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Particle accelerator measurements of ^{10}Be in marine accumulations: Intercomparison with beta counting method

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Abstract. Cosmogenic beryllium-10 activities have been measured in marine accumulations of up to ~ 6 m.y age by conventional beta counting technique and by accelerator mass spectrometry. The two sets of data at ^{10}Be levels of 10^9–10^{10} atoms/g agree within the absolute errors of the two methods. The detection limit for ^{10}Be by the accelerator mass spectrometry is about five orders of magnitude lower than that with the beta counting method.

Keywords. Particle accelerator; beryllium-10; marine sediments; manganese nodules; accumulation rates.

1. Introduction

With the advent of the accelerator mass spectrometric (AMS) technique, it has become possible to measure long-lived radionuclides at sensitivity levels of orders of magnitude better than what is possible with the conventional decay counting techniques (Mullar 1977; Gove 1978; Raisbeck *et al* 1978). Beryllium-10 is now measurable in few litres of water (Raisbeck *et al* 1979a, 1980, 1981) and in sub-gram quantities of marine sediments and ferromanganese nodules using the AMS technique (Raisbeck *et al* 1979b, 1981; Turekian *et al* 1979; Krishnaswami *et al* 1982). These studies are valuable for understanding the related geophysical/geochemical and astrophysical phenomena (Raisbeck *et al* 1981; Somayajulu 1977). Comparison between the two techniques using natural samples in the case of cosmogenic ^{14}C and with standards in the case of ^{10}Be resulted in good agreements (Krishnaswami *et al* 1982; Brown *et al* 1982; Bennett *et al* 1978; Mullar *et al* 1978; Stuiver 1978).

2. Experimental methods

^{10}Be measurements on marine sediments and on manganese nodules were carried out at the Physical Research Laboratory (PRL) using radiochemical separations followed by conventional beta counting techniques (Sharma 1982). Some of the radiochemically pure BeO samples, after beta counting, were measured at the Laboratorium fur Kernphysik, Zürich using the AMS method (Wolfli *et al* 1982). The chemical extraction of ^{10}Be was carried out by the addition of a few mg of Be carrier since the intrinsic concentrations of beryllium are low, of the order of a few ppm. This amount of carrier does not present any problems in the beta counting or in the AMS technique. Self

absorption is negligible in the first case and a sample of a few mg is even most convenient for analysis by AMS.

From a set of 8 samples (table 1) seven were measured at Zurich out of which four were meant for intercomparison; the sixth sample was known to contain ^{10}Be but its activity was too low to be precisely measured by the beta counting method. The other two samples were 'blanks' prepared to check on the contaminating activities (other than ^{10}Be) arising from chemicals used in the extraction and purification of beryllium. Sample 1 is marine sediment (red clay) from a depth of 32–48 cm of a gravity core from the central equatorial Pacific. Samples 2–6 are sliced sections from ferromanganese nodules out of which samples 2–4 belong to the sample nodule, ANTP 58D whereas 5 and 6 belong to nodule ARIES 39D; both nodules are from the Pacific Ocean.

^{10}Be measurements by the beta counting method were followed by checks like repurification, recounting and half-thickness measurements of the beta radiation. Aliquots of the pure BeO extracts were sent to Zurich for ^{10}Be measurements using the Tandem-van-deGraaf accelerator mass spectrometer (Wolfli *et al* 1982). Each sample was run independently three times and each blank twice. There is a systematic error of 2–3% and the statistical error in the case of all samples during all runs is less than 1%; for blanks it is 20–30%. The AMS ^{10}Be data presented in table 1 are the mean values of the three runs and the errors quoted are one sigma standard deviations from the means. The ^{10}Be concentrations obtained by the decay counting method are also presented in table 1. Errors quoted for these values are quadratic sums of one sigma counting statistics (1.4–8.7%), errors associated with chemical (1.2–2.5%) and counting (2.0–3.7%) efficiencies and one sigma counting statistics on the mean-blank activity including background of the detector (4–6%). Two of the PRL blanks (samples 7 and 8) were also analysed by AMS. It must be noted here that the chemical reagents introduce a beta blank of about 2 counts/ hr (cph) as determined from a large number of blank runs on ^{10}Be free samples (Sharma 1982). The blank activity is not due to ^{10}Be as shown by the accelerator results (table 1); it may be due to ^{40}K and some of the daughter nuclides of the U–Th series which may follow the chemical purification produces used for beryllium. The ^{10}Be data are blank corrected. It is seen from table 1

Table 1. Intercomparison of ^{10}Be measurements in marine deposits by decay and AMS techniques.

Sample	Depth (mm)	^{10}Be/^{9}Be (atom ratio)	AMS (atoms/g) × 10^9	Decay counting ^{10}Be (atoms/g) × 10^9
NOVA III-16* (sediment)	320–480	$(8.22 \pm 0.45) \times 10^{-10}$	(6.32 ± 0.35)	(7.2 ± 0.3)
ANTP 58D† (nodule)	0–4.8	$(7.82 \pm 0.25) \times 10^{-10}$	(2.30 ± 0.07)	(3.1 ± 0.2)
ANTP 58D (nodule)	4.8–12.8	$(7.97 \pm 0.45) \times 10^{-10}$	(1.27 ± 0.07)	(1.4 ± 0.1)
ANTP 58D (nodule)	21.0–27.8	$(5.77 \pm 0.47) \times 10^{-10}$	(1.25 ± 0.13)	(1.2 ± 0.1)
ARIES 39D† (nodule)	0–6.9	NM	NM	(7.5 ± 0.9)
ARIES 39D (nodule)	6.9–12.3	$(0.52 \pm 0.06) \times 10^{-10}$	(1.79 ± 0.18)	ND
Chemical Blank 1	—	$(6.80 \pm 0.04) \times 10^{-13}$	(1.57 ± 0.01) **	ND
Chemical Blank 2	—	$(1.70 \pm 0.50) \times 10^{-13}$	(0.40 ± 0.12) **	ND

NM = Not measured; ND = Not detected; * = Sediment sample (Lat. 0° 14.1′N, Long. 179° 07.9′W, water depth 5180 m); † = Manganese nodules, ARIES 39D (34° 15.2′N, 143° 51.2′E, 1819/1448 m) and ANTP 58D (18° 57.2′S, 135° 47.6′E. 3555/2836 m); ** = These are total ^{10}Be atoms in the blanks.

that the ^{10}Be concentrations of the blanks are two to three orders of magnitude smaller than those of the samples.

3. Results and discussion

The agreement between the results from the decay and AMS techniques for samples 1–4 (table 1) is surprisingly good, if one takes into account the fact that the uncertainty in the decay constant of ^{10}Be ($\lambda = 4.62 \times 10^{-7}\ yr^{-1}$) is of the order of 20% (adopted half-life of $^{10}Be = (1.5 \pm 0.3).10^6$ yrs) (Yiou and Raisbeck 1972), and that the $^{10}Be/^{9}Be$ ratio was measured relative to that of a standard (produced by neutron activation of ^{9}Be), whose isotopic ratio is known only within about 15%.

In sample 6, the bottom section of ARIES 39D, where there was no measurable signal above the blank level by beta assay, atom counting yields a ^{10}Be concentration of $(1.79 \pm 0.18) \times 10^9$ atoms/g nodule. Based on the decrease in the ^{10}Be activity between the two sections of ARIES 39D, its accumulation rate is deduced to be 2.0 mm/10^6 yrs (table 1, figure 1).

The beta counting data of ANTP 58D do not lead to a monotonic ^{10}Be decay with depth. There is a change in the slope of the curve with depth, indicating a drastic change in the growth rate of this nodule. Results using the AMS technique confirm this behaviour. This case is similar to that of a small nodule from the Indian Ocean (Krishnaswami *et al* 1982), but departs significantly from the exponential decrease with depth observed in the case of about a dozen nodules from the world oceans (Sharma and Somayajulu 1982).

Growth rates based on the first two points of ANTP 58D are 3.8 and 5.3 mm/ 10^6 yrs by the decay and AMS techniques, respectively. The growth rates of the two nodules reported here are in the range observed for other nodules from the world oceans (1–8

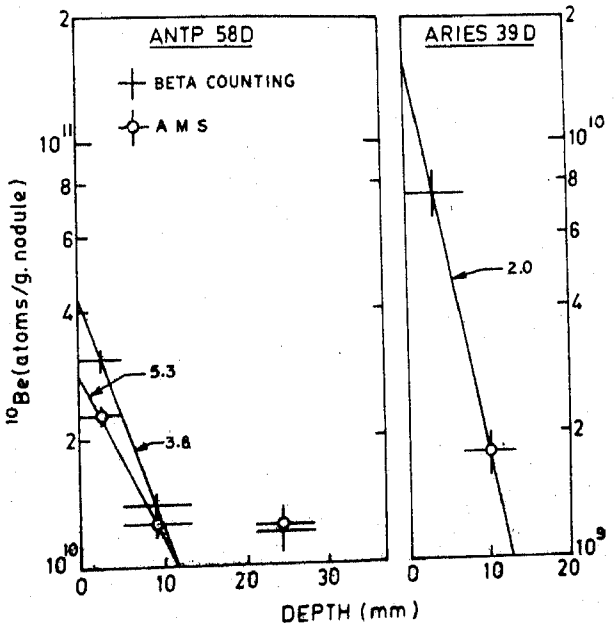

Figure 1. Plots of ^{10}Be concentration versus depth for nodules ANTP 58D and ARIES 39D. Numbers indicated by arrows are accumulation rates in units of mm/10^6 yrs.

mm/ 10^6 yrs — Ku and Broecker 1969; Sharma and Somayajulu 1982). The approximate age of the oldest nodule sample reported in this comparative study is 6 m.y.

From this study it follows that the beta counting technique can sately be used to measure samples containing 5×10^{11} atoms of ^{10}Be considering the limit placed by the activity of the chemical blank.

The present detection limit for AMS method on the other hand is about 10^7 atoms mg Be (corresponding to a ^{10}Be/ ^{9}Be atom ratio of 10^{-13} which is usually limited by the boron contamination of the samples (Brown *et al* 1982; Wolfli *et al* 1982) and the chemical blanks (table 1). Therefore, with typical accumulation rates, ^{10}Be is detectable in ~10 mg of open ocean manganese nodules and about 100 mg of Pacific red clay sediments with the AMS technique. The studies of depositional histories of manganese coatings on biological debris and of micronodules now appear feasible.

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