Reports

Aluminum-26 in Pacific Sediment: Implications

Abstract. *Aluminum-26 has been detected in a sample of sediment from the South Pacific. The disintegration rate of 0.8 disintegration per minute per kilogram of dry sediment is considerably higher than that expected from cosmic-ray spallation of atmospheric argon; it appears to result mainly from accretion of activity induced in interplanetary dust by solar-flare particles. This finding is in keeping with Wasson's published estimates regarding the magnitude of this effect, and confirms the order-of-magnitude correctness of the solar-particle flux and terrestrial accretion rate of interplanetary dust used in that calculation.*

The bulk of extraterrestrial matter accreted to Earth is in the form of micron-sized particles from the interplanetary dust cloud; very little is known about it. Estimates of the total-accretion rate range from about 10^{-6} to 10^{-8} g cm^{-2} year^{-1} (5×10^4 to 5×10^6 ton/year over the entire surface). There is no direct evidence of its chemical composition. Various investigations have been made regarding its size distribution, density, and other physical properties. A certain fraction of the dust is volatilized by interaction with Earth's atmosphere; according to Öpik (*1*) 50 percent of the total mass survives as relatively intact particles and falls to Earth's surface within a few days or weeks. Separation of these particles from sea sediments and polar-ice samples has been attempted, and there is evidence that extraterrestrial metallic particles have been successfully isolated. Meteoritic-fall statistics, however, suggest that silicate particles should be considerably more abundant than metallic ones, but very little progress has been made toward distinguishing between these objects and terrestrial contaminants.

In 1963 Wasson (*2*) proposed that the radioactivity inherent in the dust might allow indirect attack on some of the problems. He showed that solar-flare particles should be considerably more important than cosmic rays in inducing radioactivity in the dust, and he made estimates regarding the steady-state decay rates of several long-lived dust radionuclides that may be detectable at Earth's surface. These calculations are based on a composition similar to that of chondritic meteorites, a solar-proton spectrum similar to average spectra observed during the past maximum in solar activity, and an average solar-proton flux ($E > 5$ Mev) of 1000 cm^{-2} sec^{-1} at 1 astronomic unit from Sun.

The two best sources of large quantities of interplanetary dust are the polar ice caps (especially central Antarctica) and sediments from the central ocean basins (especially the South Pacific); their advantage is isolation from sources of air-borne and water-borne terrestrial "contaminants." Because our original plans called for the counting of several hundred grams of Al, we obtained a large (about 45-kg) sample of sediment (*3*), designated V-18-BBD-1, dredged on 26 November 1959 from the floor of the Pacific at 14°S, 155°W at a depth of 2660 fathoms (4860 m); it consisted chiefly of red-brown lutite. The depth of the sample below the surface of the sediment is unknown, but it could scarcely have exceeded 20 cm because of the nature of the sampling device. Sackett's (*4*) attempts to estimate the "age" of the sample by measuring the activities Pa^{231}, Th^{230}, and Th^{232} gave ambiguous results. Deposition rates in this part of the Pacific range from 4 to 40 $\times$ 10^{-5} cm/year (*4, 5*), and we can conclude that the Al^{26} in our sample has decayed at most by a factor of 2. Because the sample was collected after the start of nuclear-weapons testing, it may possibly contain bomb-produced positron emitters. The integrated production of Al^{26} and Ti^{44} should be negligible compared to natural sources, however, and our chemical procedure discriminates strongly against Na^{22}.

The sample was chemically treated (*6*) for two reasons: (i) to increase the specific activity of Al^{26}; and (ii) to remove radioactive contamination due to K^{40}, Ra^{226}, and Th^{232}. We broadly describe the procedure: The wet sample was passed through a 400-mesh sieve. The Al content was determined from a portion to be 7.6 percent on a dry-weight basis. The sample was leached at room temperature with 6*N* HCl for 16 hours and with 4*N* HCl for 6 hours, and the leached extracts were combined. These "cold" extracts removed 61 percent of the total Al. The residue was leached at 85°C with 12*N* HCl for 16 hours, with the removal of an additional 28 percent of the Al. The cold- and hot-leached fractions were then

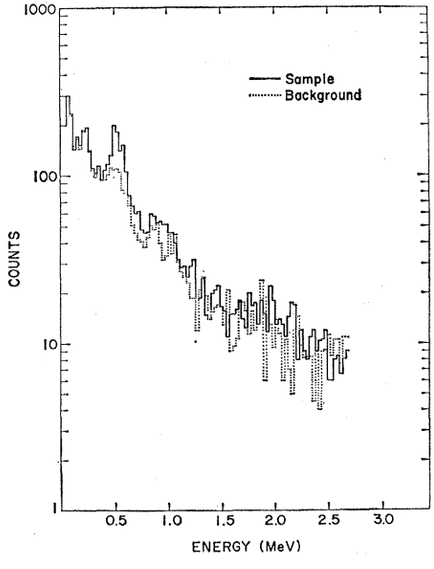

Fig. 1. Spectra of sample and background for 11650 min.

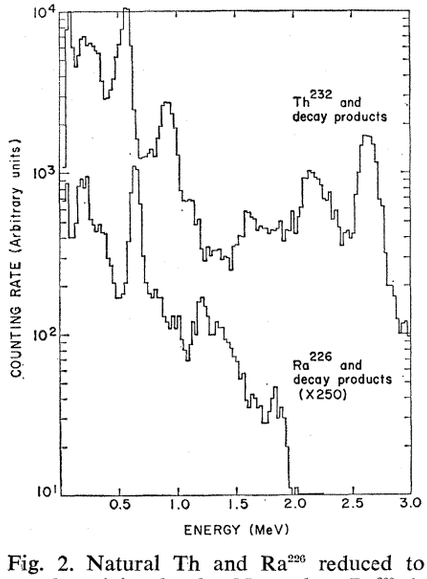

Fig. 2. Natural Th and Ra^{226} reduced to equal activity levels. Note that Ra^{226} is multiplied by a factor of 250.

Table 1. Results of measurement of a sample of Al from Pacific sediment. See text for details.

Energy range (Mev)	Count rate (cpm)			Efficiency (%)	Activity (dpm)
	Sample	Background	Net		
0.51 ± 0.035	0.0705 ± 0.0019	0.0526 ± 0.0020	0.0179 ± 0.0027	1.75	1.02 ± 0.15
1.83 ± .080	.0180 ± .0011	.0146 ± .0010	.0034 ± .0016	0.17	2.0 ± .9
2.34 ± .080	.0104 ± .0009	.0084 ± .0009	.0020 ± .0013	.17	1.2 ± .8

purified separately as follows: Aluminum (as well as other elements, especially Fe) was precipitated as the hydroxide with NH_4OH, and the precipitate was washed with hot dilute NH_4Cl solution. The precipitate was dissolved in 12*N* HCl, and Fe was removed by extraction three times with LA-1, a decanol-kerosene mixture. Thorium was removed by solvent extraction with ethyl acetate saturated with cupferron.

The aqueous layer was saturated with HCl at 7°C, and the Al was recovered as $AlCl_3 \cdot 6\ H_2O$. The precipitation step was repeated twice for additional purification, and the final precipitate was ignited to Al_2O_3 at 400°C. The oxide was mixed with charcoal, and the Al was converted to the anhydrous chloride with Cl_2 at advanced temperatures terminating at 1000°C. Metallic Na was added under an atmosphere of Ar in order to reduce Al to the metal.

Unfortunately, the Na reduction was only partially successful, and the Al metal was formed as very fine particles. In an attempt to make consolidated metal by the electrolysis of a mixture of AlF_3 and NaF, most of the Al was lost "up the stack," probably as AlF_3. The final recovery of metal amounted to 57.5 g; it was estimated to be 90 percent from the cold leach and 10 percent from the hot. Thus the recovered Al was somewhat less than 10 percent of that leached from the clay.

Aluminum-26 decays by positron emission (85 percent) and by emission of a 1.83-Mev gamma-ray (96 percent). These properties make its detection possible by either of two highly specific methods, both based on scintillation spectroscopy. The first entails the counting of large samples on a large NaI detector (*7*). The second type of counting array has two detectors operated in coincidence to detect the annihilation quanta from the positron decay; it was proposed by Anders (*8*). Our system was of this type and had been used (*9*). It consisted of two 4- by 4-inch (10- by 10-cm) NaI crystals. Detector 1 was connected to a single-channel analyzer selecting pulses within 0.035 Mev of 0.510 Mev; detector 2, to a TMC 256-channel pulse-height analyzer. The total spectrum and the coincidence spectrum were each sorted simultaneously into separate groups of 128 channels ranging from 0 to 3.4-Mev energy. The sample was roughly cylindrical in shape, 4.0 cm in diameter and 2.5 cm thick. An annular ring of high-purity Bi surrounding the sample reduced background.

For the background measurements a portion of pure Al of identical mass and similar shape was mounted between the crystals; another similar portion was cut into four pieces, between which three Mylar foils, coated with Al^{26} standard, were interspersed. The efficiency of the counting system was determined by measurements of this phantom.

Four counting runs were successful; a fifth had to be discarded because of a gain shift. In each run the sample and the background portion were measured alternately, and periodically the efficiency was checked; the sample was measured for 20150 minutes; the background, for 30050 minutes.

The counting rates of the four individual runs agree with the average counting rate within 1 σ. The sum-spectra of the first three runs for sample and background are plotted in Fig. 1. The fourth run yielded similar spectra, but because of different adjustment of the multichannel-analyzer they cannot be simply added to Fig. 1. Table 1 shows the final numerical results of the four runs. From the annihilation peak, an apparent Al^{26} activity of 1.02 ± 0.15 dpm is obtained. The low counting efficiency for the typical γ-ray of 1.83 Mev, as well as for the sum-peak, does not allow a clear identification of Al^{26}. In order to estimate possible contamination by natural Th and U, we measured natural Th and Ra^{226} under the same conditions as for the sample. The coincidence spectra are plotted in Fig. 2. For Th, typical peaks appear at 0.58, 0.92, 1.60, and 2.60 Mev; for Ra^{226}, at 0.61, 1.13, and 1.76 Mev. Table 2 shows that the net effects in these energy ranges are low. With the relative peak heights, contamination by U^{238}-Ra^{226} can be excluded, whereas about 10 percent of the 0.51-Mev sample peak seems to be due to Th contamination. The corrected decay rate is 0.91 dpm.

In order to relate this content of Al^{26} to various possible sources, one must know the magnitudes of a number of additional parameters. We have already stated that 90 percent of our 57.5-g sample came from the cold leaching, and that 61 percent of the Al (or 46.2 g of Al per dry kilogram of sediment) occurs in this fraction. We will assume that all natural Al^{26} is extracted by the cold leaching. Our Al^{26} activity is 0.81 dpm/kg. Amin *et al.* (*10*) find 0.5 to be a typical density of sediment, expressed as dry weight divided by *in situ* volume, and we shall assume a sedimentation rate at our sampling site of 10^{-4} cm/year, which should be accurate within a factor of 2 (*4, 5*). We will assume that decay of Al^{26} since deposition has been negligible. Combining these parameters with the observed decay rate in our sample, we calculate a production rate for Al^{26} of 0.40×10^{-7} dpm cm^{-2} year^{-1}. The calculated rates from interplanetary dust (*2*) and spallation of atmospheric argon (*11*) are 1.2 and 0.08×10^{-7} dpm cm^{-2} year^{-1}, respectively. The dust value is based on an assumed accretion rate of 10^{-7} g cm^{-2} year^{-1} and an assumed undersaturation of the Al^{26} in the dust by a factor of 10.

Thus the observed production rate is five times as high as that expected from spallation of atmospheric argon. If we correct for this source, the observed "dust" rate is lower by a factor of 4 than the previous estimate—excellent agreement when one considers all the uncertainties involved. The magnitude of the observed production rate is not so great that one can completely rule out the possibility that argon spallation is responsible for all the ob-

Table 2. Net counting rates of the Al sample in energy ranges typical for natural Th. Negl, negligible.

Typical energy (Mev)	Net count rate (cpm)	Est. contrib. to 0.51 peak (cpm)
0.58	0.0102 ± 0.0021	0.002
.91	.0024 ± .0017	Negl
1.60	.0010 ± .0011	Negl
2.60	.0024 ± .0010	Negl

served activity; this possibility does not seem likely, but more measurements are clearly in order.

Our results should be compared with those of Lal and co-workers (*10, 11*), who have independently reported the detection of Al^{26} in two samples of marine sediments, and with those of Fireman and Langway (*12*), who did not detect Al^{26} in particles filtered from melted Greenland ice. The former workers report an Al^{26} activity in their (dry) sediments of 0.46 dpm/kg, whereas our data yield a value of 0.81 dpm/kg. This difference could easily result from the sedimentation rates of their samples being twice the rate of our sample, or from the possibility that Al^{26} had decayed by a larger factor in their samples than in ours because their samples came from deeper within the sediment. The upper limit of Fireman and Langway, 4×10^{-7} dpm/liter^{-1}, corresponds to a production rate of Al^{26} of 1.2×10^{-8} dpm cm^{-2} year^{-1} (30 times smaller than our value). The difference could result if 97 percent of the Al^{26} were in a form that passed through the 3-μ pores of the filter used by these workers. Certainly, the 50 percent of the dust that may be volatilized (*1*) will result in soluble Al compounds, and a fairly large fraction of the remaining Al^{26} will reside in minerals that are easily soluble, or from which the Al^{26} can be easily leached by the action of H_2O. In addition, some large particles may have been lost by settling before filtration.

Lal and Venkatavaradan (*11*) have attempted to use their measurement of Al^{26} to set limits on the flux of solar protons during the last 10^5 years. Clearly our result can be used similarly, though we do so with some trepidation because of the uncertainties involved. The greatest uncertainty in the production rates probably resides in the values for the sedimentation rates, which may be incorrect by as much as a factor of 2 in either our sample or theirs (*13*). The influx of interplanetary dust is uncertain by at least an order of magnitude (*14*). The shrinkage times of the heliocentric dust-particle orbits may be shorter than previous assumptions; they depend on two unkowns: (i) the size distribution of the dust, and (ii) drag forces other than those due to solar electromagnetic radiation.

Perhaps the safest solution is to note that the apparent production rate due to dust is lower by a factor of 4 than that calculated by Wasson, and to reduce his assumed proton flux by the same factor to 250 cm^{-2} sec^{-1}, while keeping the assumed rate of dust influx of 10^{-7} g cm^{-2} year. These quantities should be within an order of magnitude of the correct values.

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Search for 21-Centimeter Radiation near Cosmic X-Ray Sources

Abstract. *A search was made for 21-centimeter wavelength radiation in the regions of seven of the cosmic x-ray sources. No new sources with flux densities greater than 5.5 flux units were found.*

The location of various cosmic x-ray sources have been reported by Giacconi *et al.* (*1*), Bowyer *et al.* (*2*), and by Fisher *et al.* (*3*). Positions (within 1.5°) and relative intensities of ten x-ray sources have been determined by Bowyer and his co-workers (*2*). Of the sources in this list, Tau XR-1 seemed to be associated with the Crab nebula; Oph XR-1, with Kepler's supernova (SN); and Sgr XR-1, possibly associated with the galactic center. The remaining sources did not seem to be associated with optical or radio objects. In January 1965 a search was made for 21-cm continuum radiation in the regions of seven of the x-ray sources in the list of Bowyer *et al.* (*2*) to determine if there were radio sources, previously undetected, at the positions of the x-ray sources. (The search was made at 21-cm because of the availability of equipment at this wavelength.) Table 1 lists the x-ray sources studied here; Taurus XR-1 was not included in the survey because of its identification with the Crab nebula; and Sco XR-2 and Sco XR-3 were too far south in declination to be observed. No new sources were found in our initial scans of the regions, and, because of the limited time available for this experiment, it was decided to discontinue the search until better positions for the x-ray sources and more time for the search were available.

More recent rocket experiments have shown, however, that Oph XR-1 does not exist at present, that Sgr XR-1 is at a position different from that originally determined, and that Sgr XR-2 is possibly variable (*4*). Because x-ray sources may be variable, it now seems that the 21-cm measurements made in January 1965 should be described.

Observations were made with the 84-foot parabolic reflector of the Naval Research Laboratory, located at the Maryland Point Observatory. The radiometer, which has been described previously (*5*), has a center frequency of 1414 Mc/sec and a bandwidth of 3.3 Mc. With a 6-second integration time, the peak-to-peak noise is 0.5 degree Kelvin. The antenna efficiency is 45 percent (*6*), which results in a minimum detectable flux density of 5.5 flux units [1 flux unit is 10^{-26} watt m^{-2} (cy/sec)$^{-1}$] for a source small with respect to the 36-minute-of-arc beam. This value for limiting flux density was determined by assuming that a source giving a deflection equal to the peak-to-peak noise could be detected in a single drift curve. For a source of size comparable to or larger than the beam, the minimum detectable flux density would be greater than 5.5 flux units.

Observations consisted of drift curves in right ascension, centered on the right ascension of the x-ray sources (*2*) and taken at intervals of 18 minutes of arc in declination (half beamwidth.) The range over which the search was carried out and the known radio sources