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SPALLOGENIC ^{14}C IN HIGH-ALTITUDE ROCKS AND IN ANTARCTIC METEORITES

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ABSTRACT. ^{14}C in high-altitude rocks and in Antarctic meteorites of long terrestrial age has been found at levels consistent with *in-situ* production by cosmic rays. Levels of 0.2–0.59 dpm/kg (or $1.0–2.6\times10^6$ $^{14}C/g$) are found in high-altitude samples ranging from 3300 to 5460m. Similar values are observed in Antarctic meteorites that have been dated by ^{81}Kr by Freundel, Schultz & Reedy (1986) as over 100 kyr old.

INTRODUCTION

Cosmic rays interact with the surface of the earth as well as its atmosphere and with objects such as meteorites in space. Due to the rapid attenuation of high-energy particles in the atmosphere, the surface flux of high-energy neutrons is 2 to 3 orders of magnitude below the free-space level (Lal & Peters, 1967). Until recently, cosmogenic ^{14}C has only been determined in meteorites of terrestrial age <30 kyr (Goel & Kohman, 1962; Suess & Wänke 1962; Born & Begemann, 1975; Boeckl, 1972; Fireman, 1978, 1979, 1983). Begemann *et al* (1972), Fireman, DeFelice and D'Amico (1976) and Fireman (1978) also studied lunar samples. Levels of ^{14}C in some older meteorites from Antarctica have been detected down to ca 1 dpm/kg (Jull *et al,* 1984; Brown *et al,* 1984; Beukens, Rucklidge & Miura, 1988). The levels determined in these samples are higher than expected from production alone (Yokoyama, Reyss & Guichard, 1977) and warrant further investigation. Sample sizes have been reduced to <1g (Jull & Donahue, 1988; Beukens, Rucklidge & Miura, 1987) from the ~10g required earlier. The enhanced sensitivity of accelerator mass spectrometry (AMS) and reduced blank levels have now allowed us to investigate cosmic-ray spallation ^{14}C in high-altitude terrestrial rocks, as well as in some Antarctic meteorites, where all extraterrestrial ^{14}C has decayed (Jull, Donahue & Linick, 1989). Studies of other "*in-situ*-produced" isotopes, such as ^{10}Be and ^{36}Cl in Libyan desert glass (Klein *et al,* 1986, Lal, 1987) and high-altitude rocks (Phillips *et al,* 1986; Leavy *et al,* 1986), indicate reasonable agreement with theoretical predictions (Lal, 1987; Yokoyama, Reyss & Guichard, 1977). If these levels can be established, the potential of dating surfaces by studying the levels of cosmic-ray-produced radionuclides may be realistic. This paper presents results of an initial study of cosmogenic ^{14}C in terrestrial rocks. A companion study of ^{14}C in rocks using acid extraction techniques will be reported elsewhere (Lal, Jull & Donahue, unpub results).

THE SAMPLES

Available mineralogic data for the terrestrial sample suite are summarized in Table 1. Seven samples were analyzed, of which 3 are granitic plutonic rocks, 3 are intermediate volcanics, and 1 (the Herkimer quartz) is a sub-surface "control sample" from a location noted for clear quartz (Sinkankas, 1976). The volcanic suite are surface "grab samples" from the upper slopes of three major volcanoes in east-central Mexico: Popocatepetl, La Malinche, and Citlaltepetl, alias Pico de Orizaba (hereafter "Orizaba").

TABLE 1

Composition of the terrestrial samples

Sample	Rock type	Major minerals	Minor minerals
003	Biotite granite	Plagioclase, K-feldspar, quartz, biotite	Myrmekite, oxide, apatite, zircon, epidote, chlorite (2)
299	Porphyritic andesite	Glass, plagioclase, oxides after amphibole	Amphibole (Trace)
090	Porphyritic andesite	Glass, plagioclase	Orthopyroxene, clinopyroxene, sulphides (5)
304	Porphyritic andesite	Plagioclase, glass, amphibole	Clinopyroxene, oxides (Trace)
SMTP-1	Granodiorite	–	–
D-1	Granodiorite	–	–
Q2A	Quartz	Quartz	–

Where petrographic data are available, the constituent minerals of the samples are listed in decreasing order of abundance (based on estimated volume percent). The 'minor' minerals are present in total amounts varying from trace to 5%.

EXPERIMENTAL

A sample of 0.5–2g rock is ground up, mixed with ca 2–4g iron-accelerator chips as a combustion flux, and placed in a low-carbon crucible. The samples are placed in a muffle furnace for 1 hr at 500°C to remove contaminants. The sample/iron mix is then placed in the RF extraction furnace and heated to fusion for up to 8 min in a flow of oxygen. The oxygen is flushed through the system every 1–2 min and collected in a liquid-nitrogen trap, after passing through a dust trap, MnO_2 and a CuO/Pt furnace at 500°C. After heating, the O_2 is pumped away, and the remaining CO_2 is separated from water by distillation at -78°C. The amount of CO_2 is measured, giving an estimate of the C content of the sample, and is then diluted to 1–2cc STP CO_2 with ^{14}C "dead" gas. The diluted CO_2 is reduced to graphite over iron (Slota *et al,* 1987). The resultant graphite powder is pressed into an accelerator target holder and mounted in a 10-position wheel along with 2 standards, normally NBS oxalic acids I and II. The methods used for accelerator measurement of ^{14}C have been described in detail by Linick *et al* (1986).

RESULTS AND DISCUSSION

CO_2 was extracted from several high-altitude rocks as a test study of *in-situ* production of ^{14}C by cosmic-ray spallation. The fraction released at temperatures above 500°C gives a reliable measure of the spallogenic C content (Desmarais, 1983; Jull & Donahue, 1988; Jull, Donahue & Linick, 1989). As discussed, the 500°C preheating step removes low-temperature weathering products and other contaminants, with the exception of calcite. Results are shown in Table 2. The data shown are given both corrected and uncorrected for the extraction system blank of $(8\pm1)\times10^5$ ^{14}C. This blank mainly results from residual ^{14}C memory in the extraction line, which has also been used for meteorites. The levels shown in Table 2 are given in dpm/kg to facilitate direct comparison with the production rates, such as those calculated by Yokoyama, Reyss & Guichard (1977), which are proportional to the cut-off rigidity, a function of geomagnetic latitude (Shea *et al,* 1987). The data are compared in Figure 1, including some additional data from three Antarctic meteorites. These three samples were dated by

TABLE 2

^{14}C content of high-altitude rocks

Sample	Site	Altitude	Geomagnetic latitude	Fraction modern ^{14}C	cc CO_2*	^{14}C dpm/kg Uncorrected	^{14}C dpm/kg Corrected
003	Mt Massive, Colorado, 2.06g	4396m	49°N	0.0561±0.002	2.24	0.41±0.01	0.31±0.03
	Mt Massive, Colorado, 0.945g			0.054±0.002	1.63	0.49±0.02	0.34±0.03
299	La Malinche, Mexico, 0.945g	4461m	28°N	0.088±0.002	1.75	1.08±0.03	0.55±0.04
090	Popocatepetl, Mexico, 1.107g	5452m	28°N	0.087±0.006	1.46	0.76±0.05	0.59±0.05
304	Orizaba, Mexico, 1.055g	4270m	28°N	0.045±0.002	1.53	0.43±0.01	0.25±0.03
SMTP-1	Mt Morgan, California, 1.454g	4000m	43°N	0.050±0.002	1.85	0.42±0.02	0.29±0.02
D-1	White Mts California, 1.698g	3300m	46°N	0.110±0.003	0.90	0.39±0.01	0.27±0.02
Blank: Q2A	Herkimer, New York, 0.967g	N/A	52°N	0.020±0.002	1.20	0.16±0.02	<0.04

*At 25°C

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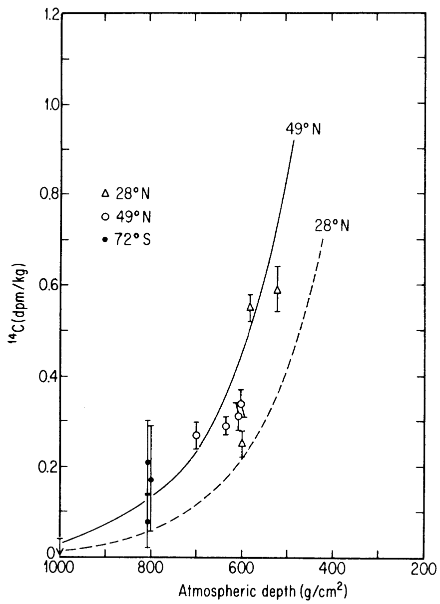

Fig 1. ^{14}C (dpm/kg) measured in rocks of different altitude (plotted as atmospheric depth in g/cm^2) and geomagnetic latitude

^{81}Kr (Freundel, Schultz & Reedy, 1986) as having a terrestrial age >100 kyr, so no extraterrestrial ^{14}C from irradiation in space should be present. The error bars on the meteorite samples are larger due to the smaller sample size (0.5–0.7g) used in these extractions, which were reported in detail (Jull, Donahue & Linick, 1988). The three meteorites plotted are Allan Hills 78132, Pecora Escarpment 82502, and Elephant Moraine 79005, which had ^{14}C contents of 0.21±0.09, 0.08±0.06 and 0.17±0.11dpm/kg, respectively (Jull & Donahue, 1988; Jull, Donahue & Linick, 1989). Values from two granitoid samples (003 and SMTP-1) plot very well when compared to the curves estimated from the data of Yokoyama, Reyss & Guichard, 1977), as does andesite sample 090 from Mexico. A granitic sample, from the White Mountains of California, with a desert varnish age of ca 10,000 yr (Dorn, pers commun) plots close to the 40°N curve from the data of Yokoyama, Reyss and Guichard (1977). However, if the desert varnish age is correct, the sample ought to be only 70% saturated in ^{14}C. The variation in the Mexican samples is almost certainly due to the young ages of some of these volcanic rocks. Sample 304 from Orizaba may be undersaturated, compared to the other samples. The latest phase of volcanism on Orizaba, in which much of the upper parts of the volcano have been constructed, occurred during the past 13,000 yr (Negendank *et al,* 1985). An age of 13,000 yr would give a value of ca 80% of saturation for ^{14}C. The Antarctic meteorite data also seem to fit the curve reasonably well, with large errors. Of course, these comparisons assume that the estimates made by Yokoyama, Reyss and Guichard (1977) are reasonable. The theoretical values of these authors rely on a few measurements of ^{22}Na and ^{24}Na in samples of metal from manmade objects exposed at altitudes of 2070–4600m in France (Yokoyama, Reyss & Guichard, 1977). Extrapolation of the production rates to other isotopes assumes knowledge of the relative cross-sections and also of the attenuation of high-energy cosmic-ray particles in the atmosphere. In the case of ^{14}C, the cross-section for neutron spallation is assumed to be similar to that of protons (Reedy & Arnold, 1972) above a few hundred MeV, with some assumptions about the excitation function in the range 50–200MeV (Reedy & Arnold, 1972). Yokoyama, Reyss and Guichard (1977) assume the atmosphere attenuates high-energy particles with depth (in g/cm^2) similar to the lunar surface, for which the best profile data are available, and a mean attenuation length of 192±5g/cm^2 was used, rather larger than the 118g/cm^2 used by Reedy & Arnold (1972) in their calculations on lunar cores. No geological evidence of erosion was observed in the samples studied. As has been discussed by Klein *et al* (1986), in the case of ^{10}Be, the effects of erosion would reduce the total concentration of ^{14}C. However, if the erosion is very low, as expected for these rock samples, the effects on radionuclides such of ^{14}C will be negligible, as is also the case for the ^{36}Cl samples studied by Leavy *et al* (1987).

CONCLUSIONS

The first measurements of spallogenic ^{14}C in terrestrial rocks, and in some Antartic meteorites, are reported. The results seem to indicate fair-to-good agreement with theoretical calculations (Lal, 1987; Yokoyama, Reyss & Guichard, 1977). In some samples, the complicating effects of a possible low rock age may result in undersaturation. If good saturated activity or production rate curves can be established for different latitudes and altitudes, then a reasonable chronology for dating of rock surfaces can be established. This will be of great benefit to geomorphology, which relies heavily on proxy estimates of age at present.

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