Influx Measurements of Extraterrestrial Material

Sea sediments, polar ice, air, and space are searched for amount and character of interplanetary debris.

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The nonplanetary "bits and pieces" of the solar system range from dust grains smaller than 1 μ in size to chunks many kilometers across, and include asteroids, comets, meteorites, meteors, and zodiacal dust. The zodiacal light and its near-sun equivalent, the F corona, provide evidence, via light scattering, for the existence of dust in space (*1*). Small particles have short lifetimes in space. Radiation pressure from sunlight rapidly repels opaque particles about 0.2 μ in radius from the solar system; it also provides a drag causing larger particles to spiral in toward the sun and to assume an increasingly circular orbit. The time for opaque particles in circular orbits to spiral into the sun as a result of this Poynting-Robertson drag is

$$T \sim 7 \times 10^6 \, s \, \delta \, a^2 \text{ years} \qquad (1)$$

where s is the radius and δ the density of the particle in centimeter-gram-second units, and a, in astronomical units, is the distance of the particle from the sun. If the particles are electrically charged, interactions with solar plasma affect their lifetimes; the magnitude of this charge and even its sign remain obscure (*2*).

With short lifetimes, continuing sources are required. Two have been particularly discussed—disintegration of comets, and collision between asteroids. Whipple has estimated that about 1 ton/sec is adequate to sustain the zodiacal cloud (*3*). His more recent estimates, in which account is taken of collisions of dust with dust, suggest a 10 to 20 tons/sec injection rate (*4*). Comets seem able to provide this material. However, the mass distribution of comets as a function of space and time is poorly known; the cometary destruction rate is also hard to estimate. Calculations of asteroidal material ground up in collisions have suggested that the debris created, ~ 100 tons/sec, might also be adequate (*5*). However, most of this is in objects far too large to account for the zodiacal cloud, and if the cloud is in steady state, then asteroidal collisions seem an inadequate source.

If we could collect dust and demonstrate it to be extraterrestrial, we could learn much about its chemistry, mineralogy, amount in space, and history and origin. Such collections have been extraordinarily difficult. The total amounts of material are small, the individual grains are tiny, methods for distinguishing the real thing from contaminants are uncertain, the velocities at satellite heights lead to vaporization on impact, and the extent of survival and preservation on earth is unknown.

Deep-Sea Spherules and Sea-Level Collections

Murray and Renard's discovery in 1876 of black magnetic "cosmic spherules" in deep-sea deposits marks the beginning of serious study of small extraterrestrial fragments. A few hundred spherules up to 200 μ in size were found in a kilogram of red clay. Two types were present: a magnetite kind, sometimes including a metal core, and a stony kind, similar to chondrules of stony meteorites. Murray and Renard suggested that the magnetite spherules are formed when molten droplets ablate from a meteorite entering the atmosphere. Pettersson and Fredriksson (*6*) found higher concentrations of spherules (several thousand per kilogram greater than 30 μ) in core surfaces; at most depths the concentration was a few hundred per kilogram. Brownlow *et al.* (*7*) also found concentrations of a few hundred per kilogram greater than 30 μ, with fluctuations as a function of depth. The fluctuations might reflect variations in influx or variations in sediment accumulation rates. Others have determined influx rates by counting spherules collected from air, ice, and ancient sediments. They obtain widely varying rates (*8*); much discussion has resulted about which spherules are handling contaminants, volcanic debris, meteoritic ablation products, cometary fragments, and so forth. Fredriksson and Boström (*9*) have noted magnetite spherules in carbonaceous chondrites, and these spherules may be found in sea sediments, although none examined from carbonaceous chondrites have metal cores.

Brownlow *et al.* (*10*) have attempted to collect extraterrestrial material from sea-level air, exposing 1-m^2 greased nylon meshes to coastal winds. Every 2 days the dust was washed off. Metallic flakes of nearly pure iron and some others of nearly pure nickel were found, together with a few magnetite spherules. These spherules were all homogeneous magnetite, whereas one-third of the deep-sea spherules are metal-centered. Later, Fredriksson found alloy compositions among the flakes, often with manganese in 1 percent amounts, and industrial contaminants were suspected. More recently, Parkin and coworkers (*11*) have made sea-level collections during a year at Barbados, with extreme precautions to minimize contamination. Three 1-m^2 meshes were suspended from a tower on coral cliffs facing the trade winds from the sea. The salty sea spray provided a sticky coating, and no grease was applied. Metallic flakes and spherules could never be eliminated, but as new precautions were introduced their numbers decreased to very low values. A blank control (an unexposed mesh washed identically to those exposed) showed comparable numbers, and therefore the low values include contaminants. No extraterrestrial dust particles (> 50 μ), magnetic or nonmagnetic, different from those on the blank were found during the year. Without blank subtraction, the spherules greater than 10 μ in diameter, if collected from the metered

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volume of wind, would have provided 0.3 ton to Earth per day. The deep-sea spherule influx to clay deposited over 10^4 to 10^5 years is about 1 ton/day. After blank subtraction, the Barbados results suggest that essentially no extraterrestrial spherules were collected. Reconciliation with the deep-sea results implies erratic arrival, as if from atmospheric ablation of meteorites arriving at random intervals. This is quite different from the spherules continuously arriving as fine-grained material which is melted and oxidized when it enters the atmosphere—suggested *(12)* to explain the observed size distribution, by means of size-dependent melting.

Öpik *(13)* and Whipple *(13)* have calculated maximum temperatures for grains entering the atmosphere. For a particle of radius s, velocity at the top of atmosphere v, and zenith angle of entry z, T_{max} is given by:

$$T_{max} = (k\, s v^3 \cos z)^{1/4} \qquad (2)$$

Small or low-velocity particles can enter without melting; if their surface-to-mass ratio is high enough they rapidly reradiate energy acquired from atmospheric slowing down. For example, at $v = 11.4$ km/sec (zero inclination, direct, circular orbits) 100-μ iron flakes could survive. The fourth-power dependence of s on T_{max} will radically discriminate in favor of refractory material.

Castaing and Fredriksson *(14)* found that the nickel and cobalt content of metal-centered deep-sea spherules was similar to that of iron meteorites—nickel-rich in the metal core but low in the magnetite. Schmidt and Keil *(15)* showed that manganese was ~ 0.01 percent, comparable to iron meteorites. Considerable data demonstrate that if an iron-nickel alloy is heated and oxidized, the nickel concentrates in the metal and is depleted in the oxide *(16)*. Marvin and Einaudi *(17)*, however, have recently observed magnetite spherules in beach sands with nickel present at meteoritic levels (~ 5 percent). Nickel may indicate extraterrestrial origin; its absence in magnetite gives little information. A further problem with the beach and deep-sea spherules mentioned above is that some most likely to have been derived by ablation of iron meteorites (significant Ni, low Mn, or metallic core) have ~ 10^2 times higher Cr abundance than the irons.

Figure 1 shows iron-manganese contents for deep-sea, volcanic, and airborne or polar spherules. Deep-sea spherules are separated from the others. Volcanic spherules have around 0.5 percent manganese; airborne and polar, about 1 percent, suggesting possible contamination. It is important to have precise blank controls. Many of the spherules earlier reported *(10, 18)*, have been questioned as contaminants *(11)*; their origin remains uncertain. There is another complication in identifying spherule origin. The best estimate for the "cosmic" ratio of manganese to iron is probably based on the relatively unfractionated type-I carbonaceous chondrites. The ratio, 8×10^{-3}, is similar for other classes of stony meteorites. But this ratio, plotted in Fig. 1, is close to the center of gravity of polar and airborne spherule distributions. For comparison the Mn/Fe ratio of 2×10^{-2} in average Earth's crust is also plotted. Evidently manganese is a poor discriminator between crustal and "cosmic" abundances, despite its low abundance in highly differentiated iron meteorites.

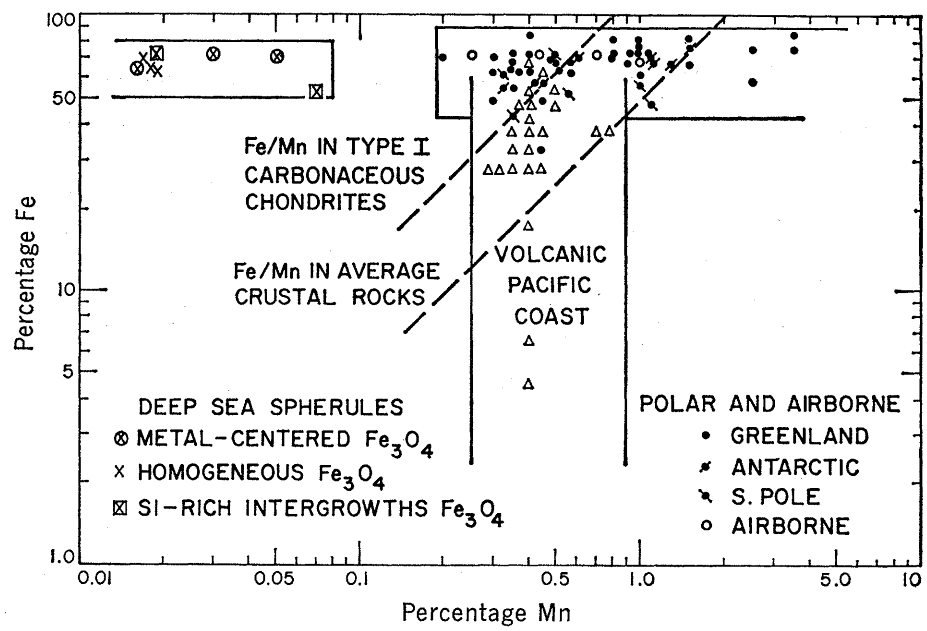

Fig. 1. Spherules from different sources. [Note: Wright *et al.* *(18)* probed only the surface of the spherules; sometimes they found < 0.5 percent manganese, and these values are plotted as if just below 0.5 percent. However, in later work, to be published, they probed polished interiors. Here manganese values are precisely stated and they are all > 0.2 percent.]

The mesh technique would not collect smoke-sized particles (0.1 μ). Parkin *et al.* *(11)* assume that particles less than 1 μ would not be retained, but that above this, 50 percent is collected (meshes have 50-percent open area). The evidence for this is: (i) Transmission coefficients, determined with several meshes in series, are 45 percent. (Cigarette smoke is little affected by a series of meshes.) (ii) Bhandari (see *11*) has determined radioactive-fallout collection efficiency, and finds rough agreement with a 50-percent factor. Fallout is mostly on 0.5- to 1-μ particles at sea level *(19)*. (iii) Calculations *(20)* suggest that at sea level for the Barbados meshes the transition from high to low collection efficiency occurs in the 1- to 10-μ-size region, and is inversely proportional to particle specific gravity.

During sampling of ~ 10^6 m^3 of air during 2-day periods, gram quantities of fine (< 10 μ) brown dust were collected, apparently from land across the Atlantic. This completely overwhelms any nonmagnetic fine dust from space. However, variable amounts of fine magnetic dust, absent from the blank, were found. In the summertime, concentrations averaged around 10 parts per million; in winter, about 0.1 ppm. X-ray diffraction analysis of the fine magnetic dust gives mainly maghemite (γ-Fe_2O_3), and neutron activation by Goles, on an August sample, shows 0.5 percent manganese. If the wintertime value were wholly extraterrestrial, the accretion would be 0.18 ton/day. This implies an upper limit of about 2 tons/day of fine extraterrestrial dust of size 1 to 50 μ, assuming a 10-percent magnetic-to-nonmagnetic ratio, typical of chondritic material. If there were mechanisms capable of varying the product of influx and fallout by a large factor, a more conservative upper limit from annual magnetic deposition is less than 600 tons/day. If most of any magnetic material is converted on atmospheric entry to smoke that is scavenged onto nonmagnetic debris, either limit is further weakened. Nonetheless, the large seasonal variation suggests a terrestrial origin for most, if not all, of the magnetic fraction.

Airplanes, Balloons, and Rockets

Windblown dirt is reduced as it leaves the Earth's surface. In June 1966 Arnold and his colleagues were successful in a stratospheric balloon flight (*20*). Here a silicone-oil-coated 10-m^2 mesh was suspended 6000 feet (1800 m) below a balloon that drifted for 10 hours at 70,000 feet. With known wind shear, 4×10^5 m^3 of air were sampled. During ascent and descent the mesh was rolled in a tightly sealed box. Before recovery, insects entered the box, and a few soil-like particles ($\sim$ 100 μ) were found. However, no fine nonmagnetic or magnetic dust was seen. Eight magnetic flakes and one black magnetic spherule observed are suspected contaminants: microprobe analysis of three gave a composition (99 percent iron, 1 percent manganese, < 1 percent nickel) similar to steel.

The total mass collected corresponds to an upper limit for influx of 550 tons/day if there were zero blank contributions and no terrestrial components. However, the experiment included a flight blank, exposed briefly but otherwise identical to the sample meshes. The mass per unit area on this blank was two to three times that found on the collection meshes, and the particle characteristics were indistinguishable. Thus the actual upper limits are lower, but it is impossible to estimate how much lower. The influx limit is independent of composition, except that it does not include materials soluble in freon (used to dissolve the oil from the meshes after flight). Smoke will not be collected unless scavenged onto larger particles, which are extremely rare at 64,000 feet.

Smoke has been observed in the stratosphere, but it is doubtful that much is extraterrestrial. Junge (*21*) found a layer of 0.15-μ particles with maximum concentration of $\sim$ 0.1 per cubic centimeter at 20 km, falling to 0.01 per cubic centimeter at 25 km. This layer seems constant with time. Sulfur is the main element. Junge thinks the particles are formed *in situ* by oxidation of SO_2 and H_2S, present in the troposphere and penetrating to the stratosphere. In a recent smoke-collecting experiment (*22*), air at 19 to 21 km was filtered with a mat of polystyrene fiber, prepared under very clean conditions. Neutron-activation analysis gave sulfur (and/or chlorine), iron, sodium, and chromium in detectable amounts. The measured Fe/Na ratio was 1.2 ± 1.2. Since the same ratio in chondrites is 36, whereas in average crustal rocks and sea salt it is 1.8 and 10^{-8}, respectively, the iron numbers can be used to place limits on the influx of submicron extraterrestrial material. These limits, assuming chondritic abundances, would correspond to < 250 tons/day if all of the iron were extraterrestrial.

Rosen (*23*) has flown a photoelectric counter and measured the dust concentration from 0 to 30 km. Light scattered from 0.25- to 1-μ dust was detected by a photomultiplier. At 20 km a layer was observed with concentration 3 per cubic centimeter. The Bali volcanic eruption (Mount Agung) in March 1963, before these flights of August 1963 and January 1964, may have perturbed the results (*24*). Above this layer, concentration was proportional to pressure, suggesting a possible extraterrestrial origin and influx of $\sim$ 10^4 tons/day. However, any volcanic source that injected fine material (or gases that formed aerosols) to altitudes above 30 km will be indistinguishable from an extraterrestrial source. Apparent layers injected by Agung were observed at heights up to 53 km in October 1963, and Junge has also observed layers between 20 and 30 km. Newkirk and Eddy, using a balloon-borne coronagraph, have also observed light scattering from particles above 25 km (*25*). They infer an influx rate of $\sim$ 10^3 tons/day for particles of less than 3 μ.

In the Venus Flytrap, flown 6 June 1961, collectors opened from a rocket nose cone at about 80 km—30 seconds after rocket burnout—and continued upward to 150 km. On descent past 110 km, the leaves were closed. Assorted particles have been reported, including irregular "fluffy" ones. Despite major technical efforts to identify when the particles were actually deposited, contamination remains uncertain. An influx of 10^4 tons/day is suggested (*26*). However, a similar flight—Luster—(Farlow *et al.*, *27*) has not confirmed

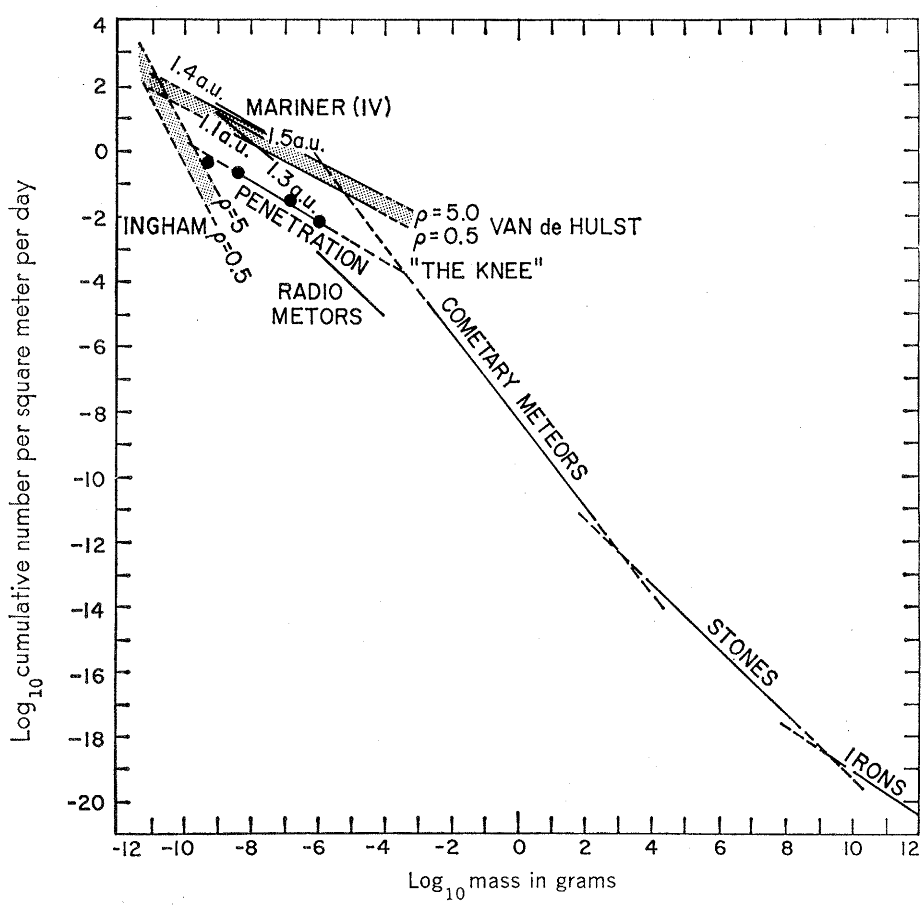

Fig. 2. Cumulative number influx to Earth of extraterrestrial material [number of particles per square meter per day with mass greater than *m* (grams)].

these results. The Luster rocket was launched during the Leonid meteor shower, 16 November 1965, and 2 m^2 of collecting surface were exposed. Great care was taken to avoid contamination, and control slides were processed identically to flight slides. Of the particles above 4 μ not identified as contaminants, upper limits were less than ~ 3 percent of apparent Flytrap fluxes. The debris on flight-exposed surfaces was also observed on nonflight controls; "more spheres and fluffy particles" were found on flight surfaces. This may measure the debris contributed by the rockets.

Satellite Experiments

The acoustic satellite results are now in doubt. The microphone methods of Alexander *et al.* (*28*) have been criticized by Nilsson (*29*). Apparently the microphones pick up noises caused by strains from thermal gradients as the satellites move in and out of Earth's shadow, and these were attributed to dust impact. The microphone results had been difficult to reconcile with astronomical findings. The microphones recorded numbers of particles $< 10^{-8}$ g greatly above those expected from the zodiacal light estimates. This led to discussion of a dust belt around the Earth. However, there appears to be no mechanism for maintaining high enhancements over interplanetary background (*2, 30*). The acoustic experiment (*31*) on the more constant-temperature Mars probe, Mariner IV, far away from Earth gave much lower impact rates than similar devices on earth-orbiting satellites. Because of uncertainty about background noises in the acoustic sensors, even these figures should perhaps be regarded as upper limits. The reported data are plotted in Fig. 2.

Naumann (*32*) has reviewed satellite penetration experiments. Explorers 16 and 23 exposed helium-filled pressure cells to dust bombardment, and registered punctures by observing loss of pressurization. Pegasus 1, 2, and 3 used thicker aluminum sheets forming one of the plates of a condenser. Penetrations shorted the condenser. To derive a mass distribution from the measurements, equations for puncture versus mass, density, and velocity of impacting projectile are used. Assuming density of 1.7 g/cm^3 and average impact velocity of 19 km/sec, Naumann and Marshall derive a mass influx in the region of 10^{-6}-g to 10^{-10}-g particles of ~ 10^2 tons/day. A cumulative mass distribution derived from penetration data is given in Fig. 2, with the velocity and density averages assumed. The assumption of particle densities of 0.4 g/cm^3, and geocentric velocities outside the Earth's field of about 5 km/sec (corresponding to low-inclination, nearly circular orbits, with impacting velocities of ~ 12 km/sec) would increase the inferred influx to ~ 500 tons/day. The penetration experiments do not detect a dust belt.

Radioactivities in Deep-Sea Sediments and Polar Snow

Meteorites, compared to terrestrial rocks, contain measurably anomalous amounts of many radioactive and stable isotopes (*33–35*). These isotopic anomalies, produced by corpuscular radiation in space, or more complex events during the meteorites' history, give definitive indications of extraterrestrial origin. They have been used to identify meteorites and other objects as having been in space. Similar measurements can be used to try to identify interplanetary dust. In the search for isotopic evidence of extraterrestrial dust, the sensitivity is optimized by collecting samples where the terrestrial sedimentation rate is low—deep-sea sediments and polar ice.

Wasson (*36*) concluded that in dust the highest production of many radioisotopes would be by solar-flare protons. These are solar material accelerated by sporadic explosive events on the sun to energies of ~100 Mev or higher. Production of many isotopes is most efficient in the range 10 to 100 Mev. The average energy of solar particles is much less than that of galactic cosmic rays; their average flux is higher. Using measured energy spectra, estimated fluxes, nuclear-reaction cross sections, and background and sensitivity for counting different radioactivities, Wasson derived "most favorable" radioactive nuclides for this search. His conclusion, that Al^{26} was the best isotope, was qualified by the possibility that dust lifetimes in space were too short for the buildup of maximum (saturation) levels of radioactivity.

Solar-flare proton flux Φ bombards dust in space and produces radioactive isotopes. Let n_i be the number of atoms per gram of dust of the *i*th parent having a production cross section σ_i for an isotope. The production of radioactive atoms is $\Phi\Sigma n_i\sigma_i$ g^{-1} sec^{-1}. With N atoms, their rate of decay is λN (λ = decay constant). Net production rate is

$$\frac{dN}{dt} = \Phi\Sigma n_i\sigma_i - \lambda N \quad (3)$$

After exposure time T the number of atoms is

$$N = \frac{\Phi\Sigma n_i\sigma_i}{\lambda}(1 - e^{-\lambda T}) \quad (4)$$

per gram of dust. Production cross sections are strongly energy-dependent, so that

$$\Phi\Sigma n_i\sigma_i = \Sigma n_i \int \bar{\phi}(E)\, \sigma_i(E)\, dE \quad (5)$$

where $\bar{\phi}(E)$ is the average flux of solar-flare particles per unit energy having energy E.

The deposition of fine-grained dust on the Earth's surface is probably latitude-dependent, and a part is soluble. We thus need a factor f, the fraction insoluble, and a factor l for the latitude effect. If I g cm^{-2} sec^{-1} is worldwide influx, then measured insoluble influx is Ifl. Let A be the specific activity per gram of terrestrial sample. The present concentration is A/λ; if the sample was deposited at time t in the past, with terrestrial sedimentation of R g cm^{-2} sec^{-1}, the initial flux of atoms was

$$\frac{A}{\lambda} R\, e^{\lambda t} \mathrm{cm}^{-2}\, \mathrm{sec}^{-1} \quad (6)$$

If Eq. 4 is multiplied by Ifl, and set equal to Eq. 6, worldwide influx of extraterrestrial material is

$$I = \frac{AR}{fl\, \Phi\, \Sigma n_i\sigma_i} \frac{e^{\lambda t}}{(1 - e^{-\lambda T})} \mathrm{g\ cm}^{-2}\, \mathrm{sec}^{-1} \quad (7)$$

As an example, consider the low level γ-γ coincidence measurements of Al^{26} in sea sediment (*37, 38*). Lal and co-workers measured Al^{26} and Be^{10} and found a Al^{26}/Be^{10} ratio of 0.12 ± 0.04. The ratio of these isotopes from cosmic-ray interactions in the atmosphere is ~ 10^{-2}. If all the Be^{10} is from the atmosphere, as calculations suggest, there appears to be excess Al^{26} in the sediment. The average Al^{26} activity is about 0.4 ± 0.2 dpm/kg (dpm = disintegrations per minute). Any decay in the core enhances the initial excess. Calculations also indicate that galactic cosmic-ray production in meteorites or dust, and nuclear reactions at the earth's surface, cannot account for this excess. However, solar cosmic rays acting on dust can explain the observations.

Combing chondritic abundances with

flare-proton energy spectra and nuclear cross-section data, a value for $\Sigma n_i \sigma_i$ of about 10^{-4} cm^2/g results for Al^{26}. The cores, according to Be^{10} measurements, have accumulated at the rate of 4 mm/10^3 years, which with a bulk density of 0.55 corresponds to $R = 7 \times 10^{-12}$ g cm^{-2} sec^{-1}. We neglect latitude effects ($l = 1$) and decay ($t = 0$); the aluminum residence time in the oceans is short ($f = 1$). We assume that saturation factor $(1 - e^{-\lambda T})$ is unity. The flare fluxes are uncertain; we assume ~ 200 protons cm^{-2} sec^{-1} (> 10 Mev) which is between the lowest and highest estimates. Then $I = 7.5 \times 10^{-8}$ g cm^{-2} year$^{-1} \approx 10^3$ tons/day to Earth. These figures would imply that 3 parts in 10^4 of the sea sediment are extraterrestrial. The influx is inversely proportional to average flare flux and saturation factor, so that if, as assumed by Lal and Venkatavaradan, the average dust lifetimes give one-fifth of saturation, the influx is increased by a factor of 5. However, the highest Al^{26} activities were found in a high latitude (57°S) core, where fallout measurements suggest several times average deposition and this tends to counterbalance the above factor. Saturation depends on lifetime in space, which depends on size, density, and orbital distributions. The proton-flux average could be too low (because of infrequent enormous flares) or too high (because the last cycle was probably the most intense in at least 100 years, and measurements are confined to part of this one cycle).

Wasson *et al.* (*39*) have also detected Al^{26} in a Pacific sediment, finding 0.9 $\pm$ 0.15 dpm/kg of sediment—consistent with the highest activity reported by the Lal group of 0.78 $\pm$ 0.34—although the sedimentation rate was not determined, and the sample was not recycled to constant activity. The results tend to support each other, since the chemical procedures of Wasson *et al.* led to metal whereas Amin *et al.* counted the oxide. However, in each case chemical purifications must be relied on to distinguish Al^{26} from other positron emitters, as the characteristic 1.83-Mev γ-ray in coincidence with the positron emission was too weak to observe. Each group has suggested other allowable combinations of solar-flare flux, saturation factor, terrestrial sedimentation rate, and influx. Fireman and Langway (*40*) searched for Al^{26} in particulates filtered from melted Greenland ice, and found upper limits of 4×10^{-7} dpm/liter and 3×10^{-6} dpm/liter of ice. The Greenland measurements are difficult to interpret because the efficiency of stirring of particles from the bottom of the ice is unknown, and because soluble or filterable (colloidal?) material was not sampled. We note that the reported particulate concentration in the Camp Century water (*41*)—2×10^{-4} g/liter—is an order of magnitude less material per square centimeter per year than that found in the oceanic sediments of lowest accumulation rate; the Greenland matter was further reduced by a factor of 5 by ashing. The published Greenland limits are difficult to compare with the sea sediments where the aluminum residence time is short. Attempts are underway (*42*) to eliminate this complication by also measuring Al^{26} in a water-soluble Greenland fraction collected with ion-exchange resins.

Schaeffer *et al.* (*43, 44*) have searched for Cl^{36} in sea sediments. With a 3×10^5 year half-life the activity should be closer to saturation than is Al^{26}. Meteoritic Cl^{36} is primarily produced by energetic cosmic rays interacting with iron and calcium, and the long-term average galactic cosmic-ray flux is well established (*33*). Washing eliminated chlorine from sea salt; remaining chlorine was extracted, purified radiochemically, and counted. The activity in two samples was $\leq 0.057 \pm 0.006$ dpm/kg in one and indistinguishable from background in another; the lower activity reflects more carbonate. Assuming chondritic composition, saturation-Cl^{36} activities, and sedimentation rates of 1 mm/10^3 years, implies an upper limit for influx of $\sim 5 \times 10^3$ tons/day. The result is uncertain because (i) the saturation factor and sedimentation rate are uncertain; (ii) recoil losses are more significant for such an energetic reaction than for Al^{26} production; (iii) the residence time of chlorine in the ocean is long (which is why the sea is salty) and so dissolved Cl^{36} is not measured. Besides spallation there may be another source of Cl^{36}—secondary neutron capture on Cl^{35}. Neutrons are produced by interactions of primary cosmic rays with solids. Although secondary neutrons will not produce much Cl^{36} in fine-grained dust, it is plausible that in hydrogen-rich material (cometary chunks?) higher in volatile chlorine than ordinary chondrites, the combined effects of efficient moderation and more chlorine may provide much higher Cl^{36} activities than in typical meteorites. If neutron capture produced significant Cl^{36}, some of which was in insoluble minerals, the spallation-based influx limits would be further reduced.

Fireman (*45*) has reported Co^{60} in particulates from several-hundred years-old arctic ice. Spallation of low-abundance nickel isotopes can produce Fe^{60}, which with a half-life of 10^5 years decays to Co^{60} of half-life 5.3 years. Unfortunately, Co^{60} is a common contaminant from bombs, reactors, and steel. But contamination Co^{60} will decay with 5.3-years' half-life, whereas that from Fe^{60} will not decay until the lifetime of Fe^{60}. Thus experiments should resolve the Co^{60} origin. Another approach is to "milk" and count Co^{60} building up from Fe^{60}. Both experiments are underway (*42, 45*). Iron-60 has a higher threshold for production from nickel than does Al^{26} from its targets. Its flare enhancement would be smaller than that of Al^{26}, although this may be counterbalanced by its shorter half-life and closer approach to saturation. The apparent levels of Co^{60} seem much too high to be consistent with observed Al^{26} in sea sediments, and to force agreement, some 10^2 times higher nickel abundance than in chondrites would have to be assumed.

Rare-Gas Measurements

A different technique, first to provide isotopic evidence of extraterrestrial dust on Earth, is mass spectrometric examination of rare gases, pioneered in sea sediments by Merrihue (*46*). A 2-g magnetic concentrate of a Pacific red clay was heated to successively higher temperatures. Each gas fraction at 600°, 1000°, and 1400°C was purified, and the amounts and isotopic abundances were determined. The ratio He^4/He^3 was 3070, 2380, 1120 at the respective temperatures, and 2540 total. This is similar to the gas-rich meteorites and not to the atmospheric He^4/He^3 ratio of 7.7×10^5. The Ar^{40}/Ar^{36} ratio was 407, 268, and 172 at the respective temperatures, and 340 total. The ratio of 172 found at 1400°C is markedly lower than the 296 ratio in the atmosphere. The Ar^{36}/Ar^{38} ratio showed no change with temperature, and was indistinguishable from the 5.35 atmospheric value. In 1 g of a nonmagnetic fraction of the clay, similar He^4/He^3 ratios were found. The He^3 measurements require confirmation.

Tilles (*47, 48*) found similar argon anomalies in a dense ($\delta > 3.2$) fraction of Greenland dusts concentrated from samples collected by Langway from a tank in the Camp Century water supply through which 10^7 liters of unfiltered melt water had passed. Other observed argon isotope ratio variations with temperature [Bieri (*49*), the Brookhaven group (*44*), and Tilles (*47, 48*)] are compared in Fig. 3 with Merrihue's values. These measurements were made on widely varying amounts of sample, which probably explains the varying anomalies observed. The anomaly-bearing material is apparently present worldwide, although it has not been observed in all sea-sediment samples examined (*49*), probably a result of varying degrees of terrestrial dilution and small sample size. No known terrestrial samples have been observed with Ar^{40}/Ar^{36} ratios less than that of air. Terrestrial rocks contain Ar^{40}/Ar^{36} ratios greater than 296 because of Ar^{40} retention from natural K^{40}.

The possibilities that cosmogenic (spallation) He^3 in dust can be used for identification, can enable inference of an influx rate, and can contribute to atmospheric He^3 were suggested by Mayne (*50*). There have been no rare-gas anomalies observed and confirmed in the dust that can be definitely attributed to cosmic-ray spallation, although Merrihue did find possible indication of excess Ne^{21}. The Ar^{36}/Ar^{38} ratios were indistinguishable from those in air, and very different from those produced by spallation. The amounts of spallation gases created during the short dust-exposure times seem too small to be easily observed in the presence of other contributions. One manganese nodule, found by Megrue to contain metallic iron-nickel spherules, was also found to contain apparent excesses of He^3 and Ne^{21}, with a Ne^{21}/Ne^{22} ratio at 600°C of about 1.3 (*44*) compared to the air ratio of 0.03. However, a second nodule showed no anomalies. The extent, if any, of incomplete outgassing of meteoritic ablation products (which initially contain major amounts of spallation isotopes) and of their incorporation into nodules is unknown.

Three plausible sources of the anomalous argon in fine-grained concentrates, all extraterrestrial, are (*46–48, 51*): (i) retention of gas trapped in mineral lattices during condensation or formation of dust, comets, or meteorites; (ii) retention of solar-wind ions stopped by grains in space; and (iii) retention of energetic ions from solar-flare streams bombarding grains in space. All should provide excess Ar^{36} and Ar^{38} in approximately atmospheric proportions, and in much larger amounts than Ar^{40}. The gas is probably in fine-grained dust that enters the atmosphere without melting, as in the Öpik-Whipple micrometeorite theory.

If we knew the concentrations of anomalous gas in "pure" extraterrestrial dust, we could use the observed anomalies to calculate influx rates for the unmelted fraction. Unfortunately we do not. Concentrations from (i) are uncertain, and widely varying meteorite observations exist (*35*). If a magnetic concentrate included 1 percent of material with meteoritic release like that, for example, found in Renazzo (see *52*) the argon observations could be explained. However, Merrihue and Tilles also found at 500° to 600°C the He^4 and Ne^{20} to be in roughly cosmic abundances relative to each other; these could not be explained in this way. Some of the gas-rich meteorites contain rare gases with resemblances to the dust (*35, 53*), although its origin in these is also uncertain. The amount of solar-wind gases stopped in dust can be calculated, and the concentration, assuming no loss, would be independent of size and density of the particles (*51*). However, for all but the finest grains, sputtering and other loss mechanisms will give saturation concentrations below this maximum; gas loss during atmospheric entry and storage is also likely (*54*). There is no compelling evidence of observation of solar-wind ions in dust, but extensive low temperature search is desirable.

Calculations for solar-flare ions, that should be retained if any extraterrestrial contributions are retained, may be useful in deriving upper limits for influx. Comparison of calculations with observations has suggested that perhaps 10 percent of the concentrates examined mass spectrometrically may be extraterrestrial (*48*). Since in the sea sediments the concentrates were about 2 percent, this corresponds to $\sim 10^{-3}$ of the sediment being extraterrestrial. This is

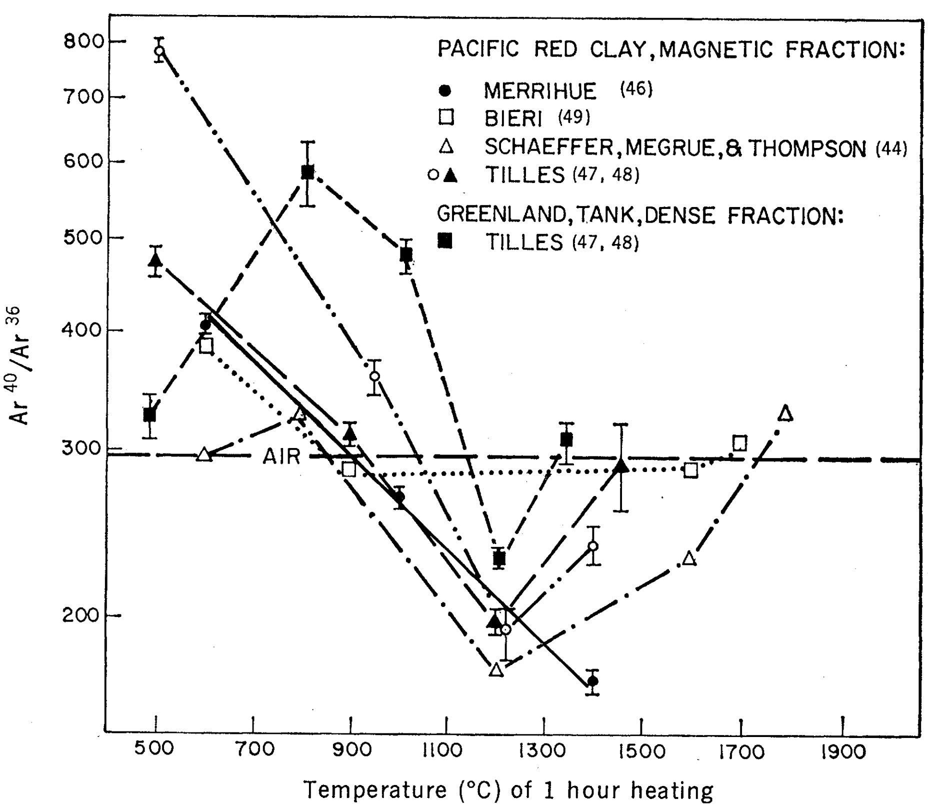

Fig. 3. Ratio of Ar^{40} to Ar^{36} in gas released from anomaly-bearing concentrates of fine-grained dust during successive 1-hour heatings. Authors and references are as given. Return toward air ratio for temperatures above about 1300°C is caused by (i) the fact that almost all of the argon has already been released at lower temperatures; (ii) increasing air argon blanks released from very hot furnace walls. Note that the mass of Bieri's sample was at least a factor of 5 smaller than any of the other samples plotted. All plotted points that gave anomalous Ar^{40}/Ar^{36} ratio had Ar^{36}/Ar^{38} ratio (within errors) indistinguishable from that in air.

comparable to the Al^{26} figure if Al^{26} is undersaturated by a factor of 3. The conclusions depend on long-term flare fluxes—a matter of uncertainty. Considering possible contributions from solar-wind and "primitive" sources and uncertainty in atmospheric gas loss, it is not possible to infer influx rate from rare-gas data. However, since these contributions would all increase the gas concentrations in "pure" extraterrestrial dust, the solar-flare-based influx may give upper limits. With an assumed sedimentation rate of 1 mm/10^3 years, and a 10 : 1 ratio for total-to-magnetic material, this would give an upper limit of 7×10^3 tons/day.

Rare-gas anomalies are by far the largest isotopic anomalies observed in dust, and can be measured in fractions of a gram compared to hundreds of grams for radioactivities. They show great potential for monitoring which materials or minerals of unmelted particulate matter are extraterrestrial. In combination with x-ray diffraction studies, they have suggested that extraterrestrial magnetite may exist prior to atmospheric entry (*47*).

Trace Elements by Neutron Activation

Paradoxically the greater the enhancement of extraterrestrial activities or isotope anomalies by flare-particle spallation, solar-ion stopping, or secondary neutrons, the easier it may be to observe extraterrestrial material, but the harder it is to infer influx rates. For the enhancements carry their own uncertainty, and the greater the enhancement the larger the uncertainty.

There have recently been some elegant attempts to determine influx rates from chemical measurements alone. The uncertainty in f (insoluble fraction) can be eliminated in polar ice by measuring soluble and insoluble fractions, and in the ocean by selecting elements of short residence time. Derived influx then depends on the chemical composition of incoming material. Contamination from terrestrial sources is likely to be serious, and is attacked in various ways. The simplest is to measure a pair of elements whose abundance ratio in Earth's crust is widely different from that in meteorites or the sun. For example, nickel relative to most elements is rarer in terrestrial rocks than in chondrites or sun by $\sim 10^2$ times. This is assumed to be a result of fractionation in the Earth, with nickel enriched in the core and depleted in the crust.

A higher concentration of nickel (0.05 percent) in deep-sea sediments than in average crustal rocks (0.02 percent) lead Pettersson and Rotschi (*55*) to attribute the excess to extraterrestrial sources. With the variability of nickel content in sediments and crust, the reality of this difference is questionable; in any case, submarine vulcanism may introduce nickel and other cations into the marine environment in relative amounts different from average crust (*56*), and preferential precipitation may alter abundances in different types of sediments (*57*).

Brocas and Picciotto (*58*) have determined nickel by neutron activation of Antarctic ice kept frozen until after irradiation. Near the South Pole they find an average concentration of 1.5×10^{-9} gram of nickel per gram of snow. Individual concentrations vary by 10^2 times, although in 13 of 15 samples total nickel varied by less than a factor of 5. Two-thirds of the nickel was insoluble; the rest passed a 0.05-μ filter. In adjacent aliquots of ice, sodium, potassium, calcium, chlorine, and magnesium were measured; the ratios Na/Ni, K/Ni, Ca/Ni and Mg/Ni were all closer to those of chondrites than to those of typical terrestrial rocks. In fact, the upper limit to the Mg/Ni ratio ($\lesssim 4$) (based on a single large-sample magnesium determination) is lower than that in meteorites by a factor of 2 or 3 but lower than that in crustal rocks by a factor of 70. The nickel blanks are less than one-third of the lowest samples, and a few percent of the average. Brocas and Picciotto consider that at least 95 percent of the nickel in the South Pole ice is probably extraterrestrial. Using chondritic abundances, they calculate worldwide influx rates, assuming accretion and fallout proportional to area, and fallout with latitude-dependence of bomb Sr^{90}. Influx values are 10^4 tons and 3×10^4 tons/day. The South Pole samples are from 1950 to 1953 snow and are from less than 10^3 cm^2 of the Earth's 5.1×10^{18}-cm^2 surface.

Nearer the coast the average nickel was 4.3×10^{-9} g/g of snow. The ratios, though also higher, were again less than those of terrestrial rocks. The snowfall here, 40 g cm^{-2} $year^{-1}$, is higher than at the South Pole, 7 g cm^{-2} $year^{-1}$, but the higher nickel cannot be caused by precipitation-scavenging alone, since direct proportionality is not observed. There is probably more windblown terrestrial dust and sea spray at the coast than at the South Pole, and the fact that at both sites the average ratio of soluble to insoluble nickel is the same is puzzling, as it suggests the same source at both locations. Perhaps a latitude effect is the explanation, as, based upon bomb-fallout studies, more extraterrestrial fallout should be deposited around 40° to 50°S, as a result of seasonal meteorological mixing of stratosphere and troposphere, than at the South Pole.

Another promising method is that of Barker (*59*), who has studied noble metals in sea sediments. The noble metals are depleted in terrestrial rocks compared to meteorites by larger factors than nickel. Barker has noted that an inverse correlation between noble-metal concentration and sea-sediment accumulation rate can give the terrestrial abundance in the sediments and the extraterrestrial fraction. The slow accumulation rates integrate over short-term fluctuations. With neutron-activation, Barker has studied iridium and osmium, and on the basis of iridium has tentatively estimated, assuming chondritic compositions of extraterrestrial material, an influx of 3×10^2 tons/day to Earth. He believes this correct within a factor of 3.

Astronomical Methods

Photographic meteor studies have been summarized (*60*) and are plotted in Fig. 2 as "cometary meteors." The cumulative number of sporadic meteors N m^{-2} day^{-1} having mass greater than m (grams) is:

$$\log_{10} N = -8.15 - 1.34 \log_{10} m \qquad (8)$$

Radar methods have extended the data to smaller masses, and are also plotted (*61*). For an estimated mass range of 10^{-4} to 10^{-6} g, the cumulative flux is (in same units as above):

$$\log_{10} N = -9.2 - 1.05 \log_{10} m \qquad (9)$$

There are significant uncertainties in the mass-luminosity calibration and the ionization efficiency required to derive the plotted curves from observations. Work is underway to extend the meteor curve to larger masses via the Prairie Network of camera stations (*62*). The mass distribution of the meteorites—that do not completely burn up—has been estimated (*60, 63*), and Hawkins' cumulative curves are plotted in Fig. 2. A variety of complex observational biases exist in reducing the known meteorite falls and finds. (For example, the estimated mass of one iron meteorite, Sikhote Alin, which fell in 1947, is 70 tons, and dominates the total mass of all historically observed falls.)

Shoemaker (*64*) has suggested, on the basis of infrasonic airwave observations, that an order of magnitude more mass enters the atmosphere in large chunks than that estimated from falls of the chondritic meteorites. The statistics for such large objects, of which the Tunguska event in Siberia is an example, are poorly known, but great short-term variations in rate are expected at the large end of the mass distribution. Such variations influence results at a single time and location. Red clay integrating 10^5 years (10-cm total depth) may have numerous horizons from such big events. If substantial chunks that are friable, volatile, soluble, or readily dispersed, enter the atmosphere, they may provide material difficult to recognize as extraterrestrial at Earth's surface. In initial sizes of a centimeter or greater this material would introduce only small amounts of flare-enhanced radioactivities or rare gases; depending on amount, size, hydrogen content, and chemical composition, such material might introduce significant amounts of neutron-capture isotopes. It would certainly provide chemical contributions.

Astronomical measurements of color, intensity, and polarization of scattered light as a function of position provide estimates of concentration, size distribution, and spatial distribution of zodiacal dust in space. These determinations depend on complex calculations involving shapes of grains, optical constants for unknown materials, surface charateristics, and albedo, all of which are unknown. Classical scattering from uniform spheres has usually been assumed—surely unrealistic. Possible solutions cannot be shown to be unique. Distinguishing light from the zodiacal cloud from other light in the night sky is difficult. Finally, a number concentration in space can be converted to a terrestrial mass influx only if the average specific gravity is known—and it is not. It is not surprising that widely varying concentration estimates exist.

Expressions for size or mass distributions usually take the form

$$dn = k\frac{ds}{s^x};\ \text{or},\ dn = C\frac{dm}{m^{(x+2)/3}} \quad (10)$$

where dn is the number of particles per cubic centimeter of space with radius s to $s + ds$ or with mass m to $m + dm$, and k and C are constants. Van de Hulst (*1*), analyzing zodiacal-light observations and eclipse data on the corona, and assuming no variation of concentration with distance from the sun, derived a value for x of 2.6. This is a fairly flat distribution with most of the mass in the larger (100-μ) particles. Assuming particle densities of 5, he derived a spatial density of 5×10^{-21} g/cm^3. If one assumes particle densities of 0.5—a figure consistent with meteor strengths and fragmentation—the spatial density is reduced.

The mass flux on unit cross-sectional area of a planet is

$$N = nu\left(1 + \frac{v_e^2}{u^2}\right) \quad (11)$$

where u is the relative velocity at great distance from a planet whose escape velocity is v_e, and n is the mass concentration in space. In the region of the Earth, particles in direct circular heliocentric orbits would largely accompany the Earth. However, differing inclinations i ($\sim 6°$) result in an average relative velocity of $u \sim 3$ km/sec. Assuming Van de Hulst spatial density (corrected for assumed density of 0.5), we have a calculated daily influx of about 250 tons/day, and the distribution is plotted in Fig. 2. More realistic assumptions about eccentric orbits will alter this, but probably not by more than a factor of 3. The assumed uniform spatial density must be modified for the Poynting-Robertson drift, which gives, for circular orbits, an inverse concentration with radial distance from the sun; a variety of other radial variations have been suggested as fitting the observations, including efforts to take account of injection and destruction mechanisms (*65, 66*).

Actually, the average spatial density, which is

$$\bar{D} = \int_{S_1}^{S_2} dn(s)\left(\frac{4}{3}\pi\rho s^3\right) ds \quad (12)$$

is, for the size distribution derived by Van de Hulst, proportional to the 1.4 power of the largest size particles included in the model. Neither observations nor model of zodiacal cloud provides a reliable determination of this quantity, and these influx estimates are thus also uncertain for this reason.

Cometary meteors have a radius distribution $ks^{-5}\,ds$ and cross the Earth's orbit. The perihelia of their parent comets are less than 1 astronomical unit (a.u.). If dust in the penetration size range is in circular orbits by the time it reaches 1 a.u., then it must have originated with perihelia substantially greater than 1 a.u. If this dust originated outside Jupiter, it would explain the "knee," in Fig. 2, since objects larger than this would be eliminated by Jupiter's strong gravitational field, whereas smaller particles would have a high enough inward-drift velocity to sift past without capture. The radar results suggest a possibility of a differential distribution of $Cm^{-2}\,dm$ or $ks^{-4}\,ds$ for the smaller cometary meteors (*61*), which, because of their high speed and their radiant positions, are assuredly noncircular and must have been recently ejected from Earth-crossing comets. For any ancient dust that is sifting past the earth in already circularized orbits, the Poynting-Robertson drift flattens the concentration distribution, compared to that in the original source, by an additional power of s (since it introduces a radial velocity proportional to s^{-1}, so that small particles migrate faster, reducing their steady-state concentration). Thus the final size distribution near the Earth of material released from comets with perihelia much outside 1 a.u. might be $ks^{-3}\,ds$ or $Cm^{-1.66}\,dm$, which would agree with Van de Hulst and penetration data. The cometary meteor-size distribution in the photographic region cannot be changed to the observed penetration-size distribution by Poynting-Robertson drift alone, as a factor s^2 would be required.

A combination of severe grinding (*67*), for example by asteroidal collisions, together with Poynting-Robertson drift, would give an $s^{-3}\,ds$ size distribution for fine material. The observable asteroid size-distribution does not give evidence of severe grinding (*68*). However, Whipple (*69*) has noted that dust-dust fragmentation may be important, and this might help explain the "fully ground" character of the fine-grained distribution. Yet according to Whipple's recent calculations (*4*) it is only for particles of mass greater than 10^{-4} to 10^{-5} g that collisional and erosional lifetimes seriously limit Poynting-Robertson lifetimes, while it is just for masses smaller than this that the "fully ground" distribution is observed. We would expect a "fully ground" $s^{-4}\,ds$ distribution for those particles with lifetimes limited by collisions, and an s^{-3} ds distribution for smaller daughter particles, created by collisions of larger particles but with lifetimes limited by the Poynting-Robertson effect. This is an alternative explanation for a change

Table 1. Influx to Earth. (For details see text.)

Recent time (days to years)		Long term ($\geqq 10^4$ years)	
Experimental technique	Amount (tons/day)	Experimental technique	Amount (tons/day)
	Small size in space (< 0.1 cm)		
Penetration satellites	100 to 500	Al^{26} (sea sediment)	200 to 10^4
		Rare gases	$\leqq 10^4$
		Zodiacal cloud:	
		Van de Hulst	250 to 2500
		Ingham and others	0.2 to 2
	Cometary meteors (10^{-4} g to 10^2 g in space)		
	200		
	"Any" size in space		
Barbados meshes:			
Spherules	< 0.3	Ni (sea sediment)	$< 10^4$
Total winter	< 2	Os (sea sediment)	3×10^2
Total annual	$\leqq 600$	Cl^{36} (sea sediment)	$< 5 \times 10^3$
Balloon meshes	$\leqq 550$	Sea-sediment spherules	1 to 10
Airplane filters	$\leqq 250$		
Balloon: dust counter	10^4		
Balloon: coronagraph	10^3		
Ni (Antarctic)	10^4 to 3×10^4		
	Large size in space		
Airwaves	10^2		
Meteorites	1 to 10		

in slope in the vicinity of the "knee" and at about the mass predicted by Whipple's calculations. However, to account for the complete change in slope from cometary meteors to penetration data still requires at least one additional mechanism—perhaps some characteristic meteoritic source-size distribution produced by evaporative fragmentation of comets in the inner solar system.

Besides the flat size distributions of the Van de Hulst type, there have been a number of attempts to fit the observations with steep size distributions: $x \sim 4$ to 5. These models [the "Ingham-Giese" type distributions (*65*)] require little total mass, mainly in small ($\sim 1\ \mu$) particles, and seem able to account for polarization observations without invoking electrons. This is desirable, as the low measured densities of electrons in the solar wind cannot account for the polarization. It may be that a combination of both kinds of models with many very small particles providing polarization but little mass, and fewer larger particles providing most of the mass but little polarization, is possible. Calculated influx rates to Earth for the Ingham-Giese type models are in the range of 0.2 to 2 tons/day. Most of this fine material is smaller than that sampled by penetration satellites.

For material of size just above the radiation-pressure limit, the orbital velocity will be reduced compared to larger bodies, a result of radiation counterbalancing solar gravity. The velocity relative to Earth will be substantial (with Earth "catching up" with these particles even if they are in circular orbits). We have plotted two of these Ingham-Giese type distributions in Fig. 2 for an assumed average relative velocity of 10 km/sec, densities of 0.5 and 5, and a differential population index of $x = 4$. More complicated models, with the fine-grained polarization-producing component replaced by surface irregularities on Van de Hulst size dust balls may also be possible (*70*); they are difficult to analyze theoretically.

Dust will "sand-blast" objects unprotected by an atmosphere. The extent of erosion on meteorites and the moon can give information about the average flux of dust. The earliest applications of this idea (*71*) were to iron meteorites, and made use of the "exposure ages," times exposed to cosmic rays as lightly shielded objects in space. These times, determined from measurements of stable and radioactive isotopes, depend on current and ancient production rates, which in turn are related to the shielding history of the object. The ductility of iron meteorites is high, and cratering deforms without efficient removal (*72*), so that the responses of irons to impact are complex, and may depend on temperature of ductile-to-brittle transition. However, combining Comerford's laboratory measurements of erosion of a chondrite with assumed relative velocity of 10 km/sec and typical chondrite exposure ages gives upper limits for dust flux to Earth of about 10^3 tons/day (*72*). Lower relative velocities would give weaker limits.

Photographs of the lunar surface provide information on the size distribution of exposed pebbles and boulders. It will be possible to determine exposure times from isotopic measurements on returned lunar samples, which will give limits for erosion rates. Data about size distributions of impacting objects can be derived from crater studies of the moon and Mars (*73*), although without direct measurements of crater ages such results remain uncertain with regard to inferred absolute influx rates.

Discussion

We attempt to combine in Table 1 an approximation to the various influx estimates and to indicate the mass ranges sampled and the time spans integrated over.

For the "long-term" observations, values range from about 1 ton/day to Earth (minimum from spherules in sea sediments) to upper limits of several times 10^4 tons/day. "Cosmic" spherules in sea sediments are in order of magnitude agreement with what is expected from ablation of meteorites (1 to 10 tons/day total) and not inconsistent with short-time spherule limits from the Barbados meshes. These latter (< 0.3 ton/day) cannot be compared with sea sediments because of the random arrival of large meteorites. The Cl^{36} and rare gas determinations give upper limits of 5×10^3 to 7×10^3 tons/day. Aluminum-26 gives influx in sizes smaller than about 0.1 cm of $\sim 10^3$ tons/day (assuming saturation activities), but with uncertainties in sedimentation, chemical compositions, lifetimes in space, long-term average flare-particle fluxes, and low specific activities, this may not be better than an order of magnitude indication. An order of magnitude lower Al^{26} influx could be accounted for by atmospheric production. The sea-sediment nickel values, although perhaps influenced by extraterrestrial contributions, could be entirely accounted for by terrestrial processes without difficulty. Thus for long-time averages, this leaves the number inferred from osmium in sea sediments—about 3×10^2 tons/day—standing nearly alone for total mass influx to Earth. This gives no information about size distribution. The Van de Hulst zodiacal-light models give 250 to 2500 tons/day, depending on assumed average specific gravity of particles, and on maximum particle size for which derived slope applies. The Ingham-Giese type models would give $\sim$ 1 ton/day.

The most direct of the "short-term" measurements are the satellite-penetration experiments. These give an influx of 100 to 500 tons/day, depending on assumed velocity and density distributions, and an apparent size distribution roughly matching the Van de Hulst type zodiacal-light models. The Antarctic nickel determinations imply a flux of order 10^4 tons/day for periods of a year or so, but give no information about size of objects in space. The balloon-borne coronagraph and dust-particle counter give 10^3 to 10^4 tons/day, but it is hard to be certain these are uninfluenced by terrestrial sources. The stratospheric filter and Barbados and stratospheric mesh experiments give upper limits $\leqq$ several times 10^2 tons/day, based on measurements of iron or magnetic fraction and assumed chondritic abundances. These experiments sample below the region at which fragile, friable, or oxidizable material will have been dispersed, although the filters sample efficiently down to very small grain size. The upper limits are consistent with the penetration satellite or osmium results. The fine magnetic dust measurements on Barbados during winter months give total influx ($> 1\ \mu$) $\leqq$ 2 tons/day. Sampling efficiency for smoke produced during atmospheric entry is unknown. Smoke, if scavenged onto large tropospheric particles, will be lost among large amounts of nonmagnetic terrestrial debris caught on the meshes. In the stratosphere, where terrestrial debris is much reduced, unattached smoke would not be collected by the meshes in any case. As far as ground-based or atmospheric collections are concerned, any amount of material oxidizable at low temperatures could exist in outer space in a wide range of particle sizes. No sample has yet been analyzed in detail for hydrocarbons. The Shoemaker airwaves imply $\sim 10^2$ tons/day average coming in big chunks, eratically distributed with perhaps an order of magnitude uncertainty in actual average in flux rate.

The total mass contributed by cometary meteors whose final form at Earth's surface may be mostly unrecognizable as submicron fragments, oxidation products, hydrocarbons, or other debris of any initial "dust-balls," is sensitive to the position of the "knee," and inversely proportional to the luminous efficiency. Higher luminous efficiency than that used in deriving the curve in Fig. 3 would shift the cometary meteor curve to lower masses, and decrease the influx. The influx is 200 tons/day if the plotted curve is extended to meet the penetration data and 700 tons/day if extrapolated to 10^{-5} g.

Measurements thus appear to be converging on 10^2 to 10^3 tons/day total current influx to Earth, with only Antarctic nickel measurements suggesting significantly higher values. The nature of most of this material is unknown, but it is desirable to search for carbonaceous material. There is some indication of its possible presence in polar snow (*10*). The relative amount contributed by small and large objects in space remains uncertain. The relationship of the long-term average to short-term measurements also remains uncertain, but should provide information about the sources of extraterrestrial debris in space. Fortunately, the chemical and isotopic techniques, as well as cosmic spherule counts, enable study of variations as a function of time over extended parts of the geologic record, particularly in sea sediments (*74*) and possibly in sedimentary rocks (*75*) and for more recent periods in ice. "Paleometeoritics" will doubtless be expanded in future years.

The identification of specific mineralogical constituents of interplanetary dust with mass spectrometric study of rare-gas anomalies appears very promising, but it is fair to say that *no* fine-grained material other than the high-nickel metal-cored spherules originally described as extraterrestrial 90 years ago has even now been completely and unambiguously identified as extraterrestrial in the presence of ubiquitous terrestrial contamination. The challenge of identifying and describing this material is perhaps now as urgent as, and more uncertain than, the question of total influx rate (*76*).

References and Notes

1. H. C. Van de Hulst, *Astrophys. J.* **105**, 471 (1947).
2. I. I. Shapiro, D. A. Lautman, G. Colombo, *J. Geophys. Res.* **71**, 5695 (1966); M. J. S. Belton, *Science* **151**, 35 (1966).
3. F. L. Whipple, *Astrophys. J.* **111**, 375 (1950); **113**, 464 (1951).
4. ———, *Smithsonian Astrophys. Obs. Spec. Rept. No. 239*, in press.
5. S. L. Piotrowski, *Acta Astron.* **5a**, 115 (1953).
6. H. Pettersson and K. Fredriksson, *Pacific Sci.* **12**, 71 (1958).
7. A. E. Brownlow, W. Hunter, D. W. Parkin, *Geophys. J.* **12**, 1 (1966).
8. R. A. Schmidt and T. J. Cohen, *Science* **145**, 924 (1964).
9. K. Boström and K. Fredriksson, *Smithsonian Inst. Misc. Collections* **151**, 1 (1966).
10. A. E. Brownlow, W. Hunter, D. W. Parkin, *Geophys. J.* **9**, 337–388 (1965).
11. D. W. Parkin, A. C. Delany (Mrs.), A. C. Delany, *Geochim. Cosmochim. Acta*, in press.
12. E. J. Öpik, *Irish Astron. J.* **4**, 84–135 (1956).
13. ———, *Publ. Obs. Astron. Univ. Tartu* **29**, 1–69 (193' → F. L. Whipple, *Proc. Natl. Acad. Sci. U.S.* **36**, 687 (1950).
14. R. Castaing and K. Fredriksson, *Geochim. Cosmochim. Acta* **14**, 114 (1958).
15. R. A. Schmidt and K. Keil, *ibid.* **30**, 471 (1966).
16. ———, and D. E. Gault, *Proc. 7th Hypervelocity Impact Sympos.* **5**, 105 (1965); H. Fechtig and K. Utech, *Ann. N.Y. Acad. Sci.* **119**, 243 (1964); A. Goresy and H. Fechtig, *Smithsonian Astrophys. Obs. Contr. Astrophys.* **11**, 391 (1967); P. Ramdohr, *Chem. Erde* **25**, 259 (1966); J. Moreau and J. Benard, *Compt. Rend.* **232**, 1842 (1951).
17. U. B. Marvin and M. T. Einaudi, *Geol. Soc. Am. N.E. Section, Program for 2nd Ann. Mtg.*, abstract, pp. 40–41 (1967); *Geochim. Cosmochim. Acta*, in press.
18. W. D. Crozier, *J. Geophys. Res.* **71**, 603 (1966); F. W. Wright, P. W. Hodge, C. C. Langway, Jr., *ibid.* **68**, 5575 (1963); P. W. Hodge and F. W. Wright, *ibid.* **69**, 2449 (1964); *ibid.*, p. 2919.
19. L. B. Lockhart, R. L. Patterson, Jr., A. W. Saunders, Jr., *J. Geophys. Res.* **70**, 6033 (1965).
20. J. R. Arnold, N. Bhandari, D. W. Parkin, *ibid.*, in press.
21. C. E. Junge, *Air Chemistry and Radioactivity* (Academic Press, New York, 1963).
22. J. P. Shedlovsky and S. Paisley, *Tellus* **18**, 499 (1966).
23. J. M. Rosen, *J. Geophys. Res.* **69**, 4673 (1964).
24. M. P. Meinel and A. B. Meinel, *Science* **142**, 582 (1963).
25. G. Newkirk, Jr., and J. A. Eddy, *J. Atmos. Sci.* **21**, 35 (1964).
26. C. L. Hemenway and R. K. Soberman, *Astron. J.* **67**, 256 (1962).
27. N. H. Farlow, M. B. Blanchard, G. V. Ferry, *J. Geophys. Res.* **71**, 5689 (1966).
28. W. M. Alexander, C. W. McCracken, L. Secretan, O. E. Berg, in *Space Research III* (North-Holland, Amsterdam, 1963), pp. 891–917.
29. C. Nilsson, *Science* **153**, 1242 (1966).
30. G. Colombo, D. A. Lautman, I. I. Shapiro, *J. Geophys. Res.* **71**, 5705 (1966); G. Colombo, I. I. Shapiro, D. A. Lautman, *ibid.*, p. 5719; D. A. Lautman, I. I. Shapiro, G. Colombo, *ibid.*, p. 5733.
31. W. M. Alexander, C. W. McCracken, J. L. Bohn, *Science* **149**, 1240 (1965).
32. R. J. Naumann, *NASA Tech. Note D-3717* (1966).
33. M. Honda and J. R. Arnold, *Science* **143**, 203 (1964).
34. J. R. Arnold, *Ann. Rev. Nucl. Sci.* **11**, 349 (1961); O. A. Schaeffer, *Ann. Rev. Phys. Chem.* **13**, 151 (1962); E. Anders, *Space Sci. Reviews* **3**, 583 (1964); ———, in *The Moon, Meteorites and Comets*, G. P. Kuiper and B. M. Middlehurst, Eds. (Univ. of Chicago Press, Chicago, 1963), pp. 402–495; J. Zähringer, *Ann. Rev. Astron. Astrophys.* **2**, 121 (1963).
35. R. O. Pepin and P. Signer, *Science* **149**, 253 (1965).
36. J. T. Wasson, *Icarus* **2**, 54–87 (1963).
37. D. Lal and V. S. Venkatavaradan, *Science* **151**, 1381 (1966).
38. B. S. Amin, D. P. Kharkar, D. Lal, *Deep-Sea Res.* **13**, 805 (1966).
39. J. T. Wasson, B. Alder, A. Oeschger, *Science* **155**, 446 (1967).
40. E. L. Fireman and C. C. Langway, Jr., *Geochim. Cosmochim. Acta* **29**, 21 (1965).
41. C. C. Langway, Jr., and E. L. Fireman, *Trans. Am. Geophys. Union* **46**, 116 (1965).
42. R. H. McCorkell, E. L. Fireman, C. C. Langway, Jr., *Trans. Am. Geophys. Union* **48**, 158 (1967).
43. O. A. Schaeffer, G. H. Megrue, S. O.

Thompson, *Ann. N.Y. Acad. Sci.* **119**, 347 (1964).
44. ———, private communication of work to be published.
45. E. L. Fireman, *Smithsonian Astrophys. Obs. Contr. to Astrophys.* **11**, 373 (1967).
46. C. M. Merrihue, *Ann. N.Y. Acad. Sci.* **119**, 351 (1964).
47. D. Tilles, *Smithsonian Astrophys. Obs. Contr. to Astrophys.* **11**, 399 (1967).
48. ———, *Science* **153**, 981 (1966).
49. R. H. Bieri, private communication.
50. K. I. Mayne, *Geochim. Cosmochim. Acta* **9**, 174 (1956).
51. D. Tilles, *Science* **148**, 1085 (1965).
52. J. H. Reynolds and G. Turner, *J. Geophys. Res.* **69**, 3263 (1964).
53. P. Signer and H. E. Suess, in *Earth Science and Meteorites* (North-Holland, Amsterdam, 1963), p. 241; H. E. Suess, H. Wänke, F. Wlotzka, *Geochim. Cosmochim. Acta* **28**, 595 (1964).
54. G. K. Wehner, C. Kenknight, D. L. Rosenburg, *Plantetary Space Sci.* **11**, 885 (1963); E. Anders and E. Mazor, *Geochim. Cosmochim. Acta*, in press; D. Tilles, *Trans. Am. Geophys. Union* **48**, 157 (1967).
55. H. Pettersson and H. Rotschi, *Geochim. Cosmochim. Acta* **2**, 81 (1952).
56. E. Bonatti and G. R. Nayudu, *Am. J. Sci.* **263**, 17 (1965); E. Bonatti, in *Researches in Geochemistry*, P. Abelson, Ed. (Wiley, New York, 1967), vol. 2.
57. E. D. Goldberg, *J. Geol.* **62**, 249 (1954).
58. J. Brocas and E. Picciotto, *J. Geophys. Res.* **72**, 2229 (1967).
59. J. L. Barker, Jr., private communication. This method was also suggested by P. A. Baedecker and W. D. Ehman, *Geochim. Cosmochim. Acta* **29**, 329 (1965).
60. G. S. Hawkins, *Ann. Review Astron. Astrophys.* **2**, 149 (1964).
61. C. S. Nilsson and R. B. Southworth, private communication.
62. R. E. McCrosky and H. Boeschenstein, Jr., *Smithsonian Astrophys. Obs. Spec. Rept. No. 173* (1965).
63. H. Brown, Jr., *J. Geophys. Res.* **65**, 1679 (1960).
64. E. Shoemaker and C. J. Lowery, *Meteoritics* **3**, 123 (1967).
65. M. F. Ingham, *Monthly Notices Roy. Astron. Soc.* **122**, 157 (1961); R. H. Giese, *Space Sci. Reviews* **1**, 589 (1962–1963); J. L. Weinberg, *Ann. Astrophys.* **27**, 718 (1964); R. D. Wolstencroft and L. J. Rose, *Astrophys. J.* **147**, 271 (1967).
66. R. B. Southworth, *Ann. N.Y. Acad. Sci.* **119**, 54 (1964); M. J. S. Belton, *Proc. Zodiacal Light and Interplanetary Medium Conf.*, Honolulu (1967).
67. G. S. Hawkins, *Astron. J.* **65**, 318 (1960).
68. E. Anders, *Icarus* **4**, 399 (1965).
69. F. L. Whipple, *Smithsonian Astrophys. Obs. Contr. to Astrophys.* **7**, 239 (1963).
70. N. B. Richter, *The Nature of Comets* (Methuen, London, 1963).
71. F. L. Whipple and E. L. Fireman, *Nature* **183**, 1351 (1959).
72. M. F. Comerford, *Geochim. Cosmochim. Acta*, in press.
73. F. L. Whipple, *Proc. Roy. Soc. London Ser. A* **296**, 304 (1966).
74. D. Tilles, *Icarus*, in press.
75. T. A. Mutch, *Earth and Plan. Sci. Letters* **1**, 325 (1966).
76. While this paper was being revised, three reviews of parts of the subject appeared [*4*, *73*, and J. F. Vedder, *Space Sci. Reviews* **6**, 365–414 (1966)]. We would particularly like to thank J. R. Arnold and N. Bhandari for their interest and advice, and R. B. Southworth, R. E. McCrosky, A. F. Cook, II, U. B. Marvin, G. S. Hawkins, and R. H. McCorkell for helpful comments on the manuscript. The recent dust-collecting expedition at Barbados was financed by Science Research Council, U.K. D.W.P. acknowledges support from NASA (grant NSG-321). We would also like to thank J. L. Barker, Jr., G. H. Megrue, R. H. Bieri, E. M. Shoemaker, and S. O. Thompson for providing data in advance of publication. This review includes results published or available through the end of April 1967.

Poisonous Principles of Mushrooms of the Genus Amanita

Four-carbon amines acting on the central nervous system and cell-destroying cyclic peptides are produced.

Theodor Wieland

The true poisonous mushrooms are of the genus *Amanita*. The best known of them, widely spread throughout the world, except for the tropic zones, is undoubtedly the fly agaric, *Amanita muscaria* (L. ex Fr.) Pers. ex Gray; its toxicity, however, is generally overestimated. Much more capable of inciting fatal poisoning is the white mushroom *A. verna* (Bull. ex Fr.) Pers. ex Vitt. sensu Fr. 1821 non al. (*A. virosa* Lam. ex Secr.). *Amanita tenuifolia* (*1*), and *A. bisporigera* Atk. (*2*) are probably closely related varieties. In Central Europe, the green death-cap *A. phalloides* (Vaill. ex Fr.) Secr., the *grüne Knollenblätterpilz*, is noted for its toxicity. The more frequently occurring yellow mushroom *A. citrina* (Schaeff.) Gray [*A. mappa* (Batsch ex Lasch) Quél.] definitely contains no peptidic toxins—unlike *A. phalloides* in this respect—but it does contain bufotenine (5-hydroxy-*N*-dimethyltryptamine) in relatively high concentration (*3*) and some other indole amines (*4*). It is of some interest that bufotenine occurs also in several other plants, and that other components of poisonous amanitas have been found also in mushrooms of different *Galerina* species (*5*). I shall outline the present state of knowledge of the components of *A. muscaria* and give a summary of the chemistry and toxicology of the poisonous peptides of *A. phalloides*.

Amanita muscaria (*6*), the fly agaric, grows from July until the end of autumn, preferably under fir or birch trees, as an egg-shaped white cap. Within 1 to 2 days the cap spreads and bursts open the outer shell, the surface then becoming brilliant red with the residue which remains as white spots regularly distributed over it. The white veil protecting the lamellae in the young mushroom now hangs at the stem as a cuticular ring. The stem, which grows up to 25 centimeters, has a cup at its bottom. The cap may reach 25 centimeters in diameter; the weight of an average mushroom is 60 to 70 grams. Different subspecies of varying colors, like the native North American one that is orange yellow, have been described.

The name "fly" agaric was derived from an insect-killing property of its extracts; however, this characteristic is inconspicuous compared to modern insecticides and can only be observed under special conditions. Wasson (*7*) interprets the "flies" as a symbol for the demonic power of the mushroom, which has been used in Siberia as a hallucinogen because of its psychotropic principles. The symptoms of intoxication from the use of *A. muscaria* are very complex and resemble those of drugs that act on the central nervous system, but with the addition of associated peripheral phenomena attributable to muscarine.

Problem of Muscarine

The search for the toxic principle of *Amanita muscaria* started over 100 years ago and led Schmiedeberg and Koppe in 1869 to a substance that excites the parasympathetic nervous system. This substance they named "muscarin" (*8*, *9*). The sensation caused among pharmacologists by the first

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