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Stratospheric and Tropospheric Components of ^{7}Be in Surface Air

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^{7}Be and ^{90}Sr concentrations were measured simultaneously in samples from NASA's Global Atmospheric Sampling Program. The samples were collected from January 1978 through June 1979 at 9 to 12 km and between 11° and 60°N. ^{7}Be concentrations ranged from 150 to 6900 fCi/m^3, while ^{90}Sr correspondingly varied from 10 to 320 fCi/m^3. The average ^{7}Be concentration in the samples collected in the lower stratosphere was 4500 fCi/m^3. In the upper troposphere, ^{7}Be concentrations were generally less than 1000 and often only a few hundred fCi/m^3. In situ production of ^{7}Be in the upper troposphere at ~11 km was estimated at 340 fCi/m^3. Since ^{90}Sr is only present in the atmosphere because of nuclear testing, except immediately after such tests, ^{90}Sr in the troposphere is due only to the subsidence of stratospheric air. Since atmospheric processes are not expected to alter $^{7}Be/^{90}Sr$ ratios during transport, surface ^{90}Sr and stratospheric $^{7}Be/^{90}Sr$ ratios can be used to estimate ^{7}Be transported from the stratosphere. Our analyses show that on an annual basis the stratosphere contributed ~25% of the observed ^{7}Be concentration. During spring and summer months, the stratospheric component can approach 40%, while on a daily basis this can be even larger.

INTRODUCTION

^{7}Be (half-life, 53.3 days) is a naturally occurring gamma emitter in the atmosphere that is produced by cosmic ray spallation reactions with nitrogen and oxygen. The production rate is maximum in the stratosphere, at about 20 km, and decreases approximately exponentially with decreasing altitude [*Lal and Peters*, 1967]. The stratospheric mean residence of 14 months [*Reiter*, 1975] exceeds the time required (6 times the half-life) to establish steady state equilibrium between ^{7}Be production and its removal by radioactive decay and atmospheric removal. Stratospheric ^{7}Be concentrations are thus expected to remain unchanged from year to year, except for small variations (<10%) resulting from solar modulation of cosmic rays. In the troposphere, however, decreased production rates and rapid mixing (mean residence time, 30 days) [*Beck and Kuroda*, 1976; *Gavini et al.*, 1974] result in ^{7}Be concentrations that are a hundredfold smaller than those in the stratosphere.

The large concentration gradient and the fact that ^{7}Be is produced naturally unaffected by nuclear detonations, have led to attempts to use it as a tracer of stratospheric air in the troposphere. *Husain et al.* [1977] showed that pulses of high ^{7}Be concentrations at Whiteface Mountain (altitude, 1.5 km), New York, were associated with air masses of stratospheric origin, as indicated by potential vorticities. Isentropic trajectories traced the air masses to ~8 km. *Reiter et al.* [1978] have also attributed the observed high ^{7}Be concentration at Zugspitze (altitude, 3 km), West Germany, to transport from the stratosphere. *Viezee and Singh*[1979] showed that the seasonal variation of ^{7}Be concentrations for sites in North America followed the occurrence of low-pressure troughs at 500 mbar. These troughs are areas most frequently associated with stratospheric intrusions. No quantitative estimates of stratospheric ^{7}Be contributions were made because in situ tropospheric production could not be determined with meaningful accuracy.

This is even more true when the attempts are made at sampling sites well within the planetary boundary layer [*Ludwick et al.*, 1976; *Husain et al.*, 1979; *Wolff et al.*, 1978]. ^{7}Be concentrations in the lower stratosphere near the tropopause average ~5000 fCi/m^3 (all activities are corrected to standard pressure and temperature) [*Dutkiewicz and Husain*, 1979]. Monthly average ^{7}Be concentrations at most ground-level sites are ~100 fCi/m^3. Thus only a small contribution of stratospheric air is needed to produce a large change in observed ^{7}Be concentration in surface air.

Large variations in surface ^{7}Be are observed in daily measurements [*Husain et al.*, 1977, 1979; *Reiter et al.*, 1978]. Peak concentrations of ~200 fCi/m^3 are often associated with the back side of high-pressure systems in the eastern United States and with enhanced surface ozone concentrations, both consistent with subsidence of air from aloft. Less frequently the ^{7}Be concentrations are higher, ~500 fCi/m^3.

One cannot assume a priori, however, that observed trophospheric ^{7}Be concentrations are entirely due to transport from the stratosphere. Long irradiation times (in which an air mass must remain undisturbed) are necessary to produce ^{7}Be concentrations>100 fCi/m^3. For example, based on calculated production rates of *Lal and Peters* [1967] at 5-km irradiation times of 7, 15, 30, and 65 days produce ^{7}Be concentrations of 80, 160, 300, and 500 fCi/m^3, respectively. In the dynamic troposphere, irradiation times in excess of a week are perhaps unrealistic. *Turekian et al.* [1983] have argued that, at the surface, tropospheric production determines the major flux rate of ^{7}Be, modulated regionally by the delivery of stratospheric air.

It is therefore important to determine unambiguously the tropospheric and stratospheric ^{7}Be contributions in surface air. Clearly, another tracer specie produced solely in either the stratosphere or trophosphere is needed for this purpose. Among the various long-lived nuclides, fissiogenic ^{90}Sr appears to be an excellent candidate. Its presence in tropospheric air is due entirely to transport from the stratosphere, except for periods of 2-4 weeks immediately after a nuclear detonation. Furthermore, since ^{7}Be and ^{90}Sr are both associated with submicron-sized particles, their fates during transport from the stratosphere are expected to be similar. A knowledge of $^{7}Be/^{90}Sr$ ratios in the stratosphere and surface ^{90}Sr concentrations could thus be used to determine stratospheric ^{7}Be contributions to surface air.

Using air filters obtained from NASA's Global Atmospheric Sampling Program (GASP), we have determined ^{7}Be and ^{90}Sr concentrations in the upper troposphere and lower stratosphere for the mid-northern latitudes. In this paper we present our data and use it to apportion the stratospheric and tropospheric components of ^{7}Be in surface air.

SAMPLING AND ANALYSIS

The GASP program utilized commercial 747 jetliners equipped with computerized aerosol sampling devices and in situ measure-

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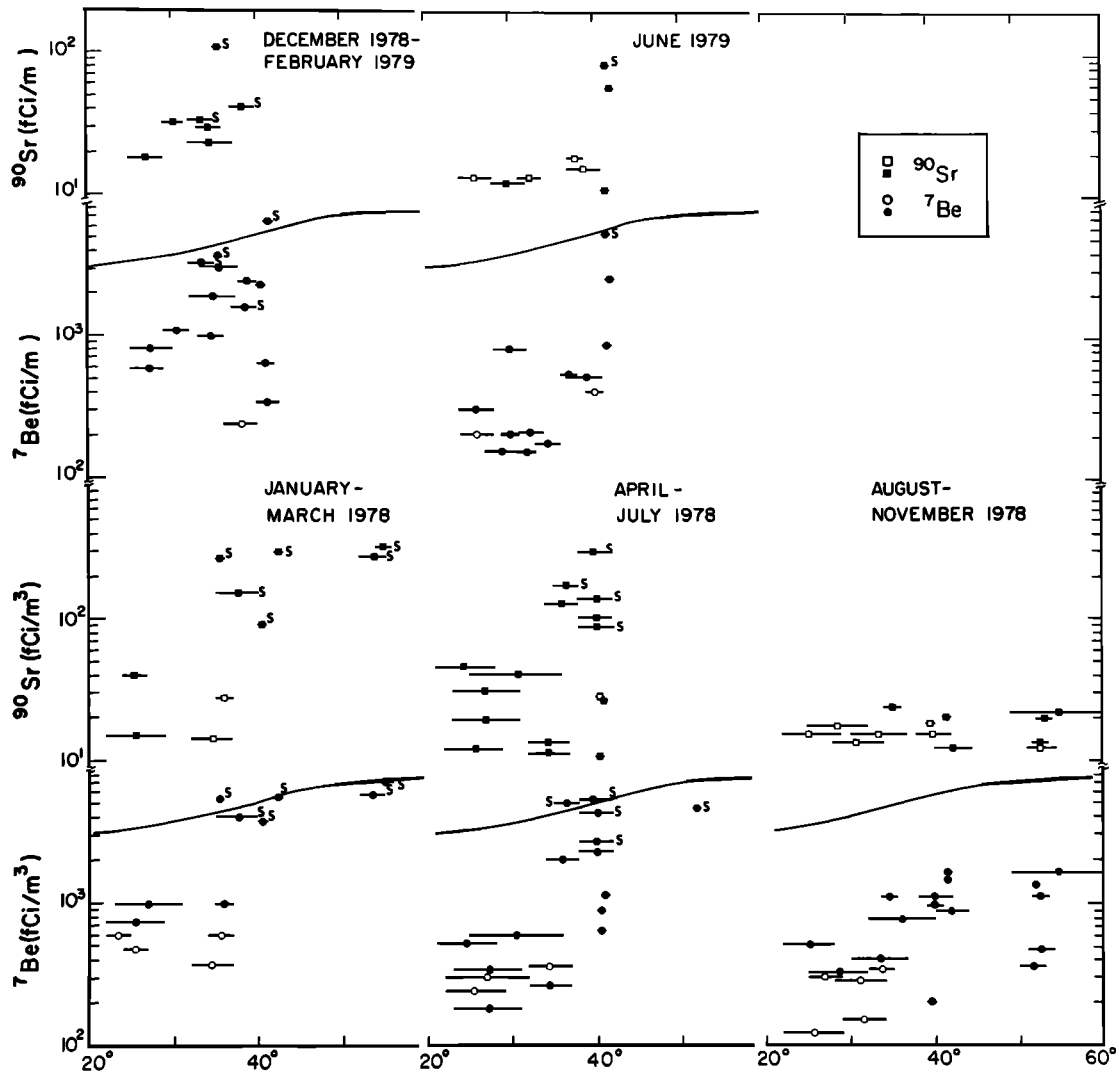

Fig. 1. ^{7}Be and ^{90}Sr concentrations in GASP samples collected at 10-12 km from January 1978 to June 1979 (s, stratospheric samples). The curve is the theoretical ^{7}Be activity for a static atmosphere at 11 km [*Lal and Peters*, 1967]. The horizontal bars correspond to the latitude range sampled and the open squares and circles correspond to upper limit concentrations of ^{90}Sr and ^{7}Be, respectively.

ment systems for a variety of trace species, including O_3 [*Perkins and Gustaffson*, 1976; *Tiefermann*, 1979]. Details of the sampling and ^{7}Be measurements have been presented earlier [*Dutkiewicz and Husain*, 1979]. Only the salient features will be discussed here, along with the procedure for ^{90}Sr measurements.

The sampling altitude ranged from 9 to 12 km; however, most samples were collected at around 11 km. Samples from both the lower stratosphere and upper troposphere were collected. The sampling inlet was automatically covered when the aircraft descended below 6.1 km to avoid contamination of the system with lower tropospheric air.

Approximately 100 aerosol samples were collected on IPC 1478 filters between January 1978 and June 1979. Exposure was generally for 2 hours, covering a distance of 1100 km and filtering ~120 standard cubic meters of air. The samples were collected between 11° and 60°N, but the latitude coverage was nonuniform: 34 samples for 25-35°N and 45 for 35-45°N, but only three samples below 25°N and seven above 45°N.

The samples were received from NASA sealed in polyethylene and were gamma counted directly to determine ^{7}Be activity [*Dutkiewicz and Husain*, 1979]. One quarter of each filter was used for ^{90}Sr determination by standard radiochemical procedures. The final precipitate was the ^{90}Sr daughter ^{90}Y in the form of yttrium oxalate, which was deposited on Whatman 42 filters precoated with an asbestos slurry. The deposit was ignited to yttrium oxide, weighed to determine the chemical yield, sealed with Mylar tape, and beta counted several times on low-background gas flow proportional counters built in this laboratory (background count rate, 0.45 ± 0.05 counts/min). ^{90}Sr activity was determined by a least squares fit of the ^{90}Y data to a 2.67-day half-life (^{90}Y was in equilibrium with ^{90}Sr prior to counting). Corrections for chemical recovery and counter efficiency were applied.

RESULTS AND DISCUSSION

The ^{7}Be measurements of the GASP samples for January-June 1978 have been reported earlier [*Dutkiewicz and Husain*, 1979]. These ^{7}Be measurements are reproduced and extended through June 1979 in Figure 1 along with ^{90}Sr measurements. For comparison the theoretical ^{7}Be concentration at 11 km corresponding to a static atmosphere [*Lal and Peters*, 1967] is also shown. The samples designated as stratospheric (S) were collected above the tropopause

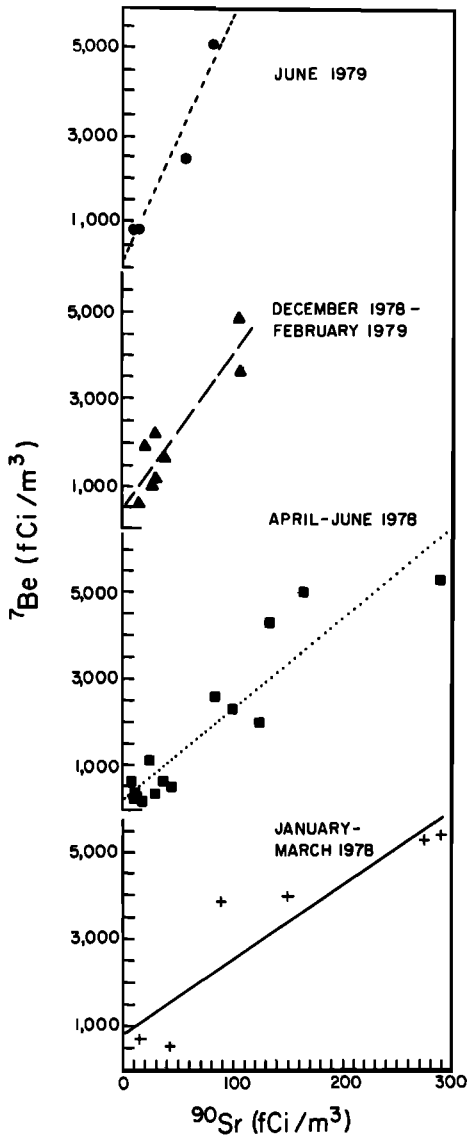

Fig. 2. Scatter plots of ^{7}Be versus ^{90}Sr for data displayed in Figure 1. The lines are linear least squares fits to the data.

height as supplied by NASA (E. A. Lezberg, personal communication, 1981). These tropopause heights were determined by time and space interpolation from the National Meteorological Center's archived data field by using the Flattery global analysis method [e.g., *Papathokas and Briehl*, 1981].

^{7}Be concentrations (Figure 1) range from 150 to 6900 fCi/m^{3}, while ^{90}Sr varied from 10 to 320 fCi/m^{3}. The samples designated as stratospheric averaged 4500 fCi ^{7}Be/m^{3} and generally had ^{7}Be concentrations close to the saturation activity. This average is slightly lower than the 5000 fCi/m^{3} reported earlier on the basis of samples collected during the first 6 months of this study [*Dutkiewicz and Husain*, 1979]. Samples collected in the upper troposphere generally had ^{7}Be concentrations <1000 fCi/m^{3}, and many were <200 fCi/m^{3}; the average was 580 fCi/m^{3}. This large variability in tropospheric ^{7}Be concentrations is most likely due to varying mixtures of stratospheric and tropospheric air, either by direct transfer of air across the tropopause or by a sampling artifact as the aircraft crossed over the tropopause during the long sampling distances.

Although the variations of ^{7}Be and ^{90}Sr concentrations were similar (Figure 1), the concentrations of ^{90}Sr relative to that of ^{7}Be decreased during the period. No significnt introduction of ^{90}Sr into the stratosphere had occurred since the nuclear detonation of November 17, 1976. Two nuclear devices were detonated during our study period (the 23rd and 24th Chinese tests of March 15 and December 14, 1978), but both devices were of low yield (20 kt), and their debris would remain predominantly in the troposphere. Even if some of the ^{90}Sr from such low-yield detonations did reach the stratosphere, it will not significantly alter the ^{90}Sr concentration. Our ^{90}Sr measurements in the stratosphere show no significant increases subsequent to these detonations.

^{7}Be concentrations for the latitudes 25-42°N are plotted against ^{90}Sr in Figure 2. Since ^{90}Sr concentration in the stratosphere was being decreased by exchange with the troposphere, the plots are limited to no more than 3-month periods. Shorter periods would have been preferable, but we had to compromise to obtain sufficient data for meaningful correlations. The lines in Figure 2 correspond to linear least squares fit to the data. The correlation coefficients (r) ranged from 0.92 to 0.96 and were all significant at $P<0.05$ [*Bevington*, 1969]. The high correlations suggest that a significant component of the ^{7}Be in the tropospheric samples originated like ^{90}Sr in the stratosphere.

Since the stratospheric ^{7}Be component would be correlated with ^{90}Sr, we assume that the ^{7}Be intercept corresponds to the tropospheric background. The intercepts in Figure 2 are (from bottom to top) 700 ± 550, 200 ± 170, 380 ± 370, and 80 ± 570 fCi/m^{3}. The uncertainties are based on the errors in the individual measurements and the quality of the least squares fit as described by *Bevington* [1969]. These uncertainties are sufficiently large to preclude any meaningful evaluation of seasonal trends. The average of these intercepts, 340 fCi/m^{3}, accounts for 60% of average ^{7}Be concentrations in samples collected below the tropopause, 580 fCi/m^{3}.

The slope of the least squares lines increase steadily with time from 17.1 for January-March 1978 to 56.2 for June 1979. This increase in the ^{7}Be/^{90}Sr ratio must be due to loss of ^{90}Sr to the troposphere as the ^{7}Be concentrations in the stratosphere remain at equilibrium.

The ^{7}Be/^{90}Sr ratios in stratospheric samples (35-55°N) are shown in Figure 3. Over the 18-month period the ^{7}Be/^{90}Sr ratio increased from ~18 to 60. During periods without large atmospheric nuclear tests, the stratospheric ^{90}Sr inventory decreases exponentially as a result of exchange with the troposphere [*Krey et al.*, 1974]. Since ^{7}Be is in equilibrium in the stratosphere, the ^{7}Be/^{90}Sr ratio would be expected to increase exponentially with an exponent equivalent to the inverse of the stratospheric mean residence time. The exponential least squares fit in Figure 3 has a correlation coefficient of 0.85, in support of the discussion above. This curve corresponds to a stratospheric mean residence time of 16 months. No measurements of the ^{90}Sr stratospheric inventory were made by the Environmental Measurement Laboratory during this study period [*Leifer et al.*, 1981]. However, this residence time is comparable to other estimates [e.g., *Reiter*, 1975].

Since ^{7}Be and ^{90}Sr are both attached to submicron-sized particles, no differential removal is expected, and the stratospheric ^{7}Be component in surface air can be determined as the product of the stratospheric ^{7}Be/^{90}Sr ratio and the surface ^{90}Sr concentration. Monthly average ^{7}Be/^{90}Sr ratios in the lower stratosphere at 35-55° can be determined for 1978 and 1979 from the exponential curve in Figure 3. During this period, ^{90}Sr and ^{7}Be concentrations were measured at four Environmental Measurement Laboratory (EML) surface sites within this latitude zone: Moosonee, Canada (51°N); New York, New York (41°N); Rocky Flats, Colorado (40°N); and

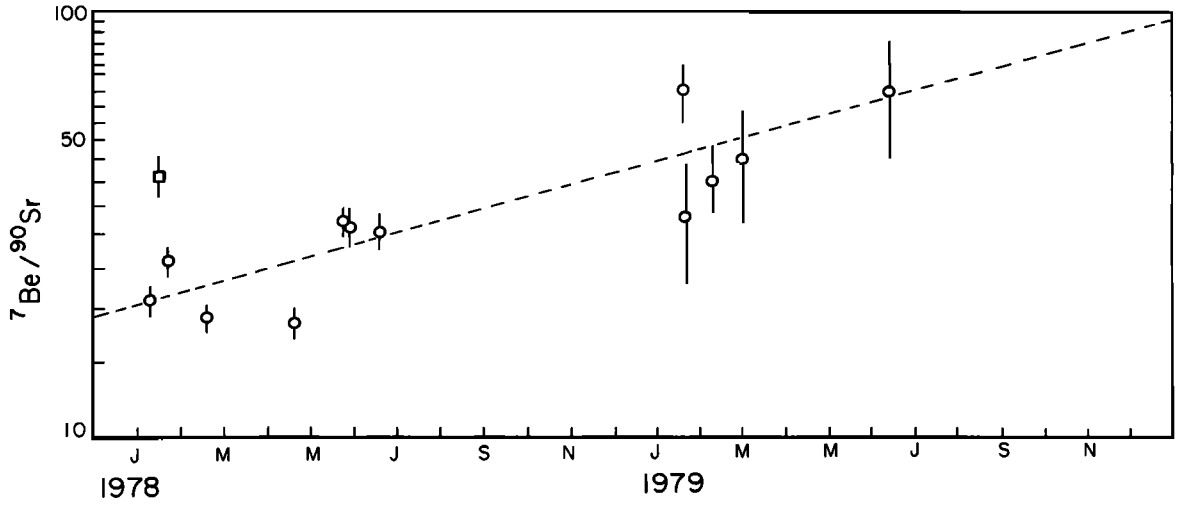

Fig. 3. ^{7}Be/^{90}Sr ratios for samples collected in the stratosphere January 1978 to June 1979. Vertical bars are uncertainties (± 1 standard deviation) based on counting statistics. The dashed line is an exponential least squares fit to the data, excluding the single point designated by □.

Livermore, California (38°N) [*Feely et al.*, 1981]. The resulting stratospheric and tropospheric components of ^{7}Be for these sites were calculated.

A few caveats are in order:

Since travel times from stratosphere to troposphere are estimated to be a few days to a week [*Danielsen*, 1980], radioactive decay of ^{7}Be during subsidence was not included. This may result in an overestimation of the stratospheric ^{7}Be component by 10%. Unfortunately, our measurements of the ^{7}Be/^{90}Sr ratios have been limited to January through June of each year, and we must rely on the least-squares fit to deduce the ratios for July through December. The data in Figure 3 give the hint of a seasonal cycle for ^{7}Be/^{90}Sr ratios in the lower stratosphere (maximum in January, minimum in March or April). If this were true, the July through December ^{7}Be/^{90}Sr ratios are likely to be larger than that indicated by the exponential curve. We do not, however, have the necessary data to determine this with confidence. At any rate this will not have any significant effect on the annual average stratospheric and tropospheric components deduced. The surface ^{90}Sr concentrations for October through December are so small that even if the ^{7}Be/^{90}Sr ratios were a factor of 2 higher it would not increase the annual average stratospheric ^{7}Be component by more than a few percentage points.

The Chinese nuclear tests were likely to introduce ^{90}Sr into the lower troposphere. ^{89}Sr/^{90}Sr ratios in precipitation at Fayetteville, Arkansas (36°N), indicated debris from the 23rd test was present in March and April 1978 and debris from the 24th test in December 1978 and January 1979 [*Burchfield et al.*, 1982]. Therefore we did not calculate the stratospheric and tropospheric ^{7}Be components for these months, except at Moosonee. Since the test site was at 40°N and the debris generally flows eastward and southward in the northern hemisphere [*Rangarajan and Eapen*, 1981], Moosonee should not have been significantly affected by these tests. The measured ^{90}Sr values at Livermore in December 1978 may have been similarly influenced by the Chinese test. However, we have used this data because it is the only December data available for that site. The calculated stratospheric ^{7}Be component does not appear to be unexpectedly high, so any ^{90}Sr contribution from the 24th Chinese test at that site must be small.

At all sites the minimum stratospheric ^{7}Be component was observed during October to December and the maximum in May and June. For example the stratospheric and tropospheric components for 1978 and 1979 at Livermore and New York are shown in Figure 4. The tropospheric component at Moosonee is similar to that shown for New York, only slightly lower (annual average, 55 and 76 fCi/m^3, respectively). The tropospheric component at Rocky Flats, on the other hand, is similar to that at Livermore (minimum concentration in early spring and maximum in fall). The annual averages (in fCi/m^3) for these sites were: Rocky Flats, 108; Livermore, 117. The difference in the seasonal profiles perhaps reflects regional differences in the degree of vertical mixing in the troposphere.

Annual averages for the sites are summarized in Table 1. On an annual basis the stratosphere accounts for 23-27% of the observed

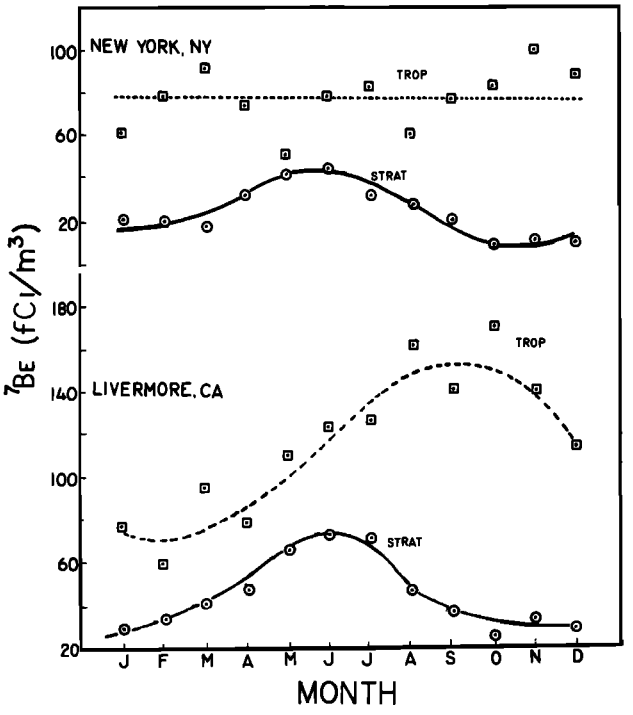

Fig. 4. Stratospheric and tropospheric components of ^{7}Be concentration for New York, N.Y., and Livermore, California. Free-hand curves have been added to aid the eye.

^{7}Be concentrations. The relative stratospheric contributions are nearly equal, even though the observed concentrations vary by more than a factor of 2.

While tropospheric production dominates surface ^{7}Be concentration on an annual basis, a strong seasonal variation in stratospheric ^{7}Be concentration was evident in Figure 4. Because of the similarity in the relative stratospheric component at all four sites, as evidenced by the nearly equal annual averages, we have combined the data into the average seasonal profile shown in Figure 5. The stratospheric ^{7}Be varies from near 10% in fall to around 40% in May and June. On a daily basis the stratospheric component may even be larger. For example, although the average ^{7}Be concentration at Whiteface Mountain, New York, is ~100 fCi/m^3, daily concentrations are highly variable and have reached 500 fCi/m^3 [*Dutkiewicz and Husain*, 1979]. Similar large pulses of ^{7}Be have been observed at Zugspitze [*Reiter et al.*, 1978]. Based on potential vorticities, *Husain et al.* [1977] demonstrated that these pulses are associated with stratospheric air masses. Further *Husain et al.* [1979] showed that short sampling periods (6 hours) often yield higher concentrations that are masked in longer sampling periods. *Danielsen* [1968] has suggested that stratospheric intrusions associated with tropopause-level cyclogenesis is the principal mechanism for transfer of stratospheric air into the troposphere. These stratospheric intrusions occur in the vicinity of the jet stream at isolated times and places, primarily in spring and summer, and can be relatively narrow [*Danielsen and Mohnen*, 1968; *Shapiro*, 1980; *Johnson and Viezee*, 1981]. Thus as these intrusions reach the boundary layer, it is expected that stratospheric air (^{7}Be) will not be delivered uniformly with time, but rather in short pulses.

SUMMARY

Measurements of ^{7}Be and ^{90}Sr are presented for the lower stratosphere and upper troposphere from GASP samples collected from January 1978 through June 1979. ^{7}Be concentrations in the lower stratosphere averaged 4500 fCi/m^3, compared to 580 below the tropopause. While the stratospheric concentrations of ^{7}Be remained relatively constant, ^{90}Sr decreased. ^{7}Be and ^{90}Sr were highly correlated (r = 0.92-0.96). Average in situ tropospheric production of ^{7}Be at around 11 km was estimated at 340 fCi/m^3 or ~60% of the observed concentration.

From ^{7}Be/^{90}Sr ratios in the stratosphere and ground-level measurements of ^{90}Sr and ^{7}Be at EML sites (38-51°N) the annual average stratospheric and tropospheric components of ^{7}Be in surface air were estimated. The tropospheric components (in fCi/m^3) were 55 at Moosonee, 76 at New York, 108 at Rocky Flats, and 117 at Livermore. Stratospheric components were 21, 24, 32, and 43 fCi/m^3, respectively. Thus the stratosphere accounted for 23-27% of the surface ^{7}Be on an annual basis. The stratospheric component was not delivered uniformly throughout the year; it accounted for around 40% of the observed ^{7}Be concentrations during late spring but only 10% during fall.

TABLE 1. Annual Average ^{7}Be Concentrations and The Stratospheric and Tropospheric Components at Four Mid-latitude Sites.

Sites	^{7}Be, fCi/m^3			Stratospheric ^{7}Be, %
	Total	Stratospheric	Tropospheric	
Moosonee (51°N)	76	21	55	27
New York (41°N)	100	24	76	24
Rocky Flats (40°N)	140	32	108	23
Livermore (38°N)	160	43	117	27

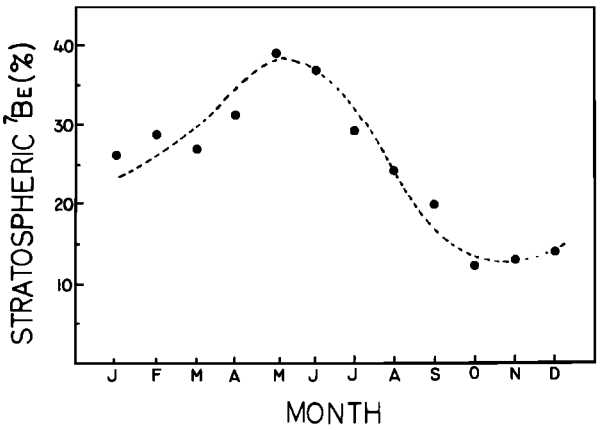

Fig. 5. Seasonal profile of the relative stratosphere ^{7}Be component for mid-latitudes (38-51°N). Free-hand curve added to aid the eye.

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