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Be-10/Be-7 tracer of atmospheric transport and stratosphere-troposphere exchange

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10\textsuperscript{Be}/7\textsuperscript{Be} tracer of atmospheric transport and stratosphere-troposphere exchange

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The 10\textsuperscript{Be}/7\textsuperscript{Be} ratio is a sensitive tracer of atmospheric transport and stratosphere-troposphere exchange (STE). Data from five NASA aircraft field missions (PEM: West A and B; Tropics A; SONEX; and SUCCESS) have been assembled to produce the largest data set of \textsuperscript{10}Be, \textsuperscript{7}Be, and their ratio collected to date (>300 samples). Ratios near 0.60 are indicative of tropospheric air with little stratospheric influence, while higher ratios are found in stratospheric air. Samples from the lower stratosphere were all collected within 2.5 km of the tropopause and had ratios >1.27. Of these lower stratosphere samples only 16% had ratios in excess of 3.0, suggesting that higher ratio air resides away from the tropopause. Seasonality observed in the \textsuperscript{10}Be/\textsuperscript{7}Be ratios results from the downwelling of air with elevated ratios from higher in the stratosphere in the spring and summer (midlatitudes) and from the decay of \textsuperscript{7}Be during descent in the winter polar vortex (high latitudes). Our results illustrate the complexity of STE and some of the mechanisms through which it occurs, including tropopause folding, mixing associated with subtropical jets, and the effect of synoptic systems such as hurricanes and northeasters. The \textsuperscript{10}Be/\textsuperscript{7}Be ratio provides important information beyond that which can be derived from studies that rely on chemical mixing ratios alone.

INDEX TERMS: 0305 Atmospheric Composition and Structure: Aerosols and particles (0345, 4801); 0368 Atmospheric Composition and Structure: Troposphere—constituent transport and chemistry; 3362 Meteorology and Atmospheric Dynamics: Stratosphere/troposphere interactions; KEYWORDS: Aerosols, radioisotopes, beryllium, stratosphere-troposphere exchange


1. Introduction

\textsuperscript{[2]} Stratosphere-troposphere exchange is a complex process that appears to be neither geographically nor seasonally uniform. For example, Folkins et al. (1999) have recently suggested that in the tropics air from the marine boundary layer at Samoa (14°S) is only convected to a height of 14 km, 3 km short of the tropopause height. They observed a clearly defined O\textsubscript{3} chemopause at this low altitude and they proposed that above this altitude, in the upper troposphere, O\textsubscript{3} is produced in situ and then ascends slowly into the stratosphere. This observation is in contrast to that of others who have suggested that elevated O\textsubscript{3} below the tropopause is due to stratospheric air that has mixed down into the troposphere [e.g., Tuck et al., 1997; Fujiwara et al., 1998].

\textsuperscript{[3]} Another mechanism for STE is via tropopause folds in midlatitudes [Danielsen, 1968; Danielsen and Hipskind, 1980; Kritz et al., 1991]. Such folds occur near the jet stream [Danielsen, 1968; Danielsen et al., 1987] when a layer of air is extruded from the lower stratosphere on the cyclonic (poleward) side of the jet. This stratospheric air enters the troposphere beneath the core of the jet and moves equatorward and downward toward the surface boundary layer [Danielsen et al., 1987]. Tropopause folds can also bring tropospheric air into the stratosphere, making the lower stratosphere a transition layer between the troposphere and the stratospheric overworld [Danielsen, 1968].

\textsuperscript{[4]} Seasonality in the source of stratospheric air entering the troposphere can occur even though the actual intensity of STE does not vary. The influence of the subtropical jet is minimal on the lowermost stratosphere, hence actual STE is fairly consistent year-round [Dethof et al., 2000]. However, as the jet strengthens in winter, exchange between the stratospheric overworld and lower stratosphere is suppressed [Dethof et al., 1999, 2000]. As a result the stratospheric air entering the troposphere in winter includes less overworld air than stratospheric air entering in spring and summer when
the weakened jet allows more overworld air to descend. Measurements made in September at 34.5°N show most of the air in the lowermost stratosphere had been transported quasi-isentropically from the troposphere [Ray et al., 1999], while observations in May and June (at 34.5°N and 64.5°N, respectively) showed the air had been predominantly advected down from the overworld [Ray et al., 1999].

[5] Synoptic systems may also affect STE. Detjof et al. [2000] suggest that mixing may be enhanced at the end of the Atlantic storm track in winter and downstream of the Asian monsoon anticyclone in summer. Enhanced 32P/32P ratios reported by Benitez-Nelson and Buesseler [1999] were attributed to synoptic systems such as hurricanes and northeasters drawing down stratospheric air (signified by high ratios) into the troposphere. Although, beryllium bound to aerosols tends to be washed out of the troposphere by precipitation, Aldahan et al. [2001] found peaks in 7Be concentrations at the end of heavy precipitation events. They attributed this to stratospheric influx.

[6] The complexity of the mixing processes associated with STE and the seasonality of the effects have made the process difficult to characterize fully. 10Be/7Be and the 10Be/7Be ratio are important tracers that can be used to provide new information about STE. 10Be and 7Be are produced by cosmic ray interactions with atmospheric oxygen and nitrogen. In 1981, Raisbeck et al. suggested that the ratio of the cosmogenic radioactive isotopes of beryllium, 10Be/7Be, could be used as a probe of atmospheric transport. Since then only a handful of studies have reported 10Be in either aerosols [Raisbeck et al., 1981; Dibb et al., 1994] or precipitation [Monaghan et al., 1986; Brown et al., 1989]. In this paper, we present the largest data set of 10Be, 7Be, and their ratio collected to date. Both isotopes are produced by cosmic ray spallation, predominantly in the midlatitude stratosphere, and then are rapidly taken up by aerosols. Aerosols can remain aloft in the stratosphere for years. Because of the vastly different half-lives of 10Be and 7Be, 1.5 × 106 years and 53 days, respectively, the ratio of these species will increase in the stratosphere as the aerosols age.

[7] Eventually, stratospheric aerosols are transported into the troposphere where they are removed from the atmosphere via scavenging and deposition. The residence time of aerosols in the troposphere is about 30–40 days [Shapiro and Forbes-Resha, 1976], which is insufficient to allow the 10Be/7Be ratio to increase appreciably above its production ratio. Since there is no evidence for isotopic fractionation during either aerosol uptake or tropospheric removal processes, the ratio will retain its stratospheric signature even if mixing ratios change significantly.

[8] In an isolated air mass where the only loss is due to radioactive decay, 10Be will accumulate with negligible decay, while 7Be will approach secular equilibrium where decay balances production. Secular equilibrium for 7Be will be reached in approximately 8 months. Once 7Be has reached secular equilibrium, the 10Be/7Be ratio will increase at a rate that depends only on the 10Be production rate. Assuming 10Be is produced at 60% of the rate of 7Be [Dibb et al., 1994; Nagai et al., 2000], the ratio will increase over time from its initial value of about 0.6 to a ratio of 5.8 after 2 years and to 11.6 after 4 years. In the stratosphere, three factors affect the observed ratio. First, if an air parcel moves from a region of high production to a region of lower production the 10Be/7Be ratio will increase as 7Be decays to the new, lower secular equilibrium value. Second, if air mixes between regions of differing age (e.g., the stratosphere and troposphere) the resulting air mass will have an intermediate ratio that reflects the relative influence of the respective source regions. Third, gravitational settling of aerosols-bound beryllium results in mid and upper stratospheric ratios that are lower than would be expected from the air parcel residence time alone, because removal from the source region keeps the ratio nearer the production ratio. These attributes make the beryllium ratio a useful tracer for stratosphere-troposphere exchange (STE).

[9] Note that there is a fourth factor to be considered. Although, for the purposes of the discussion here, the production ratio of 10Be/7Be is assumed to be constant at 0.60, it may in fact vary somewhat between the stratosphere and troposphere, and also as a function of latitude. Nagai et al. [2000] calculated the production rates of 10Be and 7Be and found the global average production rate in the stratosphere of 0.44 during solar minimum, while in the troposphere they found 0.67. They suggest multiplying these by 0.8–0.9 to better estimate long-term averages, which results in a production rate of 0.35–0.40 in the stratosphere, and 0.53–0.60 in the troposphere. Here, we will use the higher tropospheric value as a typical production ratio. This is a conservative mean value to distinguish observations of higher stratospheric ratios from lower tropospheric ones. Further, Nagai et al. [2000] calculations indicated an increase in the production ratio with latitude, for the stratosphere, this increase was observed above 80°, but in the troposphere, the increase was seen above 50°. Nonetheless, even at high latitudes, the production ratio does not exceed 1.0. Hence, ratios above this value always reflect increasing age in an air mass as 10Be accumulates while 7Be maintains its secular equilibrium.

[10] A recent modeling experiment has shown the region of the stratosphere with the highest 10Be/7Be ratios lies between 20 and 40 km in the tropical stratosphere, with the peak ratios exceeding 8.0 between 25 and 30 km [Bergmann et al., 2001]. Nearer the tropopause, the ratios are lower, approximately 2.0. The seasonal behavior described above suggests that the 10Be/7Be ratio should increase in the lower stratosphere in the spring and summer under the influence of descending stratospheric overworld air with higher 10Be/7Be ratios. In the fall and winter, as the influence of this source wanes, it is expected the ratio will decrease as tropospheric influx becomes dominant.

[11] Using the ratio of 10Be/7Be, Dibb et al. [1994] found that stratospheric influence on the Arctic troposphere was nearly continuous year-round. More importantly, they showed that the ratio peaked in late summer, indicating that the greatest stratospheric influence occurred at that time even though the mixing ratios of the two isotopes independently were at their minima. The mixing ratios alone would suggest a tropospheric air mass with little or no stratospheric input. Yet the isotope ratio clearly revealed a stratospheric signature unaffected by the processes that removed the beryllium bearing aerosols from the Arctic troposphere.

[12] We present here an overview of an extensive data set of 10Be/7Be ratios in the context of other chemical constituents and discuss the results in terms of STE. Three case studies illustrate how the 10Be/7Be ratio may lead to a better understanding of the complexities of stratosphere-tropo-
sphere exchange dynamics sketched out in the preceding discussion.

2. Methods

2.1. Sampling and Sample Selection

[13] We have measured the concentrations of $^7$Be and $^{210}$Pb in aerosols collected from the NASA DC-8 on a series of NASA campaigns as part of the Global Tropospheric Experiment (GTE) and of the Atmospheric Effects of Aviation Project Subsonic Assessment (AEAP SASS). The specific campaigns discussed here are GTE’s Pacific Exploratory Missions (PEM) West (A and B) and Tropics A, SASS Subsonic Aircraft: Cloud and Contrail Effects Special Study (SUCCESS), and the Ozone and Nitrogen Oxide Experiment (SONEX). Dibb et al. [1996, 1997, 1998, 1999, 2000] have discussed the $^7$Be and $^{210}$Pb results from these campaigns and have described the airborne aerosol sampling system and protocols that were used.

[14] An earlier investigation of $^{10}$Be/$^7$Be as an atmospheric tracer in the high Arctic relied on $^{10}$Be measurements in 8 samples collected during PEM West A and 6 collected during AGASP 3 [Dibb et al., 1994]. These results are incorporated into the present analysis, but are dwarfed in number by new measurements of $^{10}$Be during the PEM West B (32 samples), SUCCESS (58 samples), SONEX (143 samples), and PEM Tropics A (66 samples) campaigns. Targets for $^{10}$Be analysis were only prepared from samples that contained more than $5 \times 10^5$ atoms of $^7$Be. Although this selection criteria was necessary to ensure that there would be sufficient $^{10}$Be to quantify with good accuracy, it does bias our data set to include more upper tropospheric samples with significant stratospheric influence than would be representative of the study regions. The mean standard deviations for $^7$Be, $^{10}$Be, and the ratio, $^{10}$Be/$^7$Be are 12%, 9%, and 14%, respectively.

2.2. Target Preparation and Accelerator Mass Spectrometry

[15] $^{210}$Pb had previously been determined by alpha spectrometry for all of the samples discussed here, as described by Dibb et al. [1996]. The acid leach generated in this process, and the leached filter, had been retained. In the present study these filters were pulverized and leached again in 6 N HCl. This slurry was filtered and the liquid combined with the archived acid leach. Beryllium was then separated using cation exchange chromatography, Be(OH)$_2$ precipitated with NH$_4$OH and BeO prepared by igniting the purified precipitate. Targets were initially prepared by mixing pure Ag powder with the BeO (all samples from PEM West A, B and SUCCESS). Later targets were mixed with Nb instead of Ag to enhance BeO-currents in the Cs sputter source of the Lawrence Livermore National Laboratory (LLNL) FN accelerator mass spectrometer. This change in procedure was made for samples from SONEX and PEM Tropics A. Targets were sent to LLNL for analysis in the Center for Accelerator Mass Spectrometry AMS system, Davis et al. [1990]. $^{10}$Be concentrations were normalized to an ICN $^{10}$Be standard prepared by K. Nishiizumi.

3. Results

[16] Selected aerosol samples collected during five different NASA field campaigns (PEM West A and B, PEM Tropics A, SONEX, and SUCCESS) were analyzed. In addition to both $^7$Be and $^{10}$Be, ancillary data from these campaigns include O$_3$ (all missions); CO$_2$, CH$_4$, and CO (all but PEM West A); HNO$_3$ (PEM West B, PEM Tropics A, and SONEX); and N$_2$O (PEM West B, SONEX, and SUCCESS). In addition, on three missions (PEM Tropics A, SONEX, and SUCCESS) the height of the tropopause was determined from microwave temperature profile (MTP) data. Overall, the data set includes 300 samples for which the ratio of $^{10}$Be to $^7$Be has been determined.

[17] In general, both $^{10}$Be and $^7$Be increase with altitude (Figure 1). Although low mixing ratios for $^{10}$Be and $^7$Be (<2.0 × 10$^5$ atoms/m$^3$ and <0.5 × 10$^5$ atoms/m$^3$, respectively) are sometimes observed at even the highest altitudes, the maximum value observed tends to increase with height as does the number of samples that show high ratios. Below 4 km, the mixing ratios for both radioisotopes do not exceed these thresholds, reflecting the loss mechanisms that remove the beryllium bearing aerosols from the troposphere.

Figure 1. $^{10}$Be/$^7$Be, $^{10}$Be, and $^7$Be plotted versus altitude, grouped according to the value of the $^{10}$Be/$^7$Be ratio: <1.0 (group 1, squares), 1.0–2.0 (group 2, circles), 2.0–3.0 (group 3, triangles), and >3.0 (group 4, diamonds).
The $^{10}\text{Be}/^{7}\text{Be}$ ratio exhibits a different behavior (Figure 1). Relatively high ratios are seen down to 4 km and even below. Further, high ratios are observed throughout the observed altitude range, even when the mixing ratios are low (Figure 1). It is difficult to interpret the $^{10}\text{Be}/^{7}\text{Be}$ ratio as a function of altitude alone. However, if tropopause height is used to separate the samples collected in the stratosphere from those obtained in the troposphere, a clearer picture emerges of the distribution of this ratio in the atmosphere.

Tropopause height (determined by remotely sensed temperature measurements) is available for a subset (123 samples) of the overall data set (300 samples). The mean, minimum, and maximum values for a number of parameters are given for this subset in Table 1, along with the number of samples collected either above or below the tropopause. None of the stratospheric samples were collected more than 2.5 km above the tropopause. Hence, these samples reflect only the lowermost stratosphere. Also, only $^{7}\text{Be}$ samples above the $5 \times 10^5$ atom threshold were analyzed for $^{10}\text{Be}$. Since the presence of elevated $^{7}\text{Be}$ often indicates the presence of stratospheric air in the sample, there may be a slight bias in the tropospheric data (Table 1). As expected, both beryllium radioisotopes, O$_3$, and HNO$_3$, all have higher mean mixing ratios in the stratosphere than in the troposphere, while CH$_4$, CO, and N$_2$O tend to be higher in the troposphere. CO$_2$ is fairly uniformly mixed.

To evaluate the relation between the $^{10}\text{Be}$ to $^{7}\text{Be}$ ratio and stratosphere-troposphere exchange, we further subdivided the stratospheric and tropospheric samples into four subgroups according to the value of the ratio: group one ratios are less than 1.0, group two ranges from 1.0 to 2.0, group three ranges from 2.0 to 3.0, and group 4 ratios are equal to or exceed 3.0. The means, minima, and maxima for these subgroups are also given in Table 1. The results of this grouping are illustrated graphically in Figure 2 as a function of distance from the tropopause, with the data partitioned into stratospheric and tropospheric samples (Figure 2, upper panel). The middle panel shows the same data, but split into
the four subgroups. Since none of the stratospheric samples have a ratio <1.27, there is no group S1.

[21] This absence of low ratios in the stratosphere suggests that values near the 0.60 production ratio mostly or entirely result from tropospheric production. As expected, the mixing ratios of both 10Be and 7Be are correspondingly low for the tropospheric group 1 samples (T1; Table 1). Group T2 has the lowest mean and maximum mixing ratios for both beryllium isotopes. This is also true for O3, and is generally true for HNO3, except for T4 (only one sample in this group had a HNO3 measurement).

[22] The 10Be/7Be ratio tends to be higher in the stratosphere than the troposphere (Figure 2; Table 1), however some of the highest values of the 10Be/7Be ratio were found in samples collected in the troposphere. These high ratio tropospheric samples must be indicative of stratospheric air entering the troposphere, because tropospheric air cannot age sufficiently for such high ratios to develop. Only 16% of the stratospheric samples belong to the highest ratio group, suggesting that stratospheric air near the tropopause generally has a low characteristic ratio. Most of the high ratio samples in both the stratosphere and troposphere were collected during the SUCCESS field campaign, which was conducted over the central United States in April and May of 1996 (Figure 2, bottom panel). The other two field campaigns for which we have the tropopause height were carried out during northern hemisphere autumn: September–November in 1996 (PEM-Tropics A) and 1997 (SONEX). Transport scenarios reported by Detto et al. [2000] suggest that the weakening of the subtropical jets in spring and summer allows the flux of stratospheric overworld air downward into the lower stratosphere. From there, it can be exchanged isentropically with the troposphere. If this scenario is true, we would expect tropospheric samples with high ratios to be centered near the tropopause break.

The SUCCESS samples (Figure 3) do cluster around 40° N. Thus this data supports a scenario in which older overlying stratospheric air descends in spring and enters the troposphere from the lower stratosphere. This scenario agrees with prior results indicating a greater influence of stratospheric overworld air in the lower stratosphere in the spring [Ray et al., 1999].

[23] A closer examination of 10Be/7Be ratio as a function of latitude and longitude (Figure 3) reveals that most stratospheric air was found around 40° (median absolute latitude, the mean, 44° reflects five high latitude samples above 60°). The oldest air in both the stratosphere and troposphere, based on the 10Be/7Be ratio, was found at a median latitude of 40° (means of 43° and 40°, respectively). S4 air was observed at latitudes ranging from 37° to 57°, while T4 air was encountered at latitudes ranging from 36° to 48°. Clearly, there are processes leading to the influx of stratospheric air originating well above the tropopause into the troposphere at these midlatitudes.

[24] The stratospheric samples collected closest to the poles (between 60° and 71°) lie in group S2, typical of air that resides near the tropopause throughout the stratosphere. However, high ratio samples, typical of groups S3 or S4, have been observed near the high-latitude tropopause [Dibb et al., 1994; Raisbeck et al., 1981], perhaps originating from the loss of 7Be as higher altitude air descends from a region of greater 7Be and 10Be production to a region with lower production [Bergmann et al., 2001] corroborates this scenario. Their results predict a layer of enhanced ratio just above the tropopause peaking just above 3.0 and then decreasing with increasing altitude [Bergmann et al., 2001]. While the peak in ratios observed by Dibb et al. [1994] is somewhat higher (ranging from 4.0 to 6.9), slow descent is able to explain such high near-tropopause ratios in this region. Assuming a descent rate of 0.08 cm/s for the Arctic at an altitude of 19 km [Schoeberl et al., 1992], it would take 144 days for an air parcel to descend to 9 km. Dibb et al. [1994] report a ratio of 2.4 observed at 19 km in the Arctic and higher ratios, 4.0–6.9 near the tropopause. Given the 7Be half-life of 53 days, this is sufficient time for 7Be decay to increase the 10Be/7Be ratio to these values. All of the high value

Figure 2. 10Be/7Be plotted versus distance from the tropopause. Top panel shows the difference between stratospheric and tropospheric samples. Middle panel further subdivides these according to the value of the 10Be/7Be ratio, the groups defined as in Figure 1 caption. Bottom panel shows the ratios observed on three different field missions.
ratios at high latitudes were observed in spring and summer. Observations in the tropopause region during the fall ranged from 0.6–2.2. This suggests the descent is associated with the polar vortex in winter.

[25] The $^{10}$Be/$^{7}$Be ratio yields more information than either $^{7}$Be or $^{10}$Be alone. All of the $^{7}$Be T1 mixing ratios are below $0.5 \times 10^6$ atoms/m$^3$ (Figure 4, Table 1), which reflects the predominantly tropospheric origin of these samples. Similarly, the $^{10}$Be T1 mixing ratios (Figure 5) are all uniformly low, which corroborates the suggestion that T1 data is due to in situ production in the troposphere. Aerosols here are scavenged too quickly to allow for the buildup of $^{10}$Be with age. The mixing ratios for both isotopes in the T4 group, however, are not at the high end of the observed range. Mixing and removal processes have reduced the amount of beryllium in the troposphere in such a way that using the mixing ratios alone would not indicate the presence of stratospheric overworld air. For $^{7}$Be, all of the T4 samples are below $10^6$ atoms/m$^3$, as are two of the five S4 data points. The highest $^{7}$Be concentrations observed are among the T2, S2, and S3 groups ($>2 \times 10^6$ atoms/m$^3$), with only one S4 data point just barely above this threshold (Figure 4). Similarly, for $^{10}$Be, the highest mixing ratios observed in the troposphere belong to the T2 and T3 groups (Figure 5). In the stratosphere, the highest values are S3 and S4. Note, the $^{10}$Be mixing ratios should reflect mixing and removal processes alone, while the $^{7}$Be mixing ratios also reflect some decay as air moves away from high production regions.

[26] T1 samples have the lowest mean mixing ratios for $O_3$ as well as for beryllium isotopes (Figure 6). However, unlike beryllium, $O_3$ mixing ratios increase with height through the stratosphere. The highest mixing ratio occurs in an S4 sample. Similarly, the next 6 highest $O_3$ samples are all in the S3 group. Both of these observations point to downward mixing of air from an overlying region of the stratosphere. What is unlikely to be noticed from the $O_3$ data alone is the stratospheric signature in samples from the T3 and T4 groups, some of which have $O_3$ mixing ratios very similar to most T1 data points (Figure 6). Without the beryllium ratio data, some T2 samples could be interpreted to be more heavily influenced by stratospheric air than many T3 and T4 samples. The ratio of $^{10}$Be to $^{7}$Be gives

**Figure 3.** The locations where the samples were collected. Top panel shows the difference between stratospheric and tropospheric samples. Middle panel further subdivides these according to the value of the $^{10}$Be/$^{7}$Be ratio, the groups defined as in Figure 1 caption. Bottom panel divides the data according to the field mission during which the sample was obtained.

**Figure 4.** $^{7}$Be plotted versus distance from the tropopause. Top panel shows the difference between stratospheric and tropospheric samples. Bottom panel further subdivides these according to the value of the $^{10}$Be/$^{7}$Be ratio, the groups defined as in Figure 1 caption.
more information about the sources of the sampled air than the mixing ratios alone are able to reflect.

4. Discussion

[27] This data set provides information relevant to answering some of the questions about stratosphere-troposphere exchange (STE) raised in the introduction.

4.1. In Situ Production Versus Downwelling

[28] Folkins et al. [1999] suggested elevated O₃ between the chemopause at 14 km and the tropopause at 17 km was due to in situ production rather than mixing down from the stratosphere into the troposphere. There are 11 samples from the PEM-Tropics A mission (September and October 1996) collected between 12° S and 16° S (the Folkins et al. study was done at Samoa at 14° S). Unfortunately, none of the samples here was collected between 14 and 17 km, but rather were obtained between 3 and 11 km. Most of the samples (eight) were typical tropospheric samples with ¹⁰⁷Be/¹⁰⁶Be < 1.0 and O₃ < 100 ppbv. The remaining three samples had ratios of 1.2, 1.4, and 3.0, with associated O₃ of 94, 74, and 112 ppbv, respectively. It is interesting that these three samples were collected between 4.5 and 7 km, with the highest ratio sample obtained at 4.5 km. These samples suggest some introduction of stratospheric air into the troposphere occurs at this latitude. There is insufficient data to resolve the question of in situ production, since none of these samples were collected between 14 and 17 km. However, it is clear that the ratio could be used to resolve this question in the future.

4.2. Tropopause Folding - PEM-West B Flight 17

[29] The Pacific Exploratory Mission - West B (PEM-West B) was part of the series of missions conducted under NASA's Global Tropospheric Experiment (GTE) program. PEM-West A and B were designed to study the effects of Asian outflow on the free troposphere over the Pacific during two different seasons (fall and spring, respectively). Flight 17 of the PEM-West B campaign was flown through a tropopause fold. This fold has been described in detail by Dibb et al. [1997]. The flight track included long, constant altitude legs at five levels along a SW-NE line over the Sea of Japan. The samples containing ¹⁰⁷Be and ¹⁰⁶Be during this flight are shown along with O₃, CO, CH₄, CO₂, and HNO₃ data in Table 2. The first two samples were collected at 35,000 ft (10.67 km) as the plane flew SW in and above the fold. The plane turned and headed back NE, maintaining the same altitude while two more samples were collected. The remaining samples were collected at or below 20,000 ft (6.10 km), well below the tropopause fold. The highest concentrations of both Be isotopes were observed in the first four samples. The stratospheric nature of these samples is borne out by the high O₃ (ranging from 259–479 ppbv) and HNO₃ (1031–2415 pptv) and low CO (34–51 ppbv), CH₄...
The 10Be:7Be ratio of these samples ranges from 2.01 to 2.75 (all in the S3 category), which may indicate stratospheric air being pulled down toward the tropopause from higher altitudes as the jet weakens with the onset of spring.

The remaining samples are all typical tropospheric samples (Table 2) with low O3 (36–49 ppbv) and HNO3 (120–392 pptv), and high CO (144–211 ppbv), CH4 (1774–1839 ppbv), N2O (311 ppbv), and CO2 (361–365 ppmv). However, what these tracers do not reveal is the influence of stratospheric air on sample 5, with a ratio of 3.82 ± 0.78. This ratio puts this sample in the T4 group and suggests the presence of old stratospheric air that has descended into the lower stratosphere prior to mixing into the troposphere via the fold. While mixing and/or aerosol scavenging has removed the stratospheric signature from the observed concentrations, the ratio has preserved the presence of this component.

4.3. Mixing in the Stratosphere - SONEX Flight 8

[31] The SONEX (Subsonic Assessment, Ozone and Nitrogen Oxide Experiment) mission was conducted by NASA in October and November of 1997 to better understand the impact of aircraft exhaust on O3, and NOx in the free troposphere (Thompson and Singh, unpublished data, 1997, available at http://telsci.arc.nasa.gov/~sonex/pages/pg1.html). The flights were based out of Ireland, Azores, and Maine to collect samples in high air traffic regions of the North Atlantic (Thompson and Singh, unpublished data, 1997). In particular, flight 8 was a northern survey, flying from Shannon, Ireland northeast to parallel the Norwegian coast up to about 69°N. This flight path crossed the tropopause into the stratosphere (Figure 7), where a box
flight plan was used to allow sampling along four constant altitude legs in the same region, with two legs in the stratosphere, one near the tropopause, and the last in the troposphere.

Potential vorticity (PV) maps have been used to delineate the stratosphere (higher PV) from the troposphere (lower PV) [Danielsen, 1968; Danielsen and Mohnen, 1977; Danielsen and Hipskind, 1980; Danielsen et al., 1987; Browell et al., 1987]. These plots were produced routinely for the SONEX mission. The path of Flight 8 is superimposed onto a contour plot of the potential vorticity (Figure 8). The symmetry of the contours reflects the plane turning back on itself between samples 8 and 9, and again between samples 10 and 11 and finally between 12 and 13. The darkest orange contours are indicative of stratospheric air, while the blue end of the spectrum is tropospheric. The first three samples collected at 32,000 ft are south of the tropopause crossing point and are well within the troposphere. The chemistry of these samples reflect that; both Be isotopes, O3, and HNO3 have low concentrations, while CO and CH4 are high (Table 2). Increasing HNO3 and 10Be/7Be in the third sample reflects the approach to the tropopause.

The next 7 samples are stratospheric, the first 5 collected at 37,000 ft and the remainder at 33,000 ft. The PV plot (Figure 8) shows the flight path crossing a gradient, which is particularly evident in the change in the chemistry of the first three samples. O3 increases from 109 to 245 ppbv, HNO3 increases from 305 to 621 pptv, while CO drops from 62 to 31 ppbv, and CH4 drops from 1748 to 1700 ppbv (Table 2). The Be isotope concentrations also increase over this interval from $795 \times 10^{3}$ to $2716 \times 10^{3}$ atoms/m$^3$ ($^{7}$Be) and from $1304 \times 10^{3}$ to $4158 \times 10^{3}$ atoms/m$^3$ ($^{10}$Be). This reflects the transition zone nature of the near tropopause region both in the troposphere and the stratosphere. The ratio does not change much over this interval, nor does it over the entire set of stratospheric samples. The highest ratios, near 1.9, are observed nearest the peak in PV, samples 8 and 9 (Figure 8). Note, the sample numbers on Figure 8 are schematic, use the time in Table 2 to locate the sample along the flight leg using the x-axis of Figure 8. This supports the contention that the highest ratios are to be found further away from the tropopause. However, for this flight, all of the samples are S2 samples, the group that represents the youngest stratospheric samples observed. This is consistent with a suppression of downwelling of older air from the stratospheric overworld into the lower stratosphere at this time of year (late October) as discussed in the introduction.
Two of the samples during this flight that were classified as tropospheric based on the MTP data clearly have stratospheric chemical characteristics (Table 2) and would be classified as that based on PV (samples 6 and 10, Figure 8). Hence the notation in the table has been modified to S(T). It has been argued that PV is a better quantity to distinguish between stratospheric air and tropospheric air [e.g., Danielsen, 1968; Danielsen et al., 1987]. However, MTP data is available for more of the combined 10Be and 7Be data set than PV data and is therefore used here, even though it may sometimes yield misleading results.

4.4. Synoptic Systems - SONEX Flight 6

Flight 6, also from the SONEX mission, was carried out five days earlier than flight 8 described above. This flight also originated in Shannon, Ireland, but headed south to Spain, then angled out over the Atlantic. This path cut across a particularly steep gradient in tropopause heights (Figure 9). The gradient was due to strong convection associated with a trough over the eastern Atlantic evident at both 500 hPa and 250 hPa and an associated surface cyclone located off the northwestern corner of the Iberian peninsula. Surface convergence and uplift due to the cyclone on the southern portion of the flight allowed for tropospheric air to be convected above 40,000 ft (Figures 9 and 10). A cutoff low (Figure 11) to the north of this region appears to be responsible for the sharp drop in tropopause height to below 30,000 ft just to the west of the northern portion of the flight path. The lowest tropopause height encountered by the DC-8 was at 30,000 ft on the northern part of the flight track. On the outbound leg, the altitude of the plane was 29,000 ft, approaching the tropopause, but not crossing it (Figure 9; Table 2). The four samples collected on this leg are all tropospheric, however the presence of stratospheric air is evident to varying degrees. All of the concentration data indicate the greatest stratospheric influence in the first of these four samples, both Be isotopes, O3, and HNO3 are highest, while CO and CH4 are lowest for this sample. However, the beryllium ratio shows that stratospheric air has influenced all four samples, with the oldest air in the second sample, not the first. What is striking about this sample is its exceedingly high ratio of 6.46 ± 1.14. This is the highest ratio observed in the entire data set. Its presence in the troposphere suggests that old air in the stratosphere generally resides away from the tropopause where most of the stratospheric samples were collected.

Another striking feature is that this is the only T4 sample collected in the fall (20 October 1997). The others were all collected during the SUCCESS mission in spring, presumably due to downwelling from the stratospheric overworld as the jet weakened. This intrusion of stratospheric air does not appear to be a tropopause fold, because the intrusion is not drawn equatorward as it descends (Figure 12) which is characteristic of folds. Another scenario for bringing stratospheric air into the troposphere involves large storm systems. This synoptic system off the coast of Spain produced rain from the northwest coast of France to the southern end of Portugal. The DC-8 reported lightning associated with con-
vective cells (Thompson and Singh, unpublished data, 1997). It is likely that this cutoff low allowed the observed depression of the tropopause height, drawing stratospheric air down to low altitudes during this flight. It is not clear whether the very old stratospheric air observed descended solely because of the storm, or whether it had descended due to other processes and the storm drew it down from the lower stratosphere. Note, the concentrations of the Be isotopes are not atypical, so the high ratio is not an artifact of a particularly low amount of $^7$Be.

On the return leg, the aircraft altitude was sufficiently high to enter the stratosphere. Samples 14 and 17 appear to be very near the tropopause and reflect a blend of tropospheric and stratospheric air. Samples 15 and 16 appear to lie entirely in the lower stratosphere (Figure 10; Table 2). The last two samples are predominantly tropospheric. The ratios are consistent with this. And while one of the stratospheric samples has a ratio above 2.0 (S3), none of the other ratios reflect the presence of air as old as that observed on the outbound leg of the flight.

5. Summary

We have presented the largest $^{10}$Be (half-life of $1.5 \times 10^6$ years) data set to date. Combining samples from five NASA aircraft field missions (PEM: West A and B, Tropics A; SONEX; and SUCCESS) the data set is composed of 300 samples for which the $^{10}$Be/$^7$Be ratio may be determined. A subset of 123 samples for which there is also tropopause height (from temperature measurements) has been presented in detail to illustrate different properties of the ratio for stratospheric and tropospheric samples. Note that all of the stratospheric samples were obtained within 2.5 km of the tropopause; hence they represent only the lowermost stratosphere.

The absence of ratios <1.27 in the lowermost stratosphere indicates that ratios observed in the troposphere, nearer the production rate of 0.60, are characteristic of tropospheric air with minimal stratospheric influence. This is corroborated by the low mixing ratios of both beryllium isotopes for such low ratio tropospheric samples. Since, aerosols do not remain in the troposphere long enough for the ratio to increase substantially after production, the presence of high ratios in the troposphere reflects stratospheric air that has entered the troposphere. In the stratosphere, only 16% of the samples had ratios above 3.0. This suggests that high ratio air tends to reside away from the tropopause.

In the midlatitudes, the highest ratios are observed in the lowermost stratosphere and troposphere in the spring.
Figure 10. Potential vorticity (color contours) along SONEX flight 6 path (white). Time along the flight path is on the x-axis. Orange contour lines show geopotential height (m). Green contour lines show potential temperature (K). Black sample numbers indicate flight leg during which samples were collected. The blue contours show tropospheric air above 40,000 ft. While the flight path at 29,000 ft approaches the stratosphere, it does not enter it. Sample 2, collected on this flight leg, had the highest ratio observed in this data set. [This figure courtesy of Tom Kucsera, Science Systems and Applications, Inc., Mail Code 916, NASA Goddard Space Flight Center, Greenbelt, MD 20771.]

Figure 11. Geopotential height for 250 hPa shows a cutoff low to the northwest of the Iberian Peninsula. The low tropopause heights encountered on SONEX flight 6 are attributed to this low. [This figure courtesy of David Westberg, SAIC, NASA Langley Research Center, Mail Stop 483, Hampton, VA 23681, using NCEP Reanalysis data provided by the NOAA-CIRES Climate Diagnostics Center, Boulder, Colorado, USA, http://www.cdc.noaa.gov/.]
and summer months. This is attributed to the downwelling of high ratio air that resides at higher altitudes in the tropical stratosphere. This air descends as the subtropical jet weakens in spring and summer. At high latitudes, high ratios have also been observed in spring and summer in the lowermost stratosphere. However, in this case, the high ratios are attributed to the decay of $^7$Be to a lower secular equilibrium value as air slowly descends in the polar vortex in winter from a region of high beryllium production to a region with lower production.

The three studies described show that $^{10}$Be/$^7$Be ratios, when determined in the context of other chemical constituents, can provide insight into the various mechanisms of STE including, tropopause folding, changing seasonal structure of the stratosphere and the effect of synoptic systems. From this data set, it is evident that the beryllium ratio, $^{10}$Be/Be, is a sensitive tracer of stratospheric air. As such, it may be applied to various questions regarding stratospheric circulation and STE. Coupling $^{10}$Be/$^7$Be data with model transport should result in a greater understanding of the processes that drive atmospheric circulation.

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