lion km and beyond), and the roundoff error is proportionately larger. Of course, solutions by the weighted-least-squares method depend on the relative weighting of the range and Doppler data. The total RMS residual divided by the assumed error weight is about the same for both types of data (Table 2), which indicates that the relative weighting is reasonably correct.

In the future, the computation of the range data will be accomplished in extended precision, and the error weight will be reduced to the level of the measurement error. Also, the data for cruise and encounter will be combined in a single solution to obtain important information on the astronomical unit and certain elements of Earth's orbit. At present the best that can be said is that the radar value of the astronomical unit (149,597,890 km) is consistent with the Mariner V data, and that the cruise solutions accomplished to date (those which include corrections to Earth's orbital elements) indicate that significant corrections on the order of a 0.1 second of arc are needed to fit the data. One correction in particular requires that the instantaneous axis of revolution of Earth about Sun be moved by at least ½ second of arc toward Earth's axis of rotation to produce residuals as small as those indicated by Table 2.

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Aluminum-26 and Beryllium-10 in Greenland Ice

Abstract. *Activities of beryllium-10 and aluminum-26 dissolved in 200-year-old Greenland ice were found to be* $18.4\ (+\ 8.4,\ -\ 4.8) \times 10^{-6}$ *and* $3.2 \pm 0.9 \times 10^{-7}$ *disintegration per minute per liter, respectively. From these values and the precipitation rate (30 milliliters of water per square centimeter per year), the production rates of these isotopes are calculated to be* $3.6\ (+\ 1.6,\ -\ 0.9) \times 10^{-2}$ *and* $1.7 \pm 0.5 \times 10^{-4}$ *atom per second · square centimeter. These rates agree with the rates calculated for the production of these isotopes by cosmic rays in the atmosphere. Probably all the* Al^{26} *in the ice is accounted for by such atmospheric production; however, an upper limit for the influx of cosmic dust bearing aluminum-26 is calculated at* 3.2×10^{5} *tons per year for Earth. Only upper limits could be found for* Al^{26} *and* Be^{10} *in the undissolved particulate matter in the ice; their addition to the activities in the dissolved material leaves our conclusions unchanged.*

The nature and flux of extraterrestrial material accreted by Earth are highly uncertain. It has often been stated, but never proved, that the bulk of this material is in the form of micron-size particles from the interplanetary dust cloud. If extraterrestrial material is accreted uniformly over Earth's surface, sediments that accumulate at the lowest rate should contain the largest extraterrestrial component. It is clear, however (Table 1), that even the most slowly accumulating sediments are predominantly terrestrial. The problem of measuring and characterizing the extraterrestrial component is therefore not trivial.

If sufficient amounts of a sediment sufficiently rich in cosmic dust are available, isotope anomalies and radioisotopes produced by cosmic rays may be found in it (*1*) as they are in meteorites. Dust from Greenland ice and fractions of deep-sea sediments have yielded inert gases showing anomalous isotope ratios (*2*), and claims have been made for measurable amounts of Al^{26} produced by cosmic rays in deep-sea sediments (*3, 4*). However Al^{26} was not detected in the dust from Greenland ice that yielded the anomalous inert gases (*5*). This discrepancy is important. Anomalies in rare-gas isotopes may be explained by the presence of diverse types of extraterrestrial material (micron-size dust grains, cometary or asteroidal material entering the atmosphere as large chunks, or even organic material), but Al^{26}, if present, sets severe limits on the type of extraterrestrial material that can be hypothesized (*6*); it also provides a measure of the flux of extraterrestrial material reaching Earth, which the anomalies in rare-gas isotopes can do only by elaborate theoretical models (*7*).

Some Al^{26} is produced by cosmic rays in Earth's atmosphere, but the activities found in sediments by Lal and Venkatavaradan (*3*) and Wasson (*4*) are greater than would be expected from this source.

The search for Al^{26} in Greenland ice was incomplete in that only the undissolved particulate material [chiefly clay minerals and an unidentified organic component (*8*)] was collected and examined. There are many reasons for expecting that the bulk of the extraterrestrial material in the water from the ice is dissolved: extraterrestrial material may reach the ice as finely divided and readily soluble dust or smoke, the water is very pure and slightly acidic [total dissolved material, less than 1 part per million (ppm), *p*H 4.5], and the chemistry of the dissolved material in the ice indicates the presence of extraterrestrial material (*8, 9*). In any case, radioisotopes produced by cosmic rays in Earth's atmosphere may be expected to be dissolved in the ice, and a measurement of these would be of interest. We now report the measurement of Al^{26} and Be^{10} dissolved in Greenland ice; we also describe an attempt to measure Be^{10} and further attempts to find Al^{26} in the particulates. We find these isotopes only in the amounts expected from their production by cosmic rays in the atmosphere.

The facilities and methods for collecting particulate material from 100- to 300-year-old ice at Camp Century, Greenland, have been described (*5*). In our work, columns containing cation-exchange or mixed cation- and anion-exchange resins were inserted into the waterline. Later the resins were removed from the columns and homogenized, and the cation resins were leached with 4*M* HCl; the solutions so obtained have been subjected to extensive chemical study that will be reported elsewhere (*8*).

For the search for Al^{26} and Be^{10}, a solution obtained by leaching resins through which 1.2×10^{6} liters of ice water had passed was used, and carriers for Be, Ti, Mn, and Co were added. The elements were then separated by standard methods of analytical chemistry. Final purification of the aluminum, chiefly to remove thorium and its decay products, was accomplished by solvent extraction with cupferron (*4, 10*). Beryllium was purified by repeatedly precipitating it as the

Table 1. Percentages of extraterrestrial material expected in slowly accumulating sediments; an influx rate of 10^6 ton/year over the entire surface of Earth is assumed.

Sample	Input data	Sedimentation rate ($g/cm^2 \cdot yr$)	Extra-terrestrial material (%)
Sea sediment	1 mm/10^3 yr; ρ, 0.5 g/cm^3	5×10^{-5}	0.4
Particulates in Greenland ice	6×10^{-5} g/liter	1.8×10^{-6}	9.2
Dissolved in Greenland ice	4.3×10^{-4} g/liter	1.3×10^{-5}	1.5
Extraterrestrial material (assumed)		1.96×10^{-7}	100

hydroxide in the presence of ethylenediaminetetraacetic acid; Teflon laboratory ware was used.

From this solution 9.3 g of Al_2O_3 was recovered. Analysis of the ice water showed it to contain 0.017 ppm of aluminum; hence the overall chemical yield for aluminum was 25 percent. From the 111 mg of carrier added, 32.1 mg of BeO was recovered; thus the chemical yield for beryllium in this step was 30 percent. The beryllium content of the original water was too low to be measured; thus the overall chemical yield can only be estimated. The efficiency with which the resin column removed beryllium from the water was estimated to be 70 ± 20 percent from its efficiency for sodium, potassium, calcium, magnesium, and aluminum (*8*); on this basis the overall chemical yield for beryllium was 21 ± 6 percent.

The Be^{10} beta activity was counted with a flow counter having a 12.5-mm-diameter, 0.90-mg/cm^2 Mylar window. This counter has a background of 0.042 count/min when used in a low-level counting system consisting of a ring of anticoincidence counters surrounded by an iron shield 15 cm in thickness. The BeO from the resin-leach solution gave 0.446 ± 0.007 count/min; this activity had the same absorption curve as a Be^{10} standard. The half-absorption thickness in aluminum, found for the sample activity, was 30 ± 1.5 mg/cm^2, compared to 21 ± 0.3 mg/cm^2 found elsewhere (*11*) for Be^{10} and 25 ± 3 mg/cm^2 found by us for the Be^{10} standard. The BeO blanks, prepared in the same way as was the BeO from the resin-leach solution, showed no activity above background. The counting efficiency of the counter, determined by use of a Be^{10} standard of the same mass as the sample, proved to be 7.9 percent.

From the Be^{10} activity found and the overall chemical yield estimated as we have explained we calculate the Be^{10} activity in the ice to be 18.4 (+8.4, −4.8) $\times 10^{-6}$ disintegration per minute (dpm) per liter. The precipitation rate at Camp Century is well known from stratigraphic studies and Pb^{210} dating (*12*); the average over the last 150 years is about 30 ± 2 cm of water per year. Thus the precipitation rate of Be^{10} is 5.5 (+2.5, −1.4) $\times 10^{-7}$ dpm/$cm^2 \cdot$ year. This figure gives a production rate of 3.6 (+1.6, −0.9) $\times 10^{-2}$ atom/$cm^2 \cdot$ year. Lal (*13*) calculated an average global production rate of 4.5×10^{-2} atom/$cm^2 \cdot$ sec for Be^{10}.

The Al_2O_3 from the resin-leach solution was counted by gamma-gamma coincidence method using 7.5 × 7.5-cm and 5 × 5-cm NaI (thallium activated) crystals in a low-level shield (*5*). The sample was alternated every few days with an Al_2O_3 blank or a crushed sample of the Bruderheim meteorite of the same mass and geometry as the sample. As the Al^{26} activity in Bruderheim is known, the sample of it served as a standard for calculation of the counting efficiency of the crystals. The positron activity in the sample was found to be 0.093 ± 0.030 dpm with the 7.5 × 7.5-cm crystals, and 0.071 ± 0.030 dpm with the 5 × 5-cm crystals; the average is 0.082 ± 0.022 dpm. The Al_2O_3 from the resin-leach solution was also counted for Al^{26} by Wogman (*14*); his result, 0.06 ± 0.03 dpm, based on five 1000-minute counts and nine 1000-minute counts of background, agrees with our value.

From the activity in the sample and the overall chemical yield we calculate the Al^{26} activity in the ice to be $3.2 \pm 0.9 \times 10^{-7}$ dpm/liter; this figure corresponds to a precipitation rate of $0.96 \pm 0.27 \times 10^{-8}$ dpm (of Al^{26})/$cm^2 \cdot$ year, or a production rate of $1.7 \pm 0.05 \times 10^{-4}$ (Al^{26}) atom/$cm^2 \cdot$ sec.

In the atmosphere, Al^{26} is produced by cosmic-ray spallation of argon. The estimate for the cross section for this reaction is only good to about a factor of 3. On this basis Lal (*15*) has estimated the production rate of Al^{26} in the atmosphere to be 1.4×10^{-4} atom/$cm^2 \cdot$ sec, which rate agrees with our result.

After further attempts to measure Al^{26} in the particulate material from Greenland ice we have greatly reduced our previous (*5*) upper limit [1.3×10^{-8} dpm (Al^{26})/$cm^2 \cdot$ year, which is lower by a factor of 3 than that found from sea sediments (Table 2). Wasson *et al.* (*4*) made an arithmetic error when they said that their sea-sediment value differed from this by a factor of 30.] The upper limit for Al^{26} precipitation in particulate material at Camp Century now stands at 0.6×10^{-8} dpm/$cm^2 \cdot$ year. We also sought Be^{10} in 15.6 g of magnetic material that settled from roughly 2.5×10^5 liters of ice water. However, the activity found, 0.028 ± 0.005 dpm, is very uncertain as it is insufficient for determination of an absorption curve.

The surprising feature of these results is that we find no evidence of Al^{26} borne by cosmic dust. Even if we add the upper limit for Al^{26} precipitation in the particulates to that found from the dissolved Al^{26}, the result, $1.6 \pm 0.3 \times 10^{-8}$ dpm/$cm^2 \cdot$ year, could still be accounted for by production by cosmic rays in the atmosphere. Since the production rate in the atmosphere is as yet uncertain, we can-

Table 2. Gamma-gamma β^+ coincidences for particulates.

Sample weight (g)	Collection method	Water well formed by	H_2O processed (liter)	Particle-collection efficiency (%)	β^+ Decays per Minute	β^+ Decays per Minute per liter ($\times 10^{-7}$)	Al^{26} (10^{-8} dpm/$cm^2 \cdot$ yr)
37	Millipore filters	Steam	6×10^5	100	<0.1	<1.7	<0.6
330	Settling tanks	Steam	$\sim 5 \times 10^7$	~10	<1	$\lesssim 2$	$\lesssim 1.0$
20.85	Cation resin	Heat exchanger	2.4×10^6	50	<0.2	<1.7	<0.6
41	Millipore filters	Heat exchanger	1.9×10^6	100	<1	<5.3	<1.9
29.7	Cation and mixed resin	Heat exchanger	2.4×10^6	70	≤ 0.13	≤ 0.8	≤ 0.3

not rule out the possibility that some of the Al^{26} we find reached the ice in cosmic dust; nonetheless, we find a much lower precipitation rate of Al^{26} than do Wasson *et al.* and Lal *et al.*

In the work of Lal *et al.* (*3*) and Wasson *et al.* (*4*), positron activity, interpreted as that of Al^{26}, is measured in aluminum separated from pelagic red clays. This activity and the measured or estimated accumulation rate of the sediment yield an Al^{26} precipitation rate five (*4*) or ten (*3*) times that expected from atmospheric production; the "excess" is assumed to be Al^{26} from cosmic dust. We believe that the difference between these results and ours can be explained entirely by the uncertainties in their measurements. The measurements by Lal *et al.* are borderline; the positron peak they found was little above noise level and the same peak appeared in their background. Wasson *et al.* did not recycle their aluminum to see whether the positron activity followed aluminum chemistry, and the sedimentation rate they used in their calculations was an estimate. In our measurements we cannot think of any likely mechanism by which Al^{26} could have been lost, and further measurements are more likely to cause us to lower our estimate of the rate of Al^{26} precipitation than to raise it.

We can set an upper limit to the influx rate of cosmic dust bearing Al^{26} by combining our upper limit for Al^{26} precipitation with the most likely value for its fallout rate from atmospheric production. This gives $(1.6 - 0.8)10^{-8} = (0.8)10^{-8}$ dpm/cm^2 · year for Al^{26} in cosmic dust. From this figure, the assumptions of Lal *et al.* give an upper limit for the influx rate of zodiacal dust over Earth of 3.2×10^5 ton/year, and the assumptions of Wasson *et al.* give an upper limit of 1.0×10^5 ton/year. However, we emphasize that these numbers have little intrinsic value: not only are they merely upper limits; a great many very weak assumptions have been needed to turn an observed Al^{26} precipitation rate into a cosmic-dust accretion rate.

Our Be^{10} measurements are in good agreement both with the calculated atmospheric production rates and with those found from measurements of Be^{10} in sea sediments. However, to derive an average production rate of Be^{10} from our measurements, we would need to know the effect of latitude and of the nature of the precipitation (rain or snow) on the precipitation rate of isotopes from the atmosphere. This point is in some doubt, and we therefore prefer to let our results stand without further interpretation.

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Fallout from the Chinese Nuclear Explosion of 17 June 1967

Abstract. *Low-level activities of iodine-131, barium-140, and strontium-89 were found in a series of rain samples collected at Fayetteville, Arkansas, during the period from late June through August 1967. The ratios of these short-lived isotopes to strontium-90 were determined as accurately as possible. The data indicate that the debris from the Chinese nuclear explosion was injected primarily into the stratosphere.*

It was announced that a thermonuclear weapon was tested in China on 17 June 1967. Heavy fallout was expected soon after, but the preliminary indications were that the fallout was almost negligible. This indication suggested the possibility that the debris was injected primarily into the stratosphere, and that it would be some time before the fallout reached ground level. We studied the fallout of I^{131}, Ba^{140}, and Sr^{89} from this explosion at Fayetteville (94°W, 36°N), Arkansas, using large volumes of rain samples and attempting to measure the ratios of a number of fission products to Sr^{90} as accurately as possible. We now report the data obtained so far.

The rain samples were collected on the roof of the Chemistry Building of the University of Arkansas. Strontium and Ba carriers were added to 8 to 16 liters of rain samples, and the Sr and Ba fractions were radiochemically purified by a reported method (*1*, *2*). Carrier of I^- was added to 12 liters of rain samples, and the iodine fraction was radiochemically purified by a reported method (*3*). The procedure used is briefly described: the I^- was oxidized to IO_4^- with NaClO in alkaline solution; IO_4^- was then reduced to I_2 with hydroxylamine hydrochloride, and the iodine was extracted with CCl_4; the iodine was removed from the CCl_4 by shaking with $NaHSO_3$, and then was purified by another CCl_4 extraction cycle in which $NaNO_2$ was used for oxidation of I^- to I_2; this extraction cycle was repeated three times; the iodine was finally precipitated as AgI and counted. Tracerlab Omni Guard low-level background system was used for the radioactive measurements, with a background of about 0.5 count/min.

The results appear in Table 1. Presence of I^{131} and Ba^{140} was first detected in the rain sample collected on 27