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THE DISTRIBUTION OF BERYLLIUM-7 IN THE TROPOSPHERE: IMPLICATIONS ON STRATOSPHERIC/TROPOSPHERIC AIR EXCHANGE

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Abstract. The distribution of cosmogenic beryllium-7 (^{7}Be) in the lower troposphere is described based on the analysis of long-term (1973-1979) ground level ^{7}Be measurements. ^{7}Be is used as a tracer of stratospheric air to suggest that the annual mean outflow of stratospheric air in the northern hemisphere can be 20 to 30% higher than in the southern hemisphere. This asymmetry is maximum during the northern hemisphere spring and reaches its peak in April. Over North America, the ^{7}Be concentrations show a high positive correlation (linear coefficients of 0.8 to 0.9) with the occurrence of tropospheric low-pressure troughs over several latitude belts, thus supporting the postulate that tropopause folding events, associated with low-pressure troughs, are the dominant stratospheric/tropospheric (S/T) exchange mechanism. It is demonstrated that S/T air exchange shows a distinct seasonal variation that is highly dependent on the latitude. At midlatitudes (40-50°N) over North America, the S/T air exchange is maximum in summer; at low altitudes (20-30°N), it is maximum in winter and early spring. The study suggests a mechanistic model that can be used to characterize S/T air exchange at any geographical location using routinely available meteorological information.

Introduction

Because of the important role that ozone (O_3) plays in the chemistry of the troposphere, its tropospheric budget has been the subject of several studies (e.g., Danielsen and Mohnen, 1977; Singh et al., 1978; Dutkiewicz and Husain, 1979). Specifically, the contribution of natural O_3 to ground-level concentrations has been of interest (Budiansky, 1980). In the classical view, the major source of natural O_3 in the troposphere is the stratosphere. The stratospheric O_3 contribution is dictated to a large degree by two principal factors: (1) O_3 content and its variability in the lower stratosphere, and (2) the exchange of air between the stratosphere and the troposphere. The same considerations apply to tropospheric NO_x that have also been proposed to have a stratospheric source (Kley et al., 1980). While the stratospheric abundance of such trace species (O_3, NO_x) is relatively well characterized, major uncertainties in the nature and magnitude of stratospheric/tropospheric (S/T) air exchange remain. A better understanding of the S/T exchange in the two hemispheres is also necessary for a qualitative validation of photochemical models that postulate *in-situ* O_3 production from natural $CH_4/CO/NO_x$ precursors (Fishman and Crutzen, 1978).

To identify the presence of O_3-rich stratospheric air at ground level, several studies have demonstrated the feasibility of using beryllium-7 (^{7}Be) as a natural radioactive tracer of upper-atmospheric air (Ludwick et al., 1976; Reiter et al., 1976; Husain et al., 1977; Wolff et al. 1979). In this note, we report on the analyses of ^{7}Be measurements in surface air that are available for the North and South American continents, on their interpretation in terms of the difference in S/T air exchange between the two hemispheres, and on the seasonal behavior of this exchange over North America.

Analysis and Interpretation

The ^{7}Be data used in the analysis are average monthly values of concentration in surface air published by the Environmental Measurements Laboratory of the U.S. Department of Energy for 1973 to 1979 (Toonkel, 1980). As is demonstrated below, the surface air ^{7}Be measurements are representative of the surface boundary layer, which can extend to at least 5 km into the lower troposphere, depending on geographical location. Supporting meteorological data are obtained from microfilm copies of the daily 500-mbar upper-air charts analyzed by the National Meteorological Center of NOAA for the years 1975 to 1979. A master data file was created and all data were computer analyzed.

Figure 1 shows the latitudinal distribution of ground-level ^{7}Be (mean and standard deviation, 1973 to 1979) across North and South America for the northern hemisphere spring, summer, fall, and winter. The profiles extend from Moosenee, Ontario (51.27°N 80.50°W) to Balboa, Panama (8.97°N 79.57°W) and across the equator along the west coast of South America to Punta Arenas, Chile (53.13°N 70.88°W). The 12 surface measurement locations used in the analysis are listed in Table 1. Other locations with ^{7}Be measurements [e.g., Mauna Loa, Hawaii (19.47°N 155.60°W) and Easter Island (27.17°S 109.43°W)] could have been considered for inclusion into a mean meridional profile. However, these stations and others are judged to be too far removed from our longitude interval or to represent subtropical ocean areas where local deposition events may not be compatible with those of over-land areas at the same latitude (Noshkin, 1969). Also, several stations had too short (<3 yrs) a period of data record to be included in the analysis.

The latitudinal ^{7}Be profile over North America (NA) in Figure 1 shows a bimodal distribution. This bimodality is primarily the result of including the data from Richmond, California, which is the only available measurement location within the 30-40°N latitude interval. It may be argued that Richmond represents too much of a jump westward from the continental environs of Helena, Montana, and Rocky Flats, Colorado, and that the possible existence of a longitudinal gradient is projected to a non-representative latitudinal gradient. The validity of the Richmond measurements in the NA profile of Figure 1 is supported by the data listed in Table 2. The relatively small seasonal values at

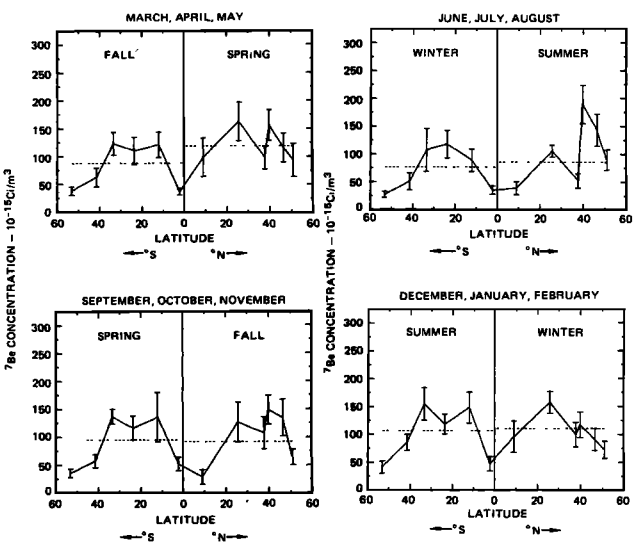

Fig. 1. Seasonal and latitudinal variations of ground-level ^{7}Be concentration across the North American and South American continents.

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TABLE 1. Locations with surface air ^{7}Be measurements used in the analysis of Figure 1.

Station	Location		Station Elevation MSL	Mean-Monthly Data Points
	Latitude	Longitude	(m)	Available
Moosenee, Canada	51.27°N	80.50°W	10	70
Helena, Montana	46.60°N	112.00°W	1187	56
Rocky Flats, Colorado	40.00°N	105.18°W	1830	72
Richmond, California	37.93°N	122.33°W	20	45
Miami, Florida	25.82°N	80.28°W	7	60
Balboa, Panama	8.97°N	79.57°W	23	41
Guayaquil, Ecuador	2.17°S	79.87°W	7	71
Lima, Peru	12.02°S	77.13°W	13	72
Antofagasta, Chile	23.62°S	70.27°W	31	62
Santiago, Chile	33.47°S	70.70°W	520	67
Puerto Montt, Chile	41.45°S	72.95°W	7	69
Punta Arenas, Chile	53.13°S	70.88°W	35	68

Richmond are compatible with independent measurements available at the nearby Lawrence Livermore Laboratory for 1973 to 1979 (Silver et al., 1979). More importantly, however, these two sets of data on the west coast compare favorably with the seasonal values obtained from east coast measurements available at Sterling, Virginia (38.97°N 77.42°W) for 1970 to 1975, except for the summer season. At Richmond and Livermore, the summer value of ^{7}Be concentration is significantly smaller than at Sterling. As clarified below in connection with Figure 3, we attribute this difference to the large latitudinal gradient in the frequency of occurence of tropospheric low-pressure troughs in the vicinity of 40°N during summer. Thus, the bimodality in the ^{7}Be profile over NA is maintained and the two maxima between 20-30°N and 40-50° N remain identifiable when the Richmond measurements are replaced by those at Sterling.

The midlatitude (40-50°N) maximum in Figure 1 peaks during summer; it then decreases significantly during fall and reaches a minimum in winter. As demonstrated below, this feature reflects the seasonal latitudinal migration of the tropospheric low-pressure troughs over NA. The subtropical (20-30°N) maximum is out of phase with the midlatitude maximum, peaks in winter and spring, and is most likely influenced by both the seasonal migration of the low-pressure troughs and the descending (subtropical) branch of the mean meridional Hadley circulation. Over South America (SA), the ^{7}Be profile is also largely bimodal but its seasonal variabilty is much less pronounced than over NA. Two relatively stationary maxima between 10-20°S and 30-40°S are present, except during winter (June, July, August), when the tropical maximum disappears.

Since ^{7}Be is also partly synthesized in the troposphere, not all ^{7}Be is considered to be of stratospheric origin. The relative minimum values analyzed near the equator are close to the theoretical ground-level values of about 45 x 10^{-15} Ci/m^3 due to tropospheric synthesis (Bhandari et al., 1966).

The data of Figure 1 also show a relatively large mean interhemispheric gradient in ^{7}Be directed from North to South America during the spring of the northern hemisphere (March, April, May). In fall and winter, however, no significant gradient is apparent. This feature is shown more explicitly in Figure 2. The upper frame of this figure illustrates the monthly variation of ^{7}Be concentration averaged from 0° to 51°N (North American average) and from 0° to 53°S (South American average). The lower frame shows the ratio of the North American to the South American average. Before computing the ratios, a value of 45 x 10^{-15} Ci/m^3 was subtracted from the hemispheric averages, assuming the well-known hemispheric symmetry in tropospheric ^{7}Be synthesis at ground level (Bhandari et al., 1966). The ratios are indicated with an ± 1 standard deviation based on the $\pm$ 20% maximum error in the ^{7}Be measurements. The largest mean ratio (2.0) occurs in April (gradient directed from north to south) and the smallest (0.8) occurs in September (gradient directed from south to north). The annual mean (dashed line in Figure 2B) implies a relatively small net gradient of 22% directed from north to south. When the data at Richmond, California (122.33°W) in the profiles of Figure 2 are replaced by the available measurements at Sterling, Virginia (77.42°W), this net gradient increases to about 30%.

Assuming no large variations in the long-term, large-scale ^{7}Be concentrations along a given latitude (i.e., zonal homogeneity), the data of Figure 2 can be interpreted to suggest that the outflow of stratospheric air into the troposphere is significantly higher in the northern hemisphere during spring, but is only marginally higher when averaged over the entire year when compared to the southern hemisphere.

Meteorological conditions involving a folded tropopause structure (stratospheric intrusion) have been identified as a key mechanism for S/T exchange. A number of studies over the last decade have shown that these stratospheric intrusions are specifically associated with low-pressure troughs in the troposphere (Danielsen, 1968; Shapiro, 1978; Viezee et al., 1979). The ^{7}Be data were analyzed to verify this dominant mechanism of S/T air exchange over NA. The results are presented in Figure 3. The left-hand frames show the seasonal variation of the monthly average ^{7}Be concentration for three typical locations across the United

TABLE 2. Comparison between seasonal values of surface air ^{7}Be measurements* from locations across the United States at comparable latitude but greatly different longitude.

Station	Lat./Lon.	Winter	Spring	Summer	Fall
Richmond, California	37.97°N 122.33°W	100	97	52	106
Livermore, California	37.68°N 121.77°W	102	115	87	111
Sterling, Virginia	38.97°N 77.42°W	107	133	118	104

*^{7}Be measured in units of 10^{-15} Ci/m^3

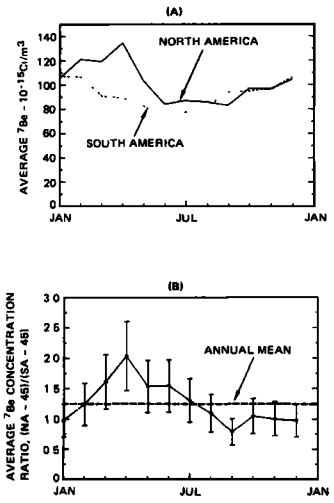

Fig. 2. Monthly variation of ground-level ^{7}Be Concentration. (A) Averaged from 0° to 51°N (North American average—solid line) and averaged from 0° to 53°S (South American Average—dashed line). (B) Ratio of North American (NA) to South American (SA) average (mean and standard deviation).

States. The right-hand frames show the seasonal variation in the number of occurrences of 500-mbar low-pressure troughs in three latitude belts from the Canadian border to the Gulf of Mexico. Each belt covers ten degrees of latitude and is considered of adequate geographical extent to contain a stratospheric intrusion event (e.g., Viezee et al., 1979). The 500-mbar troughs were identified in terms of the area of maximum wind speed and wind direction change. These areas in the trough are most frequently associated with stratospheric air intrusions (Viezee et al., 1979). At Helena, Montana (46.60°N 112.00°W), the monthly average ^{7}Be concentration is largest during summer (June through September), when the mean position and the maximum frequency of occurrence of 500-mbar troughs is between 40° and 50°N. At Miami, Florida (25.82°N 80.28°W), however, the ^{7}Be concentrations become large during winter and early spring when the position of the 500-mbar trough (and the polar front jet stream) is along the southern U.S. border (20° to 30°N). At Richmond, California (37.93°N 122.33°W), the annual cycle of ^{7}Be concentration measurements reflects most strongly the rapid decrease in the summer frequency of occurrence of 500-mbar troughs from the large relative maximum in the north to the total absence in the south. The summer minimum at Miami, Florida is not as low as suggested by the total absence of low-pressure troughs, and secondary transport mechanisms may be operative in this subtropical area.

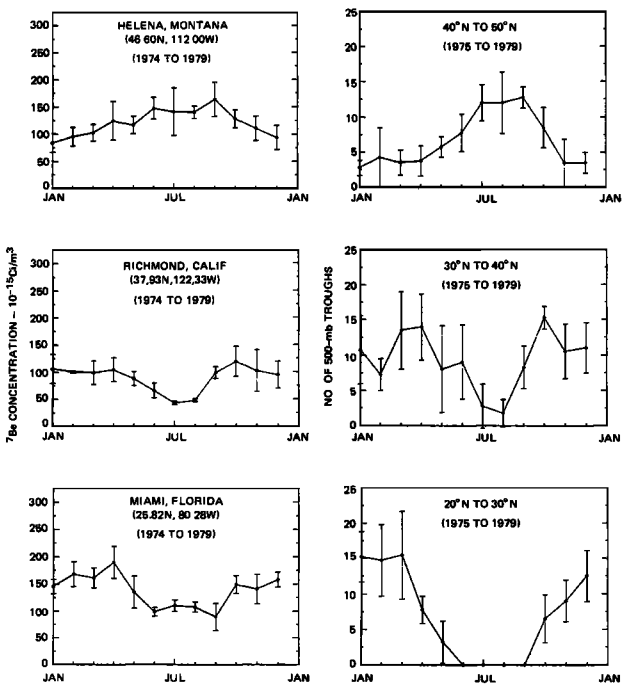

Fig. 3. Seasonal and latitudinal variation of ground-level ^{7}Be (left frames) and the frequency of occurence of 500-mb low-pressure troughs (right frames) across the continental United States.

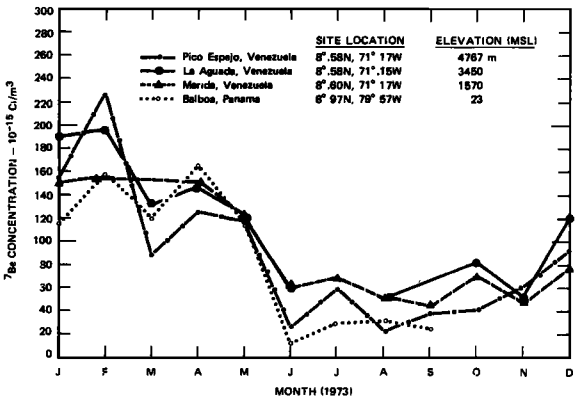

Fig. 4. Similarity in the measurements of ^{7}Be concentration in surface air at four sites with a gradual increase in elevation at approximately the same geographical location.

A linear regression between the monthly mean ^{7}Be concentrations and the monthly mean number of 500-mbar low-pressure trough occurrences in the corresponding latitude belts gives positive correlation coefficients of 0.87 for Helena, Montana, 0.85 for Richmond, California, and 0.78 for Miami, Florida. Thus, the data analysis of Figure 3 provides rather conclusive evidence of the close relation between the S/T air intrusion mechanism and ^{7}Be measurements in surface air.

The foregoing discussion is based on the premise that atmospheric mixing is efficient enough so that ^{7}Be concentrations in surface air are representative of those in the lower troposphere. This postulate is supported by the data of Figure 4, where it is shown that, for this selected geographical location, no significant difference exists between the measured ^{7}Be concentrations from 0 to nearly 5000 m above sea level throughout the entire year. Limited data from other locations support this observation.

Conclusion

Analyses of ^{7}Be data provide support for both the validity of the meteorological S/T exchange mechanism and the ability of ^{7}Be to act as a tracer of stratosphere air. Recognizing that even when as much as 40% of surface-air ^{7}Be is of troposheric origin, the major features of stratospheric ^{7}Be are maintained and are clearly identifiable. Annually averaged S/T exchange in the NH is somewhat (20 to 30%) greater than in the SH. The seasonal behavior of S/T exchange over NA is highly dependent upon latitude. At 40 to 50°N, the S/T exchange is at its maximum in summer while, at the same time, it is at its minimum between 20 to 30°N. Any analysis of S/T O_3 (or NO_x) exchange must superimpose the variabilties in the stratospheric abundance of O_3(or NO_x) on the corresponding seasonal and latitudinal variabilities in the S/T mass exchange. It is proposed that the observed summer O_3 maximum at midlatitudes (Singh et al., 1978) is at least partially attributable to the enhanced S/T air exchange over the midlatitude region.

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