NOTE

Beryllium-7 deposition at Fayetteville, Arkansas, and excess polonium-210 from the 1980 eruption of Mount St. Helens

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(Received June 11, 1985: Accepted July 21, 1985)

Radiochemical measurements of ^7Be were carried out for a total of 246 individual rain and snow samples collected at Fayetteville (36°N, 94°W), Arkansas, during the period between September 1979 and December 1984. A sharp increase in the $^{210}\text{Po}/^7\text{Be}$ ratio in rain observed during the winter and spring months of 1980–1981 was attributed to the fallout of excess ^{210}Po from the May 1980 eruption of Mount St. Helens.

INTRODUCTION

The importance of ^7Be as a radioactive tracer in the study of atmospheric phenomena has long been well recognized (see, for example, Lal and Peters, 1967; Wogman *et al.*, 1968; Perkins *et al.*, 1970; Husain *et al.*, 1977; Dutkiewicz and Husain, 1979; Furukawa and Kojima, 1982). Measurements of ^7Be concentrations in individual rain and snow samples were initiated in our laboratories in the fall of 1979 and the analyses of all the samples collected during a five-year period 1979–1984 have been completed, except for the fact that the measurements were not carried out during a period of about 8 months in 1983. While this study was in progress, the eruption of Mount St. Helens occurred in May 1980 and it was noted that there was a sharp increase in the $^{210}\text{Po}/^{210}\text{Pb}$ ratio in rain during the winter and spring months of 1980–1981 (Kuroda *et al.*, 1984). An increase in the $^{210}\text{Po}/^7\text{Be}$ ratio in rain was also noted during the course of the study on ^7Be and these results indicated that we are dealing here with an atmospheric injection of excess ^{210}Po from the volcanic eruption. An estimate of the amount of excess ^{210}Po released into the atmosphere by

the 1980 eruption of Mount St. Helens is presented in this report.

EXPERIMENTAL

The rain and snow samples were collected by means of a sampling system located on the roof of the Chemistry Building of the University of Arkansas, which is located at Fayetteville (36°N, 94°W), Arkansas. The amount of rainwater used for the analysis of ^7Be ranged from 10 to 40 liters. The rain samples collected during the period between September 1979 and December 1981 contained appreciable amounts of fission products from the Chinese nuclear weapons tests. Since the gamma-rays from fission products such as ^{103}Ru , ^{106}Ru , ^{125}Sb and ^{140}La are known to mask the 477.6 KeV peak of ^7Be , we have decided to separate and radiochemically purify the beryllium fractions before they were counted. The radiochemical purification procedure used for the analyses of ^7Be was that described by Buchanan (1958). The purification procedure was omitted, however, for the samples collected after January 1, 1982, when the levels of fission product activities in the atmosphere became extremely low.

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Fig. 1. The concentrations of ^7Be in individual rain and snow samples collected at Fayetteville (36°N , 94°W), Arkansas, during the period between September 1979 and December 1984.

The radioactivity measurements were carried out with Ortec Ge (Li) gamma-ray detector. An Ortec preamplifier and an Ortec 572 main amplifier were used in conjunction with a Nuclear Data 575 ADC and a Nuclear Data 600 analyzer. The detector photopeak response curve was established with a set of calibrated gamma-ray sources of known activities, consisting of ^{57}Co , ^{60}Co , ^{85}Sr , ^{88}Y , ^{109}Cd , ^{113}Sn , ^{137}Cs , ^{139}Ce and ^{203}Hg obtained as a mixed source in solution form and point source from the National Bureau of Standards.

RESULTS AND DISCUSSION

Figure 1 shows the seasonal variation of the concentrations of ^7Be in individual samples of rain and snow collected at Fayetteville, Arkansas during the period between September 1979 and December 1984. The dates of the eruptions of Mount St. Helens (18 May 1980) and El

Chichon volcano (28 March 1982), and also the 25th Chinese nuclear test (16 October 1980) and the fall of the nuclear-powered Soviet satellite Cosmos-1402 (7 February 1983) are also shown in Fig. 1. The ^7Be concentrations in individual rain and snow samples fluctuated markedly. The maximum value of (91.3 ± 4.9) pCi per liter was observed in the rain of 5 May 1980 and the minimum value of (4.9 ± 0.6) pCi per liter in the rain of 10 December 1982.

Table 1 shows the bi-monthly average concentrations ($\bar{C}$) of ^7Be in rain calculated by the equation

$$\bar{C} = \frac{\sum R \cdot C}{\sum R} \quad (1)$$

where R is the amount of individual rainfall and C is the concentration of ^7Be in individual samples of rainfalls which occurred during a two-month period.

Matsunami *et al.* (1974) have reported that

Table 1. Bimonthly and annual average concentrations of ^7Be in rain at Fayetteville, Arkansas, during the period between September 1979 and December 1984. (*)

Year and month	Bimonthly total rainfall (mm)	Bimonthly average concentration (pCi/l)	Annual average concentration (pCi/l)
1979			
Sep. -Oct.	44.45	18.6	
Nov. -Dec.	110.74	27.0	
1980			
Jan. -Feb.	44.08	25.5	
Mar. -Apr.	164.98	30.3	
May -June	251.34	21.8	23.3
July -Aug.	30.48	27.3	
Sep. -Oct.	161.56	24.3	
Nov. -Dec.	112.02	12.8	
1981			
Jan. -Feb.	98.88	10.8	
Mar. -Apr.	163.72	25.6	
May -June	421.12	17.5	17.3
July -Aug.	263.39	15.6	
Sep. -Oct.	222.61	13.6	
Nov. -Dec.	45.46	26.7	
1982			
Jan. -Feb.	183.7	20.4	
Mar. -Apr.	130.3	20.3	
May -June	533.7	10.3	14.5
July -Aug.	87.4	24.9	
Sep. -Oct.	80.5	21.9	
Nov. -Dec.	379.0	11.7	
1983			
Jan. -Feb.	58.3	26.7	
Mar. -Apr.	—	—	
May -June	—	—	
July -Aug.	—	—	
Sep. -Oct.	—	—	
Nov. -Dec.	169.2	23.8	
1984			
Jan. -Feb.	104.9	26.3	
Mar. -Apr.	276.6	42.9	
May -June	212.8	27.5	27.8
July -Aug.	120.8	35.6	
Sep. -Oct.	252.0	18.7	
Nov. -Dec.	282.7	18.7	

(*) Total rainfall and an overall average ^7Be concentration in rain for a 56-month period from 1979 to 1984 were 5006.7mm and 20.5pCi per liter.

the annual average rainfall in Osaka (35°N , 136°E), Japan, was 1,196mm during 1970–1973 and the average ^7Be concentration in rain was 5.7×10^6 atoms per liter. The annual average rainfall in Fayetteville (36°N , 94°W), Arkansas, for the period 1980–1984, on the other hand, was 1,156mm and the average ^7Be concentration in rain was 5.0×10^6 atoms per

liter. The annual deposition of ^7Be at Fayetteville, Arkansas, can be calculated from our data to be 5.2 (dpm/cm 2 /y). This value is in excellent agreement with the reported values of 5.5 (dpm/cm 2 /y) at Chilton (51°N) and 5.2 (dpm/cm 2 /y) at Milford Haven (51°N), England (Peirson, 1963) and is comparable to the reported values of 4.3 (dpm/cm 2 /y) at Westwood (41°N), N. J., (Walton and Fried, 1962), 9.5 (dpm/cm 2 /y) at Rijswijk (52°N), Netherlands (Bleichrodt and Van Abkoude, 1963), 7.6 (dpm/cm 2 /y) at Bombay (19°N), India (Lal *et al.*, 1979), and 8.1 (dpm/cm 2 /y) at Quillayute (49°N), Washington (Crecelius, 1981). It is worthy of note here, however, that Turekian *et al.* (1983) have recently reported the values of 17.1 (dpm/cm 2 /y) at Bermuda (33°N) and 22.7 (dpm/cm 2 /y) at New Haven (41°N), Connecticut.

Figure 2 shows a comparison of the variations of the bi-monthly average ratios of $^{90}\text{Sr}/^7\text{Be}$ in rain at Fayetteville, Arkansas, during the period of 1979–1982. The ^{90}Sr and ^{210}Po data were taken from the reports of Burchfield *et al.* (1982), Burchfield and Kuroda (1983), and Kuroda *et al.* (1984). A marked increase in the $^{90}\text{Sr}/^7\text{Be}$ ratio in rain observed during the first half of 1981 is attributable to the 25th Chinese nuclear explosion of 16 October 1980 (Burchfield *et al.*, 1982; Burchfield and Kuroda, 1983). Note that the South African incident of 22 September 1979, in which a small nuclear weapon was reported to have been tested in the southern hemisphere, had no significant effect on the ^{90}Sr concentrations in rain collected at Fayetteville, Arkansas.

Figure 2 shows that a sharp increase in the $^{210}\text{Po}/^7\text{Be}$ ratio in rain occurred toward the end of 1980 and during the months of January and February 1981, which can be attributed to an atmospheric injection of excess ^{210}Po by a series of major eruptions of Mount St. Helens, which began on 18 May 1980 (Kuroda *et al.*, 1984). A small peak in the $^{210}\text{Po}/^7\text{Be}$ ratio in rain was observed during the winter months of 1979–1980, which may be attributable to the atmospheric injection of excess ^{210}Po from the

Fig. 2. The ratios of bi-monthly concentrations of ^{90}Sr to ^{7}Be and ^{210}Po to ^{7}Be in rain at Fayetteville, Arkansas, during the period between September 1979 and December 1982. The lines ab and cd show the estimated background values of the $^{90}\text{Sr}/^{7}\text{Be}$ and $^{210}\text{Po}/^{7}\text{Be}$ ratios, respectively, in rain.

Table 2. Calculations of the values of $F(^{90}\text{Sr})$ and $F(^{210}\text{Po})$.(a) Excess ^{90}Sr from the 16 October 1980 nuclear explosion.

Period	$(^{90}\text{Sr}/^7\text{Be})_{\text{obs.}}$ (Ci/Ci)	$\delta(^{90}\text{Sr})$ (Ci/Ci)	$D(^7\text{Be})$ (pCi/cm ²)	$F(^{90}\text{Sr})$ (pCi/cm ²)
<i>1981</i>				
Jan. -Feb.	0.019	0.012	0.1068	0.00075
Mar. -Apr.	0.043	0.036	0.4191	0.01509
May -June	0.033	0.026	0.7369	0.01916
Total				0.0350

(b) Excess ^{210}Po from the 18 May 1980 eruption of Mount St. Helens

Period	$(^{210}\text{Po}/^7\text{Be})_{\text{obs.}}$ (Ci/Ci)	$\delta(^{210}\text{Po})$ (Ci/Ci)	$D(^7\text{Be})$ (pCi/cm ²)	$F(^{210}\text{Po})$ (pCi/cm ²)
<i>1980</i>				
May -June	0.034	0.004	0.5479	0.0022
July -Aug.	0.042	0.012	0.0832	0.0010
Sep. -Oct.	0.028	(-0.002)	0.3926	—
Nov. -Dec.	0.075	0.045	0.1434	0.0065
<i>1981</i>				
Jan. -Feb.	0.150	0.120	0.1068	0.0128
Mar. -Apr.	0.056	0.026	0.4191	0.0109
May -June	0.037	0.007	0.7369	0.0052
July -Aug.	0.053	0.023	0.4109	0.0095
Sep. -Oct.	0.035	0.005	0.3028	0.0015
Nov. -Dec.	0.039	0.009	0.1214	0.0011
<i>1982</i>				
Jan. -Feb.	0.032	0.002	0.3748	0.0008
Total				0.0515

11 February 1979 eruption of Bezymianny volcano in Kamchatka, U.S.S.R. (Sheng and Kuroda, 1985). The effect of Mount St. Helens on the $^{210}\text{Po}/^7\text{Be}$ ratio in rain seems to have ceased at the beginning of 1982 when the $^{210}\text{Po}/^7\text{Be}$ ratio in rain decreased to about 0.03 (Ci/Ci). A small increase in the $^{210}\text{Po}/^7\text{Be}$ ratio in rain observed after the spring of 1982 is most likely due to the 28 March 1982 eruption of El Chichon volcano in Mexico.

An order of magnitude calculation of the total amount of excess ^{210}Po from the 1980 eruption of Mount St. Helens can be carried out by the use of the equations

$$\delta(^{210}\text{Po}) = (^{210}\text{Po}/^7\text{Be})_{\text{obs.}} - (^{210}\text{Po}/^7\text{Be})_o \quad (2)$$

and

$$F(^{210}\text{Po}) = D(^7\text{Be}) \cdot \delta(^{210}\text{Po}) \quad (3)$$

where $^{210}\text{Po}/^7\text{Be}$ is the ratio of bi-monthly concentrations of ^{210}Po and ^7Be ; the subscripts *obs.* and *o* refer to the observed and background values; D is the total amount of ^7Be deposition

during a two-month period; and F is the total amount of excess ^{210}Po which was transported to the ground by rain during the same period. We also have the relationships for ^{90}Sr :

$$\delta(^{90}\text{Sr}) = (^{90}\text{Sr}/^7\text{Be})_{\text{obs.}} - (^{90}\text{Sr}/^7\text{Be})_o \quad (4)$$

and

$$F(^{90}\text{Sr}) = D(^7\text{Be}) \cdot \delta(^{90}\text{Sr}) \quad (5).$$

The values of $(^{90}\text{Sr}/^7\text{Be})_o$ and $(^{210}\text{Po}/^7\text{Be})_o$ were estimated graphically to be 0.007 (Ci/Ci) and 0.03 (Ci/Ci), respectively, and are illustrated by the lines *ab* and *cd* in Fig. 2. Table 2 shows the results of calculations. The values of $F(^{90}\text{Sr}) = 0.035$ (pCi/cm²) and $F(^{210}\text{Po}) = 0.052$ (pCi/cm²) are thus obtained for the amounts of excess ^{90}Sr and excess ^{210}Po injected into the atmosphere by the 25th Chinese nuclear explosion and the 18 May 1980 eruption of Mount St. Helens, respectively. This means that the total amount of excess ^{210}Po released from the 1980 eruption of Mount St. Helens was about 1.5 times the amount of ^{90}Sr released from the 16 October

1980 nuclear explosion. Kuroda *et al.* (1984) have estimated the total amount of ^{90}Sr injected into the atmosphere by the 16 October 1980 nuclear explosion was about $8 \times 10^4 \text{ Ci}$. They used the values of bi-monthly concentrations of ^{90}Sr , ^{210}Pb and ^{210}Po in rain and found that the total excess ^{210}Po deposition at Fayetteville, Arkansas, caused by the 1980 eruption of Mount St. Helens was about 1.5 times the value of the amount of ^{90}Sr released by the 16 October 1980 nuclear explosion. These results indicate that the total amount of excess ^{210}Po released into the atmosphere by the eruption of Mount St. Helens was approximately $8 \times 10^4 \times 1.5 = 1.2 \times 10^5 \text{ Ci}$.

Acknowledgments—This investigation was supported by the National Science Foundation under grants ATM 82-06718 and ATM 84-07618.

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