovered artificial satellites is currently of great interest (*34, 35*). Such studies have yielded some real surprises. Radioactive and stable nuclides have been detected in unexpectedly high amounts in satellites of the Discoverer series. These include tritium, Ar^{37}, Co^{57}, Ag^{106}, Xe^{127}, and Bi^{205}. Even stable He^3 has been measured in the materials of Discoverer XVII, exposed for less than a day. In other flights very small activities were seen, as would be expected in view of the very short period of bombardment. These bursts of production present many puzzling features and must, presumably, be explained either as effects of solar flares or as the result of extensive passage through the Van Allen belt. In the case of the Soviet satellite Sputnik IV, a piece of which was recovered in the United States after the satellite had been outside the earth's atmosphere for 843 days, the activation appears to have been produced mainly by cosmic rays. Since the satellite was close to the earth, the intensity was reduced by a factor of 4 to 5. There is some dispute about the extent of activation by solar particles (*35*).

There is a sense in which all of us in cosmochemistry (or theochemistry, as some call it) do nothing but prepare for the arrival of the first samples of matter from the moon and the planets. It should be easy for the reader to see, at this point, that this is true of the work we have described.

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Hormones and Sexual Behavior

Broad relationships exist between the gonadal hormones and behavior.

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Research on the relationships between the hormones and sexual behavior has not been pursued with the vigor justified by the biological, medical, and sociological importance of the subject. Explanation may lie in the stigma any activity associated with sexual behavior has long borne. In our experience, restraint has been requested in the use of the word *sex* in institutional records and in the title of research proposals. We vividly recollect that the propriety of presenting certain data at scientific meetings and seminars was questioned. Counteracting this deterrent is the stimulation which has come from colleagues in many disciplines to whom we have appealed for help, and the satisfaction we have felt in seeing a picture emerge as the pieces of the puzzle have been studied and fitted together.

Relationships in the Adult

Causal connections between gonadal hormones and the development of the capacity of infrahuman vertebrates to display sexual behavior have long been assumed, although the existence of such relationships in man is questioned (*1–3*). Doubt has also been expressed that a specific relationship exists between any one hormone (or class of hormones) and the behavior it facilitates in adults in general, from fish to man (*4–6*).

A number of explanations may be given for the uncertainty which exists. Human sexual activity is influenced by many psychologic factors, the social level, cultural background, and tradition. The many reports are not completely trustworthy. Physiological correlates with individual behavior are largely nonexistent, and controlled study in man as we know it in laboratory animals is impossible. In our opinion the many differences in behavior which in the growing child and adult are socially rather than hormonally determined have obscured the possible role of the hormones in maintaining the strength of the sexual drive. Even in lower mammals the same quantity of hormone elicits almost as many modes of response as there are individuals. This fact may have contributed to the doubt, to which we have alluded, that there is any great degree of hormonal specificity. In the human female, sexual responsiveness does not have the sharp relationship to folliculogenesis and to the functioning of the corpus luteum in the ovary that it does in most lower mammals (*7*). The degree to which this evolutionary change within the primates has been accompanied by an emancipation from the effects of hormonal action is not known.

The need for testicular androgen in the maintenance of sexual vigor in the male has been questioned by some students of the problem. In man (*5*, *8*), the dog (*9*), the domestic cat (*10*), fishes (*6*), and birds (*11*), there is, in males, a persistence of sexual activity for some weeks or months after castration which has not been explained satisfactorily; a corresponding persistence is encountered rarely if at all in females below the primates. The restoration of sexual vigor by replacement therapy also requires weeks in the male and only hours or days in the female. The longer time lapse which occurs, regardless of the direction of hormonal change, suggests that the manner of hormonal action in the male is greatly different from that in the female rather than that the strength of sexual behavior is independent of the presence of testicular androgen.

Finally, in this brief consideration of the subject, there are the important studies of deviant sexual types by Hampson and Hampson (*1*), Money (*2*), and the recent report by Völkel (*3*). The data these clinical investigators collected led them to conclude that the establishment of gender role or psychologic sex can be independent of chromosomal sex, gonadal sex, hormonal sex, internal reproductive structures, and external genital morphology. They relate the process rather to "the many experiences of growing up, including those experiences dictated by his or her own bodily equipment" (*1*).

The interest of one of us (W.C.Y.) in the relationship of the hormones to

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