[Radiocarbon, Vol 25, No. 2, 1983, P 693-700]

TANDEM ACCELERATOR MEASUREMENTS OF ^{10}BE DEPOSITION RATES

DE NELSON, RG KORTELING, JR SOUTHON*, JS VOGEL* AND I NOWIKOW*

Archaeology and Chemistry Departments, Simon Fraser University, Burnaby, British Columbia, Canada

and

TL KU, MASASHI KUSAKABE AND JL REYSS**

Department of Geological Sciences, University of Southern California, Los Angeles

ABSTRACT. A Tandem Van de Graaff accelerator has been modified for use in the direct measurement of natural abundances of ^{10}Be and ^{14}C. A description of the system is given and some ^{10}Be results on oceanographic samples are discussed.

INTRODUCTION

We present a short review of our work with the McMaster University Tandem accelerator for the study of the radioisotopes ^{10}Be and ^{14}C. We have made over 100 ^{10}Be measurements on oceanographic samples. We find ^{10}Be is an ideal isotope for testing developing systems while providing useful geophysical information. We have not yet begun natural ^{14}C measurements, as our initial system did not have sufficient sensitivity. This deficiency is now rectified, and we should soon be starting ^{14}C dating.

TECHNICAL METHODS

The general features of our system are similiar to those of most other laboratories. Negative ions are accelerated and then magnetically and electrostatically selected before detection with Faraday cups and solid-state particle detectors. Our system differs from others in that we inject, accelerate, and detect both the stable and rare isotopes simultaneously instead of sequentially. We make no attempt to determine the absolute efficiency of the system, but measure

* Also: Department of Physics, McMaster University, Hamilton, Ontario, Canada

** Permanent address: Centre des Faibles Radioactivités, 91190 Gif-sur-Yvette, France

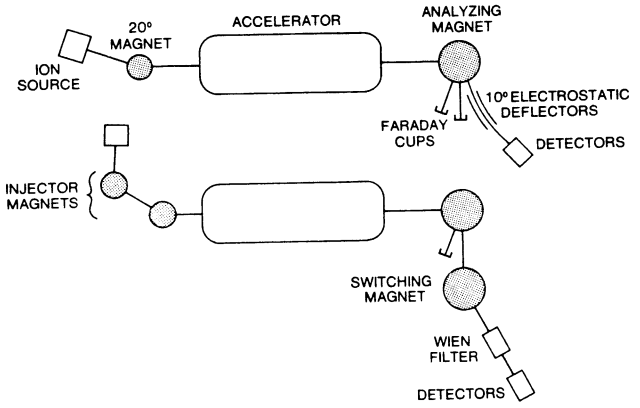

Fig 1A. (Top) The first-generation accelerator system used in making the measurements reported here. Fig 1B. (Bottom) The second-generation system which is now being completed, and which will be used in future studies.

all samples relative to a standard measured alternately with the unknowns. We operate the system as an analogue of a conventional isotope-ratio mass spectrometer.

Figure 1A shows our first-generation system. The ions from a reflected-beam Cs sputter source are injected by the 20^{o} inflection magnet. This is not ideal, as only the rare isotope can be injected on-axis, resulting in very different optical properties for the rare and major beams. This problem was compounded by the optics of the inclined-field tubes that were used in the accelerator until recently. These tubes further accentuated the differences between the beams. After acceleration, the ions were analyzed with the normal accelerator analyzing-magnet which has an enlarged vacuum chamber to accommodate the different beams. The rare isotope was further selected by a 10^{o} electrostatic deflection 80cm from the analyzing magnet exit point. Detection was done by commercial E-ΔE detectors for the rare isotope and Faraday cups for the major beams.

This system has been quite adequate for natural ^{10}Be measurements. Under typical operating conditions, for BeO samples of 0.1 to 1mg, we obtained ~0.5μA of BeO^{-} current and 30 to 40nA of analyzed $^{9}Be^{4+}$ current. Detection sensitivity was ca $^{10}Be/^{9}Be = 5 \times 10^{-13}$, limited primarily by nuclear reactions in the Al guard foil preceding the detectors and sometimes by the ^{10}Be count-rate, as not all natural samples

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

yield the same output from the source. The measurement accuracy with respect to our ^{10}Be standard ranged from 2 to 10%, and was very dependent on the condition of the accelerator.

This system did not provide sufficient sensitivity for ^{14}C measurements. ^{13}C ions from the stable-isotope beam were scattered into the rare-isotope beam by residual gas molecules in the magnet chamber. This background intensity was about equal to the intensity of ^{14}C from modern carbon, and so precluded useful measurement. Further magnetic filtering is required to eliminate these contaminant ions.

We are now completing second-generation modifications to every part of the system (fig 1B). A new injection system (Lobb et al, 1981) is nearing completion. This should simultaneously inject all isotopes of interest into the accelerator in a uniform fashion. The accelerator itself has been extensively upgraded in the past two months. The beam tubes and charging system were replaced with better designs. A new rare-isotope detection line including further magnetic and electrostatic filtering (the laboratory switching magnet and a Wien filter) is now complete. Preliminary tests show that there is now sufficient sensitivity for ^{14}C measurement. These modifications should allow us to determine ^{14}C and ^{10}Be concentrations with much better accuracy and sensitivity. Southon et al (1982) give detailed description of our system and measurement methods.

^{10}BE RESULTS

Most of our ^{10}be measurements were made on deep-sea manganese nodules as part of the manganese nodule program (MANOP) sponsored by the US National Science Foundation. We studied several of these in considerable detail and found that accelerator ^{10}Be determinations provide a powerful new tool for studying these sea-floor concretions. Much information on the long-term growth history of nodules was learned. The growth rates of mms/Myr determined from the ^{10}Be concentrations compared very well with those by uranium-series isotopes measured on the same nodules.

We also studied ^{10}Be concentrations in two manganese crusts which are similar to nodules but fixed to the ocean floor and not subject to possible roll-over. They can provide a several-million-year record of ^{10}Be deposition near the sample. If the logarithms of the ^{10}Be concentrations are a linear function of the sampling depth in the crust, it is reasonable to suggest that both the average ^{10}Be deposition and the crust accretion rate have been constant, where the average is taken over the time-interval represented by the thickness of the successive sample layers analyzed.

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

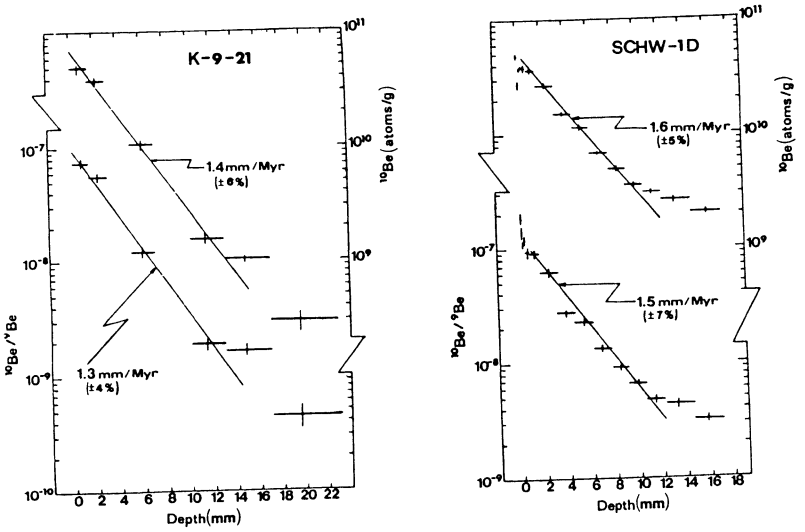

Fig 2. Plots of the ^{10}Be concentrations (on a residue-free basis) and $^{10}Be/^{9}Be$ ratios in two Mn crusts. Least squares fits for the linear segments of the plots give estimates for the accretion rates. The horizontal lines on each point give the depth intervals analyzed, and the positions of the vertical lines (measurement uncertainty) are placed at the effective measurement depths.

Figure 2 shows both the ^{10}Be concentrations (upper plots) and the $^{10}Be/^{9}Be$ ratios in a crust from the equatorial Atlantic (K-9-21 at $7^{o}58'N$, $21^{o}02'W$) and one from the North Pacific (SCHW-1D at $30^{o}N$, $140^{o}W$). The ^{9}Be measurements that were required to obtain the $^{10}Be/^{9}Be$ ratios were made by atomic absorption spectroscopy. Except near the very surface of sample SCHW-1D, the sampling intervals on these crusts represent depositions averaged over time spans of the order of a million years.

For both crusts, the plots are log-linear for the first ~10mm of sample. This indicates that the ^{10}Be production and deposition rate and the crustal growth rate have remained quite constant (the data suggest to within $\sim\pm$6-7%) for the past 7-9 Myr. Prior to that time, the crusts from both oceans show a very similar deviation, which could be due either to a higher ^{10}Be deposition rate in both oceans prior to 7-9 Myr ago or to a global change in the oceans which would similarly change the growth rates of these two crusts. The growth rate

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

change is deemed unlikely, though, because no discernible major changes in the Fe/Mn ratio or in the concentrations of ^{9}Be and alumino-silicates occur across the 7-9 Myr boundary in these crusts (Ku et al, 1982). Ciesielski et al (1982) suggested that the formation of the West Antarctic ice shelf at ca 7-9 Myr ago caused a permanent change in the global abyssal circulation; perhaps these crusts recorded that event.

The detailed study of the Pacific crust shows that ^{10}Be concentration changes markedly in the outermost millimeter. There was not sufficient sampling resolution to detect such a change in the Atlantic crust, but we saw this feature in all the manganese nodules we studied. We cannot determine from these data whether this indicates a gradual decrease in the ^{10}Be production rate from 0.5 Myr ago to the present, or whether it is due to the nature of the chemical processes by which the Be is incorporated into the manganese deposits. Kusakabe and Ku (ms in preparation) have developed a model which explains but does not prove the latter possibility.

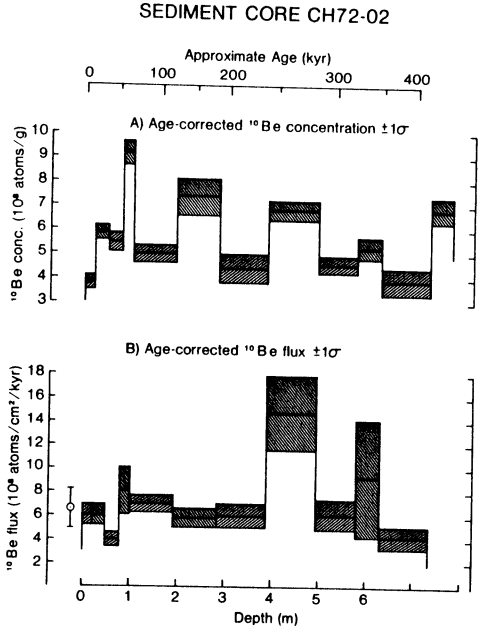

Fig 3. Plots of the age-corrected ^{10}Be concentrations in, and the calculated ^{10}Be fluxes into, a deep-sea sediment core. Each step in the histogram represents the average value for oxygen-isotope states I-XII, reading from left to right. An approximate age-scale is given at the top.

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

On the million-year time-scale, then, ^{10}Be can provide a good chronometer for oceanographic studies. For shorter periods, we have information from a preliminary analysis of ^{10}Be concentrations in a sediment core (CH72-02 from 40°36'N, 21°42'W; 3485m) from the Atlantic, representing ~500 kyr of depositional history (fig 3) as determined by ^{230}Th and oxygen-isotope measurements. The core was divided into sections corresponding closely to the oxygen-isotope stages. The upper histogram of figure 3 gives the age-corrected average ^{10}Be concentrations as a function of down-core depth. For this core, age estimates as a function of depth were determined from the ^{230}Th data, from a comparison of the oxygen-isotope record of this core with the isotope record of Morley and Hays (1981) and from the first appearance of the micro-organism, E Huxleyi. Linear-regressions of the age-depth data gave high correlation coefficients ($r \sim 0.996$). At first glance, this suggests that the sedimentation rate for the core is constant, and that the ^{10}Be concentration histogram also gives the variation with time of the ^{10}Be deposition rate at this location. We see, eg, a variation between isotope stages 1 and 2 that is very similar to that found by Raisbeck et al (1981) in an Antarctic ice core, and it is tempting to conclude that this indicates a global change in ^{10}Be production.

We believe this interpretation is incorrect. A plot of the non-carbonate fraction (or the "clay" fraction) of our core vs depth gives a histogram identical in shape to the ^{10}Be concentration histogram, indicating that the ^{10}Be concentration variations primarily reflect the changes in biogenic carbonate production and not ^{10}Be production. (The effect of carbonate dissolution should be minor, as this core is well above the lysocline depth. The constancy of the core sedimentation rate is more apparent than real.

The time-dependent fluxes of ^{10}Be into the core are given in the lower histogram of figure 3. We have used the isotope-stage boundary ages of Morley and Hays (1981) to determine the average sedimentation rate for each stage. We then calculated the ^{10}Be fluxes with these average rates, the age-corrected ^{10}Be concentrations, and the in situ sediment densities. The uncertainties include an estimate of the uncertainty in transposing the Morley and Hays' ages to this particular core, the uncertainty in the ^{10}Be measurement, and the uncertainty in the core-density estimates.

Any examination of this histogram for evidence of change in ^{10}Be (and thus, ^{14}C) production rates must be done with the caution that the values plotted here depend strongly on the ages of the stage boundaries. Perhaps it is preferable to base the flux calculations on the ^{230}Th ages measured for this core, but these determinations have analytical uncertainties

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

(especially for the older samples) that are quite large. The age values we have chosen from the literature (Morley and Hays, 1981) are derived from a variety of methods and are perhaps the best current estimates, but an assessment of the absolute accuracy of these ages has not been made.

The point plotted to the left of this histogram gives the ^{10}Be production-rate predicted by Reyss, Yokoyama and Guichard (1981) from spallation cross-sections and cosmic-ray intensities. There is very good agreement between this predicted value and the observed flux into the core.

If the age estimates are correct, the histogram gives no evidence that the ^{10}Be flux averaged over each isotope stage (hence, climatic cycle) has changed more than ca 25 to 30% over the past 1/2 Myr, with the possible exception of stages 3 and 8. Further, whether we use the isotope-record ages or the ^{230}Th ages, there is no evidence of differences between the average ^{10}Be fluxes for stages 1 and 2.

We are also attempting to examine ^{10}Be production on a shorter time scale, with detailed ^{14}C and ^{10}Be measurements on a 20,000 year-old lake core from Tennessee. The topography indicates that the catchment area of the pond has not changed a great deal during that time. Only preliminary determinations on two segments of the core have been made; one for a modern sample and one for a sample of age ~14,000 ^{14}C years. The results indicate that, 1) the ^{10}Be concentration in the modern sample is 3 times higher than that in the 14,000-year-old sample, 2) the ^{10}Be flux into the core is about a factor of 4 lower in modern times than at 14,000 years ago, and 3) the $^{10}Be/^{9}Be$ ratio in the modern sample is approximately a factor of two lower than the ratio in the 14,000-year-old sample.

These data are apparently inconsistent and do not provide any reliable general information on ^{10}Be production and distribution of these times. In order to extract such information from this core, we will at least have to derive very accurate estimates for the sedimentation rates, so that the ^{10}Be fluxes can be accurately determined. At present, the only method by which this can be done is ^{14}C dating, and a calibration scale for that technique exists only for the past 8000 years.

CONCLUSION

These studies have examined the production and distribution of the radioisotope ^{10}Be, both to evaluate its potential as a dating tool, and to gain information on the production rate variation. We have found that measurements of ^{10}Be concentrations do not always provide unambiguous information on ^{10}Be production rates. To translate ^{10}Be concentra-

https://doi.org/10.1017/S0033822200006032 Published online by Cambridge University Press

tions in a sample to ^{10}Be flux for short time scales compared to the mean-life of ^{10}Be, we require accurate, independent dating techniques.

ACKNOWLEDGMENTS

We thank the staff of the McMaster University Accelerator Laboratory for their continuing support of our program, Tom Brown for permission to discuss his preliminary lake-core data, and Gord Richards, John Sheppard, and Nadine Bohna for technical assistance. Funding is provided by the NSF Manganese Nodule Program, by NSERC of Canada, and by Simon Fraser and McMaster Universities.

REFERENCES

Ciesielski, PF, Ledbetter, MT, and Ellwood, BB, 1982, The development of Antarctic glaciation and the Neogene paleo-environment of the Maurice Ewing Bank: Marine Geol, v 46, p 1-51.

Ku, TL, Kusakabe, M, Nelson, DE, Southon, J, Korteling, RG, Vogel, J, and Nowikow, I, 1982, Constancy of oceanic deposition of ^{10}Be as recorded in Mn crusts: Nature v 299, p 240-242.

Lobb, DE, Southon, JR, Nelson, DE, Wiesehahn, W, and Korteling, RG, 1980, An ion injection system for use in tandem accelerator radioisotope dating devices: Nuclear Instruments Methods, v 179 p 171-180.

Morley, JJ and Hays, JD, 1981, Towards a high-resolution, global, deep-sea chronology for the last 750,000 years: Earth Planetary Sci Letters, v 53, p 279-295.

Raisbeck, GM, Yiou, F, Fruneau, M, Loiseaux, JM, Lieuvin, M, Ravel, JC, and Lorius, C, 1981, Cosmogenic ^{10}Be concentrations in Antarctic ice during the past 30,000 years: Nature, v 292, p 825-826.

Reyss, J-L, Yokoyama, Y, and Guichard, F, 1981, Production cross-sections of ^{26}Al, ^{22}Na, ^{7}Be from argon and of ^{10}Be, ^{7}Be from nitrogen: implications for production rates of ^{26}Al and ^{10}Be in the atmosphere: Earth Planetary Sci Letters, v 53, p 203-210.

Southon, JR, Vogel, JS, Nowikow, I, Nelson, DE, and Korteling, RG, 1983, The measurement of ^{10}Be concentrations with a Tandem accelerator: Nuclear Instruments Methods, in press.