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Atmospheric Deposition of Beryllium 7 in the Chesapeake Bay Region

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Bulk (combined wet and dry) atmospheric deposition of the cosmogenic isotope ⁷Be was monitored at Solomons, Maryland (38°19'N, 76°27'W), from March 1986 to November 1987. The observed deposition of ⁷Be supported an inventory ranging from 2 to 4 dpm (disintegrations per minute)/cm². The supported inventory and monthly fluxes of ⁷Be showed maxima in late spring (March–May). The peak in ⁷Be deposition was very similar in 1986 and 1987 despite 1986 having a very dry spring. It thus appears that given even modest amounts of precipitation, there is strong seasonality in the atmospheric deposition of ⁷Be near 40°N. The timing and magnitude of the spring peak in ⁷Be deposition are in close agreement with estimates of the seasonal variation in the injection of stratospheric air into the troposphere, suggesting that on a monthly time scale the bulk deposition of ⁷Be closely reflects the tropospheric inventory of ⁷Be.

INTRODUCTION

Beryllium 7 is a short-lived (half-life equals 53.3 days) cosmogenic isotope formed by spallation of N and O, predominantly in the stratosphere [Arnold and Al-Salih, 1955; Lal et al., 1958, 1960]. Once formed, ⁷Be rapidly attaches to particles in the atmosphere. Scavenging by precipitation is the main process bringing ⁷Be to the Earth's surface [Matsunami et al., 1979; Olsen et al., 1985]. The initial focus of investigations of ⁷Be was its usefulness as a tracer of atmospheric mixing (the works by Arnold and Al-Salih [1955], Lal et al. [1958], Gustafson et al. [1961], Junge [1963], Viezee and Singh [1980], and Dutkiewicz and Husain [1985] are a very limited sample of the rich literature). More recently, ⁷Be has come to be recognized as a good tracer of mixing and transport in aquatic and sedimentary systems [Silker, 1972; Young and Silker, 1974; Krishnaswami et al., 1980; Rice, 1986; Olsen et al., 1986; Martin et al., 1986; Dibb and Rice, 1989]. A program monitoring ⁷Be deposition at Solomons, Maryland, on the western shore of the Chesapeake Bay, was initiated primarily to provide a measure of the atmospheric input for a study of the dynamic behavior of ⁷Be in the Chesapeake Bay [Dibb and Rice, 1989]. The results of the first 19 months of the monitoring program also yield considerable insight into the temporal variability of ⁷Be deposition and how that variability is related to atmospheric processes.

METHODS AND MATERIALS

Bulk atmospheric deposition (combined wet and dry) of ⁷Be was collected in a fiberglass tank (surface area equals 2.685 m²) deployed 10 m above ground on the roof of a building at the Chesapeake Biological Laboratory (CBL) in Solomons, Maryland (38°19'N, 76°27'W). Atmospheric deposition was sampled by periodically draining the tank into polyethylene buckets, rinsing the tank thoroughly two times (with first 0.2 N HNO₃ and then distilled water) and then adding the rinses to the sample. Stable Be yield tracer (100–800 μg in dilute

HNO₃) and Fe (as a 20% wt/vol solution of FeCl₃ in dilute HNO₃) were stirred into the sample, which was then allowed to stand for at least 3 hours. Beryllium 7 and Be yield tracer were concentrated by coprecipitation with Fe by raising the pH to 9.5 ± 0.5 with NH₄OH or NaOH [Olsen et al., 1986]. The precipitate was allowed to settle at least overnight, the bulk of the overlying water was carefully decanted, and the precipitate was collected on 9.0-cm-diameter Whatman GF/C filters by vacuum filtration. The filters were mounted in 10-cm × 1.5-cm polystyrene petri dishes for gamma spectrometry.

Initially, sampling was intended to be monthly; however, beginning in April 1986, samples were taken after each major precipitation event or weekly during dry periods to monitor fission products from the reactor accident at Chernobyl in April 1986 [Devell et al., 1986; Thomas and Martin, 1986; Aoyama et al., 1986; Bondiotti and Brantley, 1986; Dibb and Rice, 1988]. By December 1986 the sampling program had evolved to an approximately biweekly schedule.

In 1987 there were heavy snowfalls on January 22 and 25. Snow collected in the tank was transferred to buckets and melted and then was treated as outlined above. To examine the variability of ⁷Be flux in a single precipitation event, additional samples were collected by carefully digging a trench in an area apparently unaffected by drifting, and then sampling from the wall of the trench at three depth intervals: 0–7, 7–14, and 14–26 cm. A prominent ice crust layer at 14 cm separated snow from the two storms. These snow samples were also melted and treated as above.

An additional attempt to determine variability of ⁷Be flux in a single precipitation event was made in July 1987. The collector was drained and cleaned prior to an afternoon thunderstorm. Two samples were collected from the storm, which had a total duration of 50 min. In the first 30 min, 1890 mL of rain was collected, and 1660 mL was obtained in the final 20 min. These samples were processed as above.

All ⁷Be determinations were made using a reversed electrode coaxial intrinsic germanium detector and a computer-based multichannel analyzer. Detector calibration and calculation of sample activities followed the procedure outlined by Larsen and Cutshall [1981]. Prior to March 1987, National Bureau of Standards sediment standard 4350B, in various geometries to match samples, was the primary standard. Counting efficiency at the 477.6-keV ⁷Be photopeak was calculated by linear interpolation between the 351.9-keV ²¹⁴Pb photo-

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TABLE 1. Beryllium 7 Activity in the Collector at the End of Each Sampling Interval

Sampling Date	Precipitation, mm	⁷ Be Activity, dpm/cm ²
March 19, 1986		collector deployed
April 7, 1986	6.35	0.568 ± 1.1*
April 23, 1986	14.73	0.768 ± 1.4
May 8, 1986	0.00	0.089 ± 0.5
May 16, 1986	2.54	0.179 ± 0.8
May 20, 1986	11.43	0.259 ± 1.0
May 22, 1986	45.47	1.046 ± 1.7
May 29, 1986	0.00	0.059 ± 0.5
June 5, 1986	6.35	0.149 ± 0.9
June 12, 1986	0.76	0.084 ± 0.6
June 20, 1986	0.25	0.021 ± 0.4
June 27, 1986	2.79	0.307 ± 1.2
July 2, 1986	35.81	0.421 ± 1.7
July 10, 1986	4.57	0.208 ± 1.1
July 17, 1986	22.61	0.390 ± 1.9
July 24, 1986	12.45	0.349 ± 1.1
July 31, 1986	4.32	0.152 ± 1.5
Aug. 7, 1986	14.48	0.407 ± 2.3
Aug. 15, 1986	11.94	0.181 ± 1.3
Aug. 18, 1986	66.80	0.472 ± 1.7
Aug. 21, 1986	32.51	0.194 ± 1.3
Aug. 28, 1986	27.43	0.238 ± 1.5
Sept. 3, 1986	12.70	0.178 ± 1.4
Sept. 8, 1986	19.05	0.256 ± 1.6
Sept. 25, 1986	0.00	0.020 ± 0.6
Oct. 6, 1986	1.52	0.039 ± 0.9
Oct. 14, 1986	34.54	0.303 ± 2.3
Oct. 18, 1986	27.94	0.338 ± 2.4
Nov. 6, 1986	16.26	0.480 ± 3.1
Nov. 12, 1986	23.37	0.438 ± 3.0
Nov. 19, 1986	8.38	0.109 ± 1.7
Dec. 1, 1986	27.43	0.474 ± 4.4
Dec. 11, 1986	57.91	0.541 ± 4.9
Dec. 23, 1986	34.54	0.441 ± 5.1
Dec. 29, 1986	70.61	0.187 ± 4.1
Jan. 14, 1987	50.04	0.262 ± 3.9
Jan. 27, 1987	85.85	0.386 ± 3.0
Feb. 19, 1987	7.37	0.058 ± 2.0
March 5, 1987	70.36	0.508 ± 5.4
March 19, 1987	22.86	0.527 ± 4.0
April 7, 1987	45.72	0.809 ± 6.9
April 21, 1987	27.43	0.505 ± 6.2
May 5, 1987	104.39	0.968 ± 11.4
May 21, 1987	75.95	1.067 ± 3.3
June 3, 1987	30.23	0.317 ± 2.3
June 18, 1987	51.05	0.388 ± 3.1
June 29, 1987	44.20	0.394 ± 3.1
July 13, 1987	12.45	0.279 ± 2.9
July 27, 1987	9.65	0.229 ± 2.9
July 29, 1987	10.67	0.304 ± 2.6
Aug. 25, 1987	27.43	0.630 ± 4.2
Sept. 8, 1987	11.18	0.279 ± 6.0
Sept. 14, 1987	93.98	0.661 ± 3.3
Sept. 28, 1987	29.97	0.412 ± 6.7
Oct. 20, 1987	42.67	0.581 ± 2.3
Nov. 3, 1987	19.81	0.254 ± 1.9

Activities are decay corrected to sampling date, and uncertainties represent 1σ counting uncertainty.

*Uncertainties only are multiplied by 1000.

peak and the 661.7-keV ¹³⁷Cs photopeak. After March 1987 a ⁷Be standard from the Environmental Protection Agency allowed direct determination of ⁷Be counting efficiency. Activities of all samples were corrected for decay between the sampling and counting dates.

After counting, the Fe(OH)₃ precipitate was redissolved in 0.2 N HNO₃, and the Be content was determined by atomic

absorption spectrophotometry to estimate the recovery of ⁷Be in the concentration process.

RESULTS

Between April 1986 and September 1987, 52 samples of atmospheric deposition were collected. Beryllium 7 activities in the collector at the end of each interval ranged from ~0.02 to ~1.1 dpm (disintegrations per minute)/cm² (Table 1). An atmospherically supported ⁷Be inventory $I(t_i)$ at each sampling time t_i has been calculated from these data and the equation

$$I(t_i) = I(t_{i-1})e^{-\lambda\Delta t_i} + D(t_i) \quad i = 0, 1, 2, \dots \quad (1)$$

where λ is the radioactive decay constant of ⁷Be (0.013 d⁻¹), $\Delta t_i = t_i - t_{i-1}$ is the interval length (in days), and $D(t_i)$ is the ⁷Be that accumulated in the tank between sampling times t_{i-1} and t_i (in disintegrations per minute per square centimeter) (Figure 1). The calculated inventory assumed $I(t_0 = 0) = 0$ (i.e., no ⁷Be inventory present before the collection program began); hence it underestimates inventories for the first 100 days or so of the record. After this initial run-up period the atmospherically supported ⁷Be inventory ranged from 2 to 4 dpm/cm² (uncertainty ~6%).

Monthly amounts of precipitation and ⁷Be deposition were derived by summing the values obtained for the shorter sampling intervals (after correcting ⁷Be activities to the middle of each month). Over the length of this investigation the average monthly precipitation was 80 mm, and ⁷Be deposition averaged 1.13 dpm/cm². Although ⁷Be monthly deposition was only weakly correlated with precipitation ($r = 0.34$), the temporal trends in deposition and precipitation showed an interesting relationship (Figure 2). In 1986 and 1987 the bulk of ⁷Be deposition occurred in spring and early summer, in spite of near-drought conditions in the spring of 1986. Relatively wet months in late summer (August 1986 and September 1987) resulted in above average deposition of ⁷Be, but even heavier precipitation in the winter (December and January) carried down relatively little ⁷Be.

Beryllium 7 specific activities in the serial samples of single precipitation events decreased with time (Figure 3). The higher ⁷Be concentrations in the rain samples may indicate that rain, particularly in thunderstorms, is more effective than snow in stripping ⁷Be from the atmosphere, but it seems likely that some of the increase reflects seasonal differences in the abun-

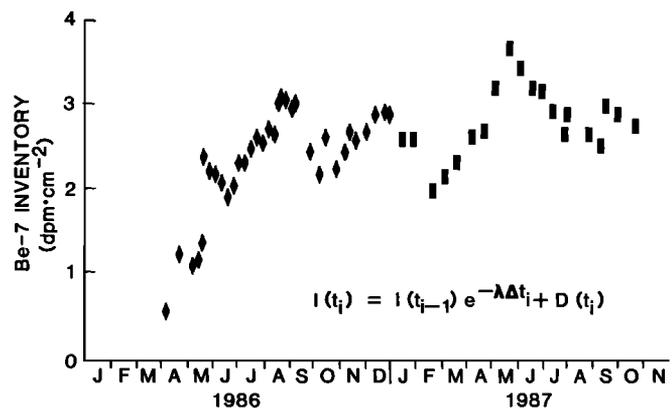


Fig. 1. Calculated atmospherically supported ⁷Be inventory. If an uncertainty of 2% is assumed for the flux during each interval $D(t_i)$, the uncertainty in the calculated inventory is ~6%.

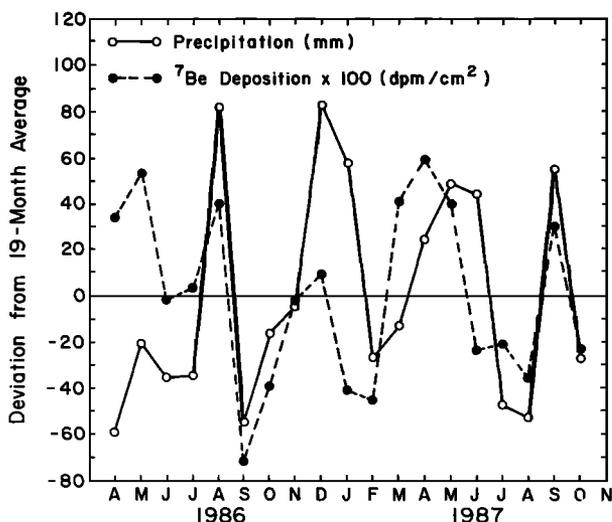


Fig. 2. Monthly deviations of ^7Be deposition ($\times 100$) and precipitation from the average for 19 months. Monthly values were calculated by summing deposition for the shorter sampling intervals after decay correcting the ^7Be deposition to the middle of each month. Average precipitation was 80 mm, and ^7Be averaged 1.13 dpm/cm 2 . Uncertainty in the flux of ^7Be is about 5%.

dance of ^7Be in the air masses scavenged by the precipitation (see discussion below).

The early "washing out" of atmospheric ^7Be revealed by the serial sampling of single precipitation events suggests that the concentration of ^7Be in precipitation should decrease as the amount of precipitation increases. The volume of water in the collector was not routinely measured, but an estimate of the volume of precipitation delivered to the tank during each sampling interval was made from the rainfall measured at a weather station approximately 50 m from the collector. Beryllium

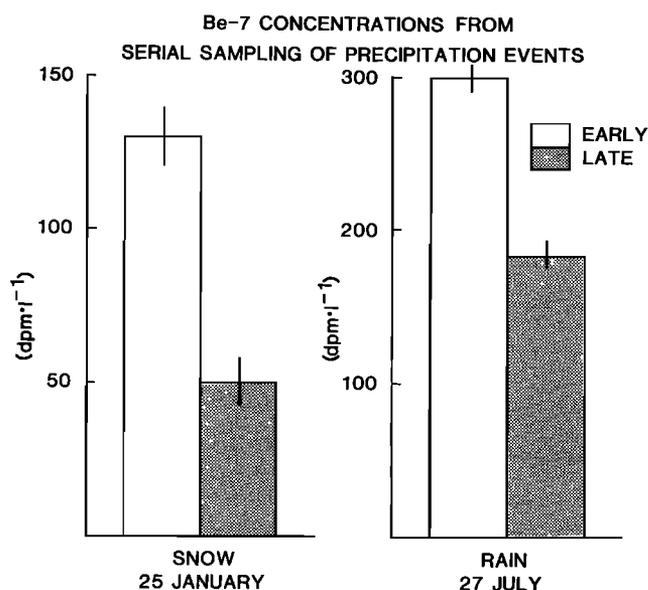


Fig. 3. Beryllium 7 concentrations in serial samples of single precipitation events. For the snow pit samples the 0- to 7-cm-depth interval represents snow falling late in the January 25 storm, and the 7- to 14-cm interval is from the early part of the storm. The early sample of the July 27 rain shower was collected in the first 30 min (volume equals 1890 mL), and the late sample was from the final 20 min of the shower (volume equals 1660 mL). Vertical bars represent 1σ counting uncertainties.

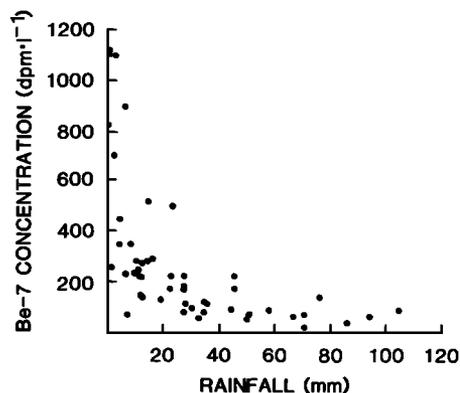


Fig. 4. Estimated ^7Be concentration in precipitation plotted against the amount of precipitation. Concentrations were approximated by dividing the activity of ^7Be in the collector at the end of each sampling interval by the amount of rain measured at a weather station ~ 50 m from the collector during the same interval. The estimates are clearly low, since no corrections for decay between the precipitation event(s) and sampling date were made.

^7Be concentrations estimated from the observed flux and the approximated volume of precipitation must be underestimates of the actual concentration in the precipitation, since no corrections were made for decay between the precipitation event(s) and sampling. In general, sampling intervals were longer, and hence the underestimation of ^7Be concentration was greater, during those periods when little rain or snow fell. However, this crude approach clearly shows ^7Be concentrations to decrease with increasing amount of precipitation (Figure 4).

DISCUSSION

The deposition of ^7Be measured at Solomons, Maryland, compares well with published values based on total deposition into bucket type collectors. The 19 monthly fluxes allow estimation of a yearly deposition of 13.6 dpm/cm 2 , which falls in the range of 4.3–22.7 dpm/cm 2 tabulated for the latitude range 19°–52°N [Turekian *et al.*, 1983]. The average monthly flux of 1.13 dpm/cm 2 is higher than the average (0.69 dpm/cm 2) reported for two locations in Japan during 1977–1979 [Matsunami *et al.*, 1979] but is very similar to the averages during 1982–1984 at Oak Ridge, Tennessee (1.0 dpm/cm 2), and Norfolk, Virginia (1.1 dpm/cm 2) [Olsen *et al.*, 1985]. The calculated atmospherically supported inventory in Maryland is also similar to that in Tennessee and Virginia [Olsen *et al.*, 1985] and to what was measured in the Swiss Alps during 1982–1983 [Dominik *et al.*, 1987]. It is notable that Olsen *et al.* [1985] observed a pronounced seasonality in the deposition of ^7Be , with 40–45% of the annual total occurring during March–May.

Bulk atmospheric deposition of ^7Be provides an integrated signal of at least two interrelated processes: the accumulation of ^7Be in the atmosphere and its removal by dryfall and precipitation. Where the climate is not extremely arid, precipitation generally carries down most of the ^7Be [Matsunami *et al.*, 1979; Olsen *et al.*, 1985]. However, it is clear that a given amount of precipitation can contain widely varying amounts of ^7Be (Figures 2–4). On a precipitation event basis, variability in both the concentration and total flux of ^7Be can result from differences in the type of event (intensity, duration, etc.) and, presumably, the length of time between precipitation events from a single air mass. However, the apparent constancy in

TABLE 2. Monthly Average Concentration of ^7Be in Ground Level Air in Sterling, Virginia, as Reported by EML for the Period May 1970 to April 1974

Month	Concentration, dpm/m ³
Jan.	0.24
Feb.	0.26
March	0.27
April	0.31
May	0.30
June	0.26
July	0.27
Aug.	0.26
Sept.	0.23
Oct.	0.25
Nov.	0.20
Dec.	0.21

the empirical deposition velocity of ^7Be [Young and Silker, 1974, 1980; Crecelius, 1981] suggests that there is little or no seasonal trend in the efficiency of the processes removing ^7Be from the atmosphere. It thus appears that the seasonality in ^7Be deposition is due to variation in the amount of ^7Be available to the removal processes.

Theoretical [Lal *et al.*, 1958] and experimental [Lal *et al.*, 1960; Rama, 1963; Bhandari *et al.*, 1966; Bhandari, 1970] studies have established the large-scale features of ^7Be production in the atmosphere. For the yearly and shorter time scales of interest here there are no temporal variations in the production of ^7Be . In a given latitude range the most important variation in ^7Be production is a strong increase with altitude to a maximum at about 12–15 km [Bhandari *et al.*, 1970]. This results in stratospheric air usually having ^7Be concentrations roughly an order of magnitude higher than upper tropospheric air and approximately 50 times greater than ground level air [Dutkiewicz and Husain, 1979, 1985]. The long residence time of stratospheric aerosols (14 months [Reiter, 1975] compared to the 53.3-day half-life of ^7Be) suggests that much of the ^7Be reaching the ground is produced in the troposphere [Turekian *et al.*, 1983], but periodic increases in the fraction of stratospheric air mixing below the tropopause greatly increase the inventory of ^7Be in the troposphere [Gustafson *et al.*, 1961; Dutkiewicz and Husain, 1979, 1985] and should be reflected in increased deposition of ^7Be .

Gustafson *et al.* [1961] suggested that the annual rising of the tropopause in summer would incorporate some stratospheric air into the troposphere as well as increase the tropospheric production rate by an increase in the cosmic ray flux reaching the troposphere. The combination of these effects was estimated to increase the tropospheric inventory of ^7Be by about 50%. The tropopause near 40°N reaches its maximum altitude in mid to late summer [Reiter, 1975], so this mechanism might account for the above average deposition of ^7Be in August 1986 and September 1987, but it does not seem capable of explaining the greater than twofold increase in deposition between January and May 1987. (It should be noted that precipitation in these two months was above average and very similar at 129 mm in January and 137 mm in May (Figure 2).)

Another mechanism bringing stratospheric air into the troposphere is disruption or folding of the tropopause. A number of studies, using ^7Be , ozone, and fission products from bomb tests as tracers of stratospheric air, have demonstrated that injection of stratospheric air into the troposphere by this

type of process has a pronounced annual cycle [Junge, 1963; Rangarajan and Gopalakrishnan, 1970; Dutkiewicz and Husain, 1979, 1985; Staley, 1982]. Near 40°N the mixing of stratospheric air into the troposphere reaches a maximum, at 2–3 times the annual minimum, during the late spring (late April to early June) [Staley, 1982; Dutkiewicz and Husain, 1985]. Such an increase in the tropospheric inventory of ^7Be would explain the similarity in the spring peaks in deposition in Maryland in 2 years of greatly different precipitation and would appear to be a major factor causing similar spring peaks in ^7Be deposition in Virginia and Tennessee [Olsen *et al.*, 1985].

It would obviously be of considerable value to be able to compare ^7Be deposition to a measure of its tropospheric inventory over time. The time and expense of repeatedly determining ^7Be concentrations at different heights in the atmosphere preclude the routine monitoring of ^7Be tropospheric inventories, but the time series of ground level ^7Be concentrations measured at the Environmental Measurements Laboratory's (EML) network of stations do provide at least some information about tropospheric levels of ^7Be . From May 1970 to April 1974, EML maintained a station at Sterling, Virginia (38°58'N, 77°25'W), which is probably close enough to CBL to be affected by the same mesoscale and large-scale meteorologic processes. The ^7Be concentrations in ground level air at Sterling showed an annual pattern, with monthly average values peaking near 0.3 dpm/m³ in April and May and minimum values of about 0.2 dpm/m³ occurring in November and December (Table 2). It is not certain that the pattern seen during the early 1970s at Sterling has continued through 1987, but the monthly deposition of ^7Be at CBL during this study was more strongly correlated with the average ground level air concentrations measured at Sterling 10–15 years previously ($r = 0.51$) than it was with precipitation measured at CBL at the same time ($r = 0.34$).

CONCLUSIONS

The atmospheric deposition of ^7Be exhibits wide variability on time scales ranging from within a single precipitation event to seasonal changes. This is a result of the interaction between the processes building up and depleting the tropospheric inventory of ^7Be . Deposition of ^7Be over monthly intervals appears to closely reflect large-scale variations in the tropospheric inventory of ^7Be . Sampling during 2 years with markedly different precipitation patterns revealed that while precipitation is very important in carrying down ^7Be , the usual spring rains in northern mid-latitudes are greatly in excess of the amount of precipitation required to bring down most of the ^7Be present in the troposphere during this season. It thus appears that previously reported high correlations between ^7Be deposition and precipitation reflect, to some extent, the coincidence of annual patterns of precipitation and variability in the tropospheric inventory of ^7Be rather than a direct dependence of ^7Be deposition on the amount of precipitation. The timing of the annual peak in ^7Be deposition in Maryland, Virginia, and Tennessee suggests that the spring maximum in injection of stratospheric air into the troposphere is the predominant cause of the cyclic pattern of ^7Be deposition seen in this region.

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