

Note added in proof: We have recently found evidence with Cocksackie B₁ virus and mengovirus that all or nearly all viral proteins are cleaved from precursor protein molecules of high molecular weight (10). This probably explains the constant ratio of viral gene products observed here.

JOHN J. HOLLAND

Department of Molecular and Cell Biology, University of California, Irvine 92664

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Aluminum-26 and Beryllium-10 in Marine Sediment

Abstract. Activities of aluminum-26 and beryllium-10 in marine sediment were measured at 0.01 ± 0.13 and 4.4 ± 0.9 disintegrations per minute, per kilogram dry weight, respectively. Only an upper limit of 0.03 could be determined for the ratio of aluminum-26 to beryllium-10 in the sediment. The ratio is probably explained by production by cosmic rays in the atmosphere.

Pelagic sediment is one of the best sources of information on radioactivities produced by cosmic rays. Generally two types of processes are expected to cause accumulation of long-lived radioactive products in marine sediment: (i) precipitation of spallation products resulting from nuclear interactions of high-energy galactic cosmic rays with atmospheric substances; and (ii) sedimentation of cosmic dust, when radioactivities induced by low-energy solar protons may be expected. It has been proposed that simultaneous pairs of measurements of Al²⁶ and Be¹⁰ would be an effective way to investigate these processes (1).

Lal and Peters (2) have estimated the global rates of production of Al²⁶ and Be¹⁰ by spallation of atmospheric constituents at 1.4×10^{-4} and 4.5×10^{-2} atom/cm², respectively; these values correspond to a ratio of about 0.01 for the specific activities of Al²⁶ and Be¹⁰ in marine sediment. The estimated production rate for Be¹⁰ generally accords with the observed occurrence of 1 to 10 dpm/kg (dry weight) (3) (dpm, disintegrations per minute). Wasson's estimate (4) of the Al²⁶ introduced to marine sediment by process (ii) suggests that this contribution to the content exceeds the contribution by

process (i) by more than an order of magnitude; a higher Al²⁶:Be¹⁰ ratio is expected by virtue of the fact that little Be¹⁰ can be produced by process (ii). However, there are still large ambiguities in his estimate because of the lack of accurate knowledge of both the rate of accretion to Earth of cosmic dust and the flux of solar protons.

Amin *et al.* (5) have reported the mean specific activities of Al²⁶ and Be¹⁰ in two cores from the Pacific basin at 0.5 and 4.0 dpm/kg (dry weight), which correspond to an average Al²⁶:Be¹⁰ ratio of 0.12. Wasson *et al.* (6) have reported an Al²⁶ content of 0.8 dpm/kg (dry weight). Both results imply that the Al²⁶ content is higher by about an order of magnitude than that expected from the spallation of

Table 1. Measurements of Be¹⁰ in four samples of sediment; the chemical yield of Be was determined to be 40 ± 5 percent by separate tracer experiments.

No.	Sample		Be ¹⁰ [dpm/kg (dry wt)]
	Depth in core (cm)	Dry clay(g)	
1A	0-23	156	6.1 ± 1.6
1B	23-47	182	3.7 ± 1.3
2A	47-73	132	4.4 ± 1.8
2B	73-100	144	3.5 ± 1.7

atmospheric argon. The high Al²⁶ content was taken to originate from cosmic dust accreted by Earth, and the necessary flux of solar protons and the accretion rate of cosmic dust were discussed (7). On the other hand, Yokoyama (8) says that the Al²⁶:Be¹⁰ ratio of around 0.1 can be explained as resulting only from spallation of atmospheric nuclei, in the light of new cross-sectional data from Bernas *et al.*

However, the important point is that there seem to be uncertainties in the earlier Al²⁶ measurements, resulting mainly from difficulties in obtaining a counting sample completely free from radioactive impurities. While investigating Al²⁶ and Be¹⁰ in marine sediment, we have obtained a value for Al²⁶ lower than the earlier results.

Our core sample of red clay was 7.8 cm in diameter and 1 m long; it came from a depth of 5439 m at 23°07'N, 135°45'E. The core was halved vertically, one half being available for our experiments. The sample was cut into four sections, each about 25 cm long. The Al₂O₃ content was vertically uniform throughout the core: 16.38 ± 0.32 percent (dry weight).

Dried clay was completely dissolved by treatment with HF and subsequent fusion of the residue with sodium carbonate. From the solution, Be and Al were separated and purified (9). The purified Be was deposited on a platinum disk, and Be¹⁰ was counted with a small flow-type Geiger counter. Because of the paucity of the sample and the rather high background of the counter, the statistical errors in the Be¹⁰ measurements (Table 1) are relatively high. However, the observed count rates accord with previously reported values. Sample 1A (Table 1) was subjected to absorption measurement with an aluminum absorber, which gave a half-thickness of 20 mg in fair agreement with the maximum energy of Be¹⁰.

The purified Al was finally converted to Al₂O₃, and Al²⁶ was counted with a low-level gamma-gamma coincidence spectrometer developed by us (10). The spectrometer comprises two quartz-faced NaI crystals, each 7.5 by 7.5 cm, positioned face to face. The coincidence spectrometry was performed with gating at 465 to 550 kev, and the counting efficiency for positrons was determined to be 2.8 percent.

Figure 1 illustrates the gamma-ray coincidence spectra of samples of Al₂O₃ after subtraction of background.

No peaks specific to Al^{26} (0.51 and 1.83 Mev) were observed. The numerical results for positron activities in samples of sediment appear in Table 2. The differences between count rates of sample and background were within statistical errors (1σ) of counting for the entire energy range analyzed; this finding proves that the counting samples were not contaminated with other radioactive impurities, and that selection of the background sample was quite adequate.

Our results are summarized in Table 3 along with earlier results. Only an upper limit of 0.03 was determined for the $\text{Al}^{26}:\text{Be}^{10}$ ratio, but this value is very important. Obviously the Al^{26} content in our sample is quite small in comparison with the earlier results. The present upper limit of the ratio is well accounted for by production by cosmic rays in the atmosphere, while the earlier higher Al^{26} contents, especially that of Wasson *et al.*, should rather be attributed to extraterrestrial origin.

Table 2. Measurements of Al^{26} in samples of sediment; cpm, counts per minute. The chemical yield of Al was 72.2 ± 2.8 percent. Background measurements were made from Al_2O_3 purified from a pure Al metal (99.999 percent).

Sample No.	Counting time (min)	Count rate at 465 to 550 kev ($\times 10^{-3}$ cpm)		Al^{26} [dpm/kg (dry wt)]
		Gross	Net	
1 (1A + 1B)	10921	27.00 ± 1.57	1.01 ± 2.01	0.14 ± 0.27
2 (2A + 2B)	14592	25.50 ± 1.32	-0.49 ± 1.81	$-0.08 \pm .32$
	16393	Background 25.99 ± 1.25		

Table 3. Summary of our results and those of others. Our sample was an average of samples 1 and 2 (Table 2).

Investigators	Disintegrations per minute, per kilogram (dry wt)		$\text{Al}^{26}:\text{Be}^{10}$
	Al^{26}	Be^{10}	
Tanaka <i>et al.</i>	0.01 ± 0.13	4.4 ± 0.9	≤ 0.03
Amin <i>et al.</i>	$.46 \pm .17$	$3.9 \pm .2$	$.12 \pm 0.04$
Wasson <i>et al.</i>	$.81 \pm .12$		

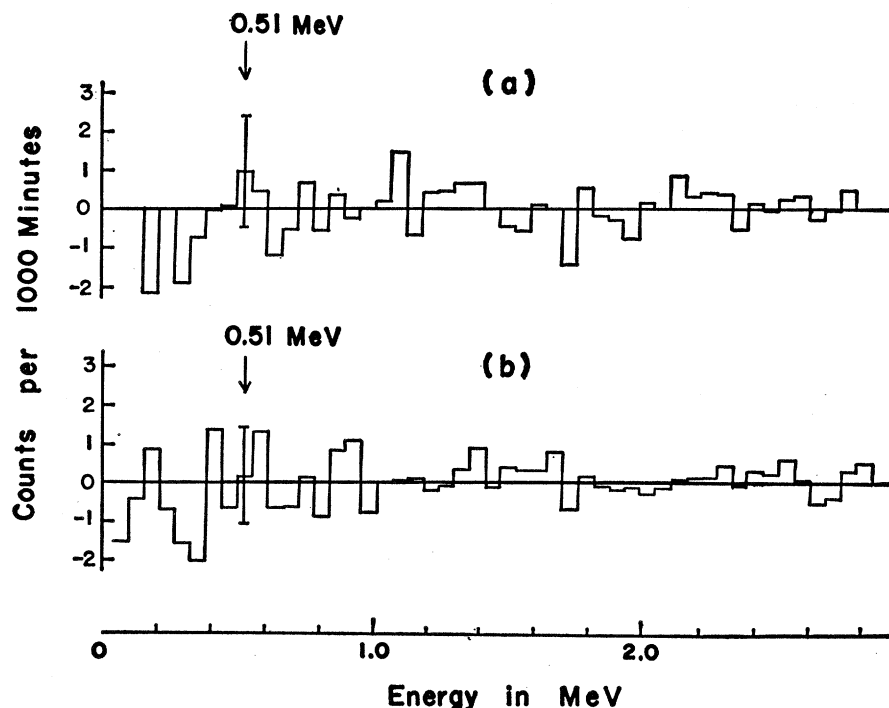


Fig. 1. Coincidence spectra (less background) of sample 1 (a) and sample 2 (b). Samples 1A and 1B (Table 1) were combined to form counting sample 1; 2A and 2B, to form sample 2. Total times of counting are indicated in Table 2. Errors represent statistical errors in counting.

Very recently McCorkell *et al.* (11) analyzed the contents of dissolved Al^{26} and Be^{10} in 200-year-old Greenland ice, determining their respective precipitation rates at 0.96×10^{-8} and 5.5×10^{-7} dpm/cm²·year, corresponding to an $\text{Al}^{26}:\text{Be}^{10}$ ratio of 0.017. According to them the upper limit of the ratio can be set at 0.03 if the contribution of Al^{26} from undissolved particulate matter in the ice is taken into account. Their result for the ratio, in good agreement with ours, conflicts with those reported by Amin *et al.* and Wasson *et al.* McCorkell *et al.* pointed to the possibility that the previous Al^{26} measurements might have been erroneous; most probably contributions by radioactive impurities to the 0.51-Mev peak were underestimated.

We believe that the reported high values for Al^{26} in marine sediments are in some doubt, and there is no conclusive evidence of extraterrestrial origin. We must emphasize that more experiments are required before we can determine the $\text{Al}^{26}:\text{Be}^{10}$ ratio in marine sediments and discuss the origins of these isotopes. Thus we prefer to avoid further discussion at present.

S. TANAKA, K. SAKAMOTO

J. TAKAGI, M. TSUCHIMOTO

Institute for Nuclear Study, University of Tokyo, Tanashi, Tokyo, Japan

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9. From the solution, hydroxide aggregate was precipitated at pH 5. The precipitate was dissolved in warm concentrated solution of NaOH, and the supernatant was neutralized with HCl until quite a small fraction of precipitate appeared in the solution (pH > 10). Most Be was coprecipitated with this fraction. The Be was extracted from the precipitate and purified by five cycles of solvent extraction. See J. R. Merrill, M. Honda, J. R. Arnold, *Anal. Chem.* **32**, 1420 (1960). The Al was purified by the procedure that consists principally in repetition of fractional recrystallization of aluminum alum. See S. Tanaka, K. Sakamoto, J. Takagi, M. Tsuchimoto, *J. Geophys. Res.*, in press.
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