Estimation of stratospheric input to the Arctic troposphere: ^{7}Be and ^{10}Be in aerosols at Alert, Canada

Jack E. Dibb and L. David Meeker

Glacier Research Group, Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham

Robert C. Finkel, John R. Southon, and Marc W. Caffee

Lawrence Livermore National Laboratory, Livermore, California

Leonard A. Barrie

Atmospheric Environment Service, Downsview, Ontario, Canada

Abstract. Concentrations of ^{7}Be and ^{210}Pb in 2 years of weekly high-volume aerosol samples collected at Alert, Northwest Territories, Canada, showed pronounced seasonal variations. We observed a broad winter peak in ^{210}Pb concentration and a spring peak in ^{7}Be. These peaks were similar in magnitude and duration to previously reported results for a number of stations in the Arctic Basin. Beryllium 10 concentrations (determined only during the first year of this study) were well correlated with those of ^{7}Be; the atom ratio ^{10}Be/^{7}Be was nearly constant at 2.2 throughout the year. This relatively high value of ^{10}Be/^{7}Be indicates that the stratosphere must constitute an important source of both Be isotopes in the Arctic troposphere throughout the year. A simple mixing model based on the small seasonal variations of ^{10}Be/^{7}Be indicates an approximately twofold increase of stratospheric influence in the free troposphere in late summer. The spring maxima in concentrations of both Be isotopes at the surface apparently reflect vertical mixing in rather than stratospheric injections into the troposphere. We have merged the results of the Be-based mixing model with weekly O$_3$ soundings to assess Arctic stratospheric impact on the surface O$_3$ budget at Alert. The resulting estimates indicate that stratospheric inputs can account for a maximum of 10-15% of the O$_3$ at the surface in spring and for less during the rest of the year. These estimates are most uncertain during the winter. The combination of Be isotopic measurements and O$_3$ vertical profiles could allow quantification of the contributions of O$_3$ from the Arctic stratosphere and lower latitude regions to the O$_3$ budget in the Arctic troposphere. Although at present the lack of a quantitative understanding of the temporal variation of O$_3$ lifetime in the Arctic troposphere precludes making definitive calculations, qualitative examples of the power of this approach are given.

Introduction

The chemistry and dynamics of the Arctic atmosphere have received a great deal of attention following the discovery of Arctic haze about 15 years ago [*Barrie*, 1986]. Arctic haze, which occurs every year in the winter-spring season, consists of visible haze layers associated with high levels of pollution-derived aerosols. In an effort to understand the origin of this annual pollution event, a number of surface-based monitoring stations were established and a series of airborne measurement campaigns have been conducted. The winter atmosphere has been characterized through missions flown in late winter 1983, 1986, 1989, and 1992 under the auspices of the NOAA Arctic Gas and Aerosol Sampling Program (AGASP). Summer conditions have been studied as part of NASA's Global Tropospheric Experiment, which mounted two combined airborne and ground-based campaigns, Arctic Boundary Layer Experiment

(ABLE) 3A in 1988 and ABLE 3B in 1990. Recently, the potential for large-scale, anthropogenically induced depletion of stratospheric ozone has been investigated in two NASA Airborne Arctic Stratosphere Experiment campaigns: January-February 1989 and through the winter of 1991-1992.

These studies have shown that Arctic haze is a basin-wide pollution event which is caused by low-level transport from midlatitudes to the Arctic. The reduced efficiency of pollution removal processes during the Arctic winter and spring and a maximum in meridional transport into the Arctic basin during the same season allow pollution concentrations in the lower troposphere to grow quite high (see the following special issues on Arctic haze: *Geophysical Research Letters*, 11 (5) 1984; *Atmospheric Environment*, 19 (12) 1985; and *Journal of Atmospheric Chemistry*, 9 1989). In the summertime, measurements at the surface indicate very "clean" air in the Arctic boundary layer. In contrast, airborne measurements between 2 and 6 km reveal enhanced concentrations of several trace species, with stratospherically derived air apparently an important source of O$_3$, NO$_y$, ^{7}Be, and perhaps SO$_4^{2-}$ and ^{210}Pb in the Arctic troposphere [*Browell et al.*, 1992, 1994; *Dibb et al.*, 1992a;

Gregory et al., 1992; *Sandholm et al.*, 1992; *Talbot et al.*, 1992; *Wofsy et al.*, 1992; *Anderson et al.*, 1994; *Bachmeier et al.*, 1994]. It is somewhat surprising that similar stratospheric influences on the free troposphere have not been shown to be significant during the winter-spring season as well, since measurements and modeling clearly show pronounced stratospheric subsidence in the winter Arctic vortex [e.g., *Schoeberl et al.*, 1992; *Toon et al.*, 1992; *Tuck et al.*, 1992]. Episodic intrusions of stratospheric air into the winter Arctic troposphere have been documented [e.g., *Shapiro et al.*, 1984; *Raatz et al.*, 1985] and it was suggested that the O_3 transported downward in such events could constitute an important term in the winter tropospheric O_3 budget [*Oltmans et al.*, 1989].

Here we use time series of near-surface concentrations of three natural radionuclides at Alert, Canada (82.5°N, 62.3°W), to examine the origins of tropospheric air in the Arctic Basin. Temporal variations of ^7Be and ^{210}Pb from late September, 1990 through September 1992 follow those previously reported from all other low-altitude Arctic sites where these two radionuclides have been measured [*Feely et al.*, 1988; *Larsen and Sanderson*, 1990]. We thus feel that Alert is representative of a large region of the western Arctic. The greatly different sources of these two radionuclides (decay of ^{222}Rn emitted from continental surfaces for ^{210}Pb and cosmic-ray-induced spallation in the upper troposphere and stratosphere for ^7Be) suggest that these radionuclides might serve as tracers of continental versus upper tropospheric or stratospheric air masses, respectively. The use of ^{10}Be in atmospheric studies is a recent development, as its measurement at low concentrations was made possible only by recent improvements in accelerator mass spectrometric techniques. *Raisbeck et al.* [1981] demonstrated the potential of ^{10}Be in such studies but analyzed only a small number of aerosol samples. *Brown et al.* [1989] and *Monaghan et al.* [1986] extended this work to investigate global properties of the atmosphere through analysis of precipitation samples. Wahlen and coworkers (M. Wahlen, personal communication, 1990) have measured aerosol filters collected on high-altitude flights.

A simple model based on the pioneering work of *Raisbeck et al.* [1981] is used to quantify the temporal pattern of stratospheric influence on the concentrations of the Be isotopes in surface air at Alert. Results from this Be-based model are then used to estimate the impact of stratospherically derived air on the surface ozone budget throughout the year.

Methods

Sampling

Weekly high-volume aerosol samples from the routine monitoring program at Alert are collected on 20 x 25 cm^2 Whatman 41 filters. These filters have been shown to be less than 100% efficient for the submicron-size aerosols carrying ^{210}Pb and both Be isotopes [*Lockhart et al.* 1963; *Turekian et al.*, 1989]. Concentrations reported later in this paper may thus be 10-20% low, but ratios will not be affected. The site, experimental protocols, and other routine measurements have been described previously [*Barrie and Hoff*, 1985]. The subsamples for radionuclide determinations (1/8 of each filter) contained the particulates from 1600-2000 m^3 STP of air. At intervals of 4 weeks, the filters (plus a monthly blank) were mailed to Toronto for subsampling and archiving, and radionuclide aliquots were forwarded to the University of New Hampshire for determination of ^7Be and ^{210}Pb concentrations.

^7Be and ^{210}Pb Analyses

Upon receipt at the University of New Hampshire, filter strips were pressed into 4-mL polyethylene vials for nondestructive gamma spectrometry. The counting system consists of a germanium well detector coupled to a 4-kilobyte computer-interfaced multichannel analyzer. Lead 210 and ^7Be activities were determined from the areas of the 46.5- and 477.6-keV photopeaks, respectively. Calibration of the counting system was accomplished by frequent recounting of a blank filter strip spiked with certified ^{210}Pb and ^7Be standard solution that had been similarly pressed into a vial. Beryllium 7 activities were corrected (to the midpoint of each sampling interval) for decay between sampling and counting. This time period was generally near 45 days for the oldest filter in each group but occasionally approached 60 days. Despite this interval between sampling and analysis, ^7Be activities were high enough in all samples that uncertainty due to counting statistics was ≤5% for 10-hour counting times.

^{10}Be Analyses

After gamma counting, the filters were sent to Lawrence Livermore National Laboratory (LLNL), where they were leached, in the presence of Be carrier, with 50% HNO_3. Several releaching experiments demonstrated that the leaches were >95% effective. The leached Be was purified using ion-exchange chromatography, and BeO was prepared by igniting the purified ion-exchange fraction. Beryllium 10 concentrations were determined using the LLNL Center for Accelerator Mass spectrometry (CAMS) accelerator mass spectrometer [*Davis et al.*, 1990]. In addition to the samples from Alert, six samples collected in the lower stratosphere during AGASP 3 [*Dibb et al.*, 1992a] and eight samples collected in the upper troposphere and lower stratosphere over the Pacific ocean during the Pacific Exploratory Mission (PEM) WEST A (J.E. Dibb, R.W. Talbot, K.I. Klemm, G.L. Gregory, H.B. Singh, J.D. Bradshaw, and S.T. Sandholm, Asian influences over the western North Pacific during the fall season: Inferences from lead 210, soluble ionic species, and ozone, submitted to *Journal of Geophysical Research*, 1994) (hereinafter referred to as Dibb et al., submitted manuscript, 1994) were analyzed for ^{10}Be as part of this investigation. The overall uncertainty in the ^{10}Be measurements for the Alert samples is approximately 5%; for the very small volumes of air sampled during the airborne campaigns, the uncertainty can reach 100%. Additional uncertainty in the determination of air volumes sampled will have an impact on the absolute concentrations of both Be isotopes but not their ratio.

Results

Beryllium 7 and ^{210}Pb concentrations in surface-level air at Alert experienced large seasonal variations that were quite similar in the 2 years we studied (Figure 1). Lead 210 concentrations during the June-September minimum were tenfold lower than during the broad maximum, which occurred from November through March. Peak ^{210}Pb concentrations occurred in January and February both years, and were more than 15 times higher than concentrations in summer. Beryllium 7 concentrations were also lowest during the June-September period, with a recovery beginning in October that closely paralleled that of ^{210}Pb. However, peak ^7Be concentrations occurred a bit later in the year than peak ^{210}Pb concentrations, that is, in March-April 1991, while in 1992 the annual ^7Be peak
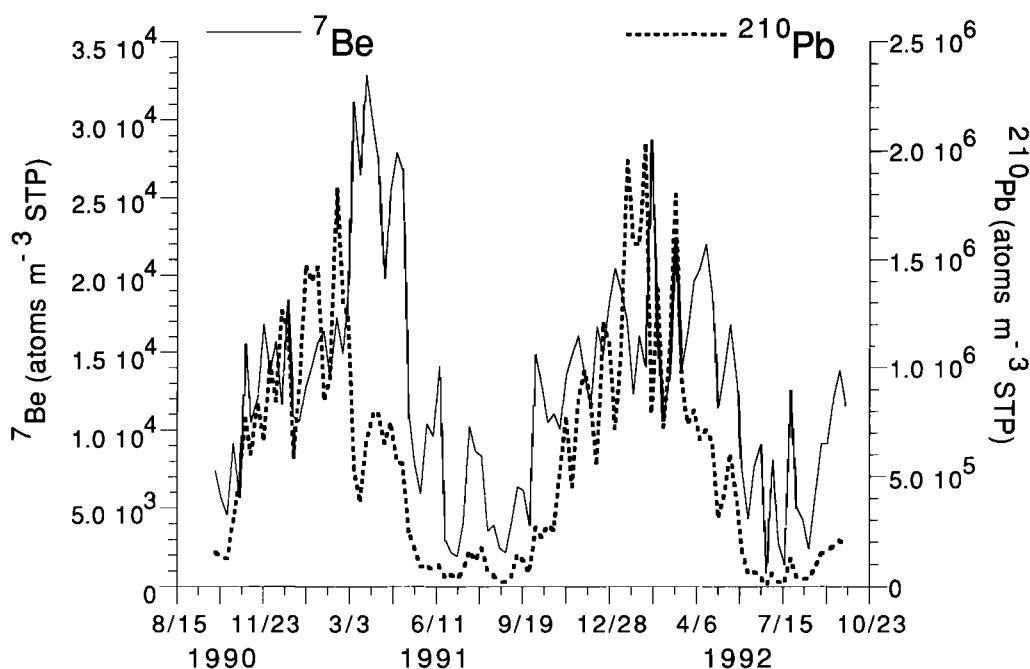
Figure 1. Concentrations of ^7Be and ^{210}Pb in surface-level air at Alert from September 25, 1990 to September 28, 1992. Sampling interval was weekly, with sampled volumes ranging from 1600 to 2000 m^3 STP. Average uncertainties are 3% for ^7Be and 6% for ^{210}Pb.

was lower and broader but persisted through April, when ^{210}Pb levels were steadily declining.

Beryllium 10 concentrations at Alert during the first year of sampling closely followed those of ^7Be (Figure 2). Our measured ^{10}Be concentrations, a few times 10^4 atoms m^{-3} STP, compare well with the few other tropospheric measurements available [*Raisbeck et al.*, 1981; S. Harder, personal communication, 1994]. The $^{10}Be/^7Be$ ratio measured at Alert averaged 2.2 ± 0.3 but varied seasonally, reaching maximum values near 3.0 in July-August, with fall-winter minima closer to 2.0. Concentrations of ^{10}Be but not ^7Be in the atmosphere can be locally enhanced by resuspension of surface soils. We estimated the potential contribution of resuspended ^{10}Be at Alert through the year from Al concentrations measured by neutron activation analysis of separate strips of each weekly filter, assuming that Al constitutes 7% (by mass) of dust [*Mason*, 1966; *Bowen*, 1979] and that dust contains 5 x 10^8 atoms ^{10}Be g^{-1} [*Pavich et al.*, 1984]. The ratio (estimated resuspended ^{10}Be/measured ^{10}Be) was generally << 0.05 (mean, 0.04 ± 0.08, median, 0.01). Eight samples with values of >0.05, including four ranging from 0.25 to 0.36, elevate the mean and cause the large standard deviation. All of the samples in which the estimated contribution of resuspended ^{10}Be exceeded 25% were collected in September and October, when the $^{10}Be/^7Be$ ratio was relatively low (Figure 2). This observation suggests that resuspended ^{10}Be is even less important than we have estimated, so no corrections for resuspended ^{10}Be were applied to the present data.

Beryllium concentrations in high-altitude Arctic air and lower latitude air are presented in Table 1. Concentrations of both Be isotopes at the base of the Arctic stratosphere during AGASP 3 were threefold lower than those at 10.7 km in 1978, but the average $^{10}Be/^7Be$ ratio (5.1 ± 1.0) was nearly the same (Table 1). Concentrations increase markedly with altitude in the Arctic stratosphere (at least up to 19 km), particularly for ^7Be. The $^{10}Be/^7Be$ ratio decreases significantly with height above the

tropopause at high northern latitudes (Table 1). *Raisbeck et al.* [1981] suggested that the apparent $^{10}Be/^7Be$ gradient in the lower stratosphere was due to descent (and concurrent mixing) from higher-production (higher altitude) regions at rates slow enough for decay to significantly decrease ^7Be concentrations. If this conceptual model is valid, detailed $^{10}Be/^7Be$ profiles could yield refined understanding of stratospheric stratification and mixing. Our new data for the Arctic cover only a very small altitude region just above the tropopause, but they do suggest that the high $^{10}Be/^7Be$ ratio in the earlier sample from 10.7 km was not anomalous.

Beryllium isotopic data are even more sparse for high altitude air from lower latitudes (Table 1). The PEM WEST A samples were all collected on transit flights, so only a single altitude was sampled in each geographic region. The first three samples collected in September were clearly from the lower stratosphere. The fourth September sample and all samples from October were from upper tropospheric air with stratospheric influence (E. V. Browell et al., Influence of stratospheric intrusions on chemical composition of the troposphere over the western Pacific during PEM WEST, submitted to *Journal of Geophysical Research*, 1994). The tropical sample from 16.8 km is felt to be stratospheric but may include tropospheric inputs from Hadley circulation [*Raisbeck et al.*, 1981]. This limited data set cannot be considered definitive, but it does suggest that the high $^{10}Be/^7Be$ ratios found near the Arctic tropopause may be restricted to high latitudes. Such a finding would be consistent with poleward transport and increasing age of stratospheric air masses.

Discussion

The seasonal variations of ^7Be and ^{210}Pb concentrations at Alert are strikingly similar to previously reported observations at a number of sites in the Arctic Basin, particularly Barrow, Alaska
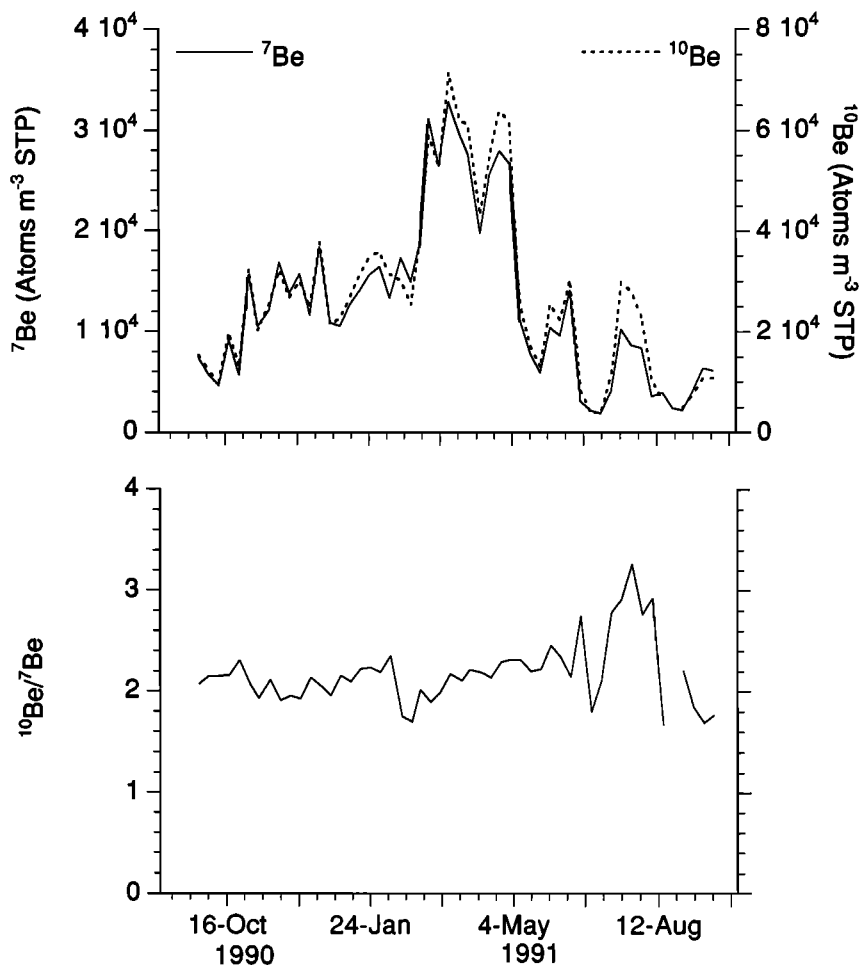
Figure 2. (top) Beryllium 10 concentrations in samples collected the first year of this study compared to ^{7}Be concentrations ; (bottom) ^{10}Be/^{7}Be ratios in the same samples . Average uncertainties in ^{10}Be measurement are 5% We point out that this data set differs slightly from that presented earlier [*Dibb et al.*, 1992c]. Analysis of new aliquots from four filters that we originally reported had anomalously high/low ^{10}Be/^{7}Be ratios has confirmed that accelerator mass spectrometry targets had been mislabeled (i.e., the ^{10}Be values were not registered to the correct samples). We offer apologies for any confusion.

(Table 2). *Rahn and McCaffrey* [1980] proposed a simple transport and aging model that qualitatively explained the elevated levels of ^{210}Pb and the anthropogenic component of Arctic haze reaching Barrow in late winter-spring. Very briefly, this model invokes relatively frequent meridional surges of midlatitude tropospheric air (predominantly originating over Europe) into the Arctic during the winter. Low scavenging rates in the Arctic Basin due to reduced precipitation in winter result in elevated concentrations of ^{210}Pb and SO$_4^{2-}$ from decay of ^{222}Rn and oxidation of SO$_2$, respectively, which are transported from industrialized continental source regions by the meridional surges. Modeling of ^{222}Rn and ^{210}Pb tropospheric distributions using a three-dimensional chemical tracer model based on climatologic meteorology from the NASA Goddard Institute for Space Sciences general circulation model also reproduces the very high ^{210}Pb concentrations seen at Barrow but not at Alert during winter [*Balkanski et al.*, 1993]. In the Balkanski et al. model, the plume of elevated ^{210}Pb concentrations originates over Siberia rather than over Europe, but both modeling efforts agree that winter-spring elevated ^{210}Pb concentrations in the western Arctic Basin are the result of long-range lower tropospheric transport from Eurasia.

The source of seasonal ^{7}Be variations is not, a priori, so obvious. At both Alert and Barrow, the ^{7}Be and ^{210}Pb variations are broadly similar (Table 2). As we have noted, at Alert the annual ^{7}Be peak in March-April clearly follows the ^{210}Pb peak in January-February. This distinction is not so apparent at Barrow; ^{7}Be does peak in March-April, but ^{210}Pb is roughly constant from December through March or April.

It is tempting to ascribe the spring peak in ^{7}Be at all of the Arctic sites (Table 2) to increased stratosphere-troposphere exchange and/or increased vertical (downward) mixing of upper tropospheric air. However, ^{7}Be concentrations begin increasing in October or November with ^{210}Pb, and it seems reasonably well established that the ^{210}Pb increase reflects tropospheric transport and lower removal rates. *Feely et al.* [1988] have examined ^{7}Be aerosol data from a large network of sites and noted a pronounced decrease in concentration along a transect from the continental United States into the Arctic. This led them to suggest that most ^{7}Be enters the Arctic troposphere by horizontal advection from lower latitudes. However, the recent research suggesting a strong stratospheric influence on the chemistry of the Arctic troposphere, noted above, suggests that this hypothesis is less than completely satisfactory. The concurrent ^{10}Be and ^{7}Be

Table 1. Beryllium 10 and ^{7}Be Concentrations in Stratospheric Air

	Latitude, °N	Altitude, km	Concentration, 10^4 atoms m^{-3} STP		
			^{10}Be	^{7}Be	^{10}Be/^{7}Be
Arctic Region					
July 1978	65	19.2	1290±210	533±112	2.4
July 1978	65	16.8	1190±155	502±55	2.4
July 1978	65	13.7	791±126	316±57	2.5
July 1978	65	10.7	663±113	117±13	5.7
March 16, 1989	68-72	8.1	55±5	14±1	4.0
March 18, 1989	71-74	8.2	86±1	18±1	4.9
March 21, 1989	68-75	8.9	132±1	25±1	5.2
March 23, 1989	71-72	8.3	282±5	68±2	4.1
March 29, 1989	68-82	8.1	265±1	38±1	6.9
March 30, 1989	68-75	9.1	176±7	33±2	5.4
Lower Latitudes					
September 16, 1991	56	10.7	131±26	96±10	1.4
September 17, 1991	47	10.7	210±5	120±4	1.8
September 18, 1991	41	10.7	100±9	65±4	1.5
September 18, 1991	38	10.8	5±5	9±1	0.6
October 21, 1991	36	10.7	14±5	6±1	2.2
October 21, 1991	32	9.8	33±5	35±3	0.9
October 21, 1991	30	9.8	18±9	28±3	0.6
October 21, 1991	30	9.8	21±7	23±2	0.9
July, 1978	9	16.8	181±40	97±18	1.9

All 1991 samples are from PEM WEST A (Dibb et al., submitted manuscript, 1994), all 1989 samples are from AGASP 3 [*Dibb et al.*, 1992a], and all 1978 samples are from *Raisbeck et al.* [1981].

measurements on aerosols collected at Alert provide new insight into this question.

General Model for Be Isotopes in the Arctic Troposphere

Following *Raisbeck et al.* [1981], we suggest that the tropospheric concentrations of the Be isotopes are described by

$$dC_{10}/dt = -\lambda_r C_{10} + (Q_{10} + P_{10}) \tag{1}$$

$$dC_7/dt = -(\lambda_r + \lambda_7)C_7 + (Q_7 + P_7) \tag{2}$$

where λ_r is removal by scavenging and deposition (assumed to be first order and the same for both isotopes), λ_7 is the decay constant of ^{7}Be, Q is Be injected from the stratosphere, and P is Be produced in the troposphere. For convenience we define

$$Q_{10} = SrQ_7 \tag{3}$$

$$P_{10} = PrP_7 \tag{4}$$

where Sr is the ^{10}Be/^{7}Be ratio in the stratospheric source, and Pr is the ^{10}Be/^{7}Be production ratio. With the initial condition, $C_{10} = C_7 = 0$ at $t = 0$, (1) and (2) yield the general solution for ^{10}Be/^{7}Be given by *Raisbeck et al.* [1981]:

$$\frac{C_{10}}{C_7} = R = \frac{(PrP_7 + SrQ_7)(\lambda_r + \lambda_7)(1 - e^{-\lambda_r t})}{(P_7 + Q_7)\lambda_r(1 - e^{-(\lambda_r + \lambda_7)t})} \tag{5}$$

Rearranging (5) gives the more useful form:

$$\frac{Q_7}{P_7} = \frac{R(\lambda_r/(\lambda_r + \lambda_7))(1 - e^{-(\lambda_r + \lambda_7)t}) - Pr(1 - e^{-\lambda_r t})}{Sr(1 - e^{-\lambda_r t}) - R(\lambda_r/(\lambda_r + \lambda_7))(1 - e^{-(\lambda_r + \lambda_7)t})} \tag{6}$$

We assume that the value of R measured at Alert is representative of the free troposphere (since removal processes do not fractionate the two Be isotopes). As a first approximation, the small variation of R about its mean of 2.2 (Figure 2) suggests that the tropospheric budgets of both Be isotopes were nearly in equilibrium throughout the year of our measurements. The steady state value of Q_7/P_7 can be evaluated by letting t go to infinity in (6), but we also require estimates of Sr, Pr, and λ_r.

Air from just above the tropopause appears to be the most likely source of stratospheric injections into the troposphere. The polar front tends to separate the Arctic troposphere from lower latitudes, suggesting that injections of Arctic stratospheric air into the Arctic troposphere will dominate over meridional advection of stratospheric air injected into the troposphere south of the polar front. Thus the mean value, 5, of the ^{10}Be/^{7}Be ratio in the six AGASP filters (Table 1), provides an estimate for Sr.

Atmospheric ^{10}Be and ^{7}Be production rates are difficult to estimate, because most of the measured cross sections are for proton-induced reactions, while production in the atmosphere is dominated by neutron-induced reactions. Measurements in the beam stop at LAMPF gave a ^{10}Be/^{7}Be production ratio induced by neutrons on oxygen of 0.6 for a particle spectrum similar to

Table 2. Monthly Average Concentrations of ^{7}Be and ^{210}Pb in the Arctic Basin

	Average Concentration, fCi m^{-3} STP[a]			
	Alert	Barrow[b,c]	Nord[b]	Thule[b]
Beryllium 7				
Jan.	61	63	82	125
Feb.	67	64	78	130
March	90	77	98	153
April	92	80	110	126
May	51	57	64	87
June	26	27	44	57
July	25	26	42	58
Aug.	19	18	35	51
Sept.	34	26	48	59
Oct.	38	39	66	84
Nov.	56	57	83	100
Dec.	60	49	139	110
Lead 210				
Jan.	36.1	22.3		
Feb.	32.8	20.6		
March	24.7	23.2		
April	18.7	19.6		
May	8.1	8.8		
June	1.5	2.5		
July	2.0	2.2		
Aug.	1.8	1.6		
Sept.	3.7	2.8		
Oct.	6.8	6.4		
Nov.	19.1	13.4		
Dec.	25.4	21.2		

[a] 1 fCi = 245.9 atoms of ^{7}Be or 37,569 atoms of ^{210}Pb.
[b] Data are from *Feely et al.* [1988].
[c] Additional, more recent data are from *Larsen and Sanderson* [1990].

that in the atmosphere [*Klein et al.*, 1988]. Data for proton reactions on nitrogen gave ^{10}Be/^{7}Be production ratios of 0.1-0.18, depending on the energy [*Raisbeck and Yiou*, 1974]. There are no published neutron cross sections for nitrogen, but recent measurements of cross sections for ^{14}N + n suggest that the ^{10}Be/^{7}Be production ratio may be as high as 1.2-1.3 (M. Imamura, personal communication, 1993). We believe that a ^{10}Be/^{7}Be production ratio as high as 1.2 is difficult to reconcile with measured values of ^{10}Be/^{7}Be in the environment that are less than 1.2, since this ratio can increase with time only through decay of ^{7}Be and/or resuspension of ^{10}Be into the atmosphere. We measured ^{10}Be/^{7}Be ratios of <1.0 in four of eight samples collected in the upper troposphere/lower stratosphere over the North Pacific in 1991, albeit with large uncertainties (Table 1). In addition, *Brown et al.* [1989] found ^{10}Be/^{7}Be ratios of <1.2 in about 20% of 68 precipitation samples collected in Illinois, including three samples for which the ratio was <0.7. We use a ^{10}Be/^{7}Be atmospheric production ratio of 0.6 $\pm$ 0.1 in the calculations in this paper.

Rather than assuming a value for λ_r, we calculate steady state values of Q_7/P_7 as a function of aerosol mean lifetime T_r ($T_r = 1/\lambda_r$). As the aerosol lifetime increases, decay of ^{7}Be becomes significant and causes R to increase. However, if aerosol lifetimes are less than about 200 days, stratospheric contributions are required to maintain R at 2.2 (Figures 3a and 3b). Previous estimates of ^{7}Be residence time in the troposphere converge on 30-40 days [*Shapiro and Forbes-Resha*, 1976, and references therein], suggesting that a 200-day lifetime is implausibly long. It thus appears that frequent injections of stratospheric air are required to maintain the ^{10}Be/^{7}Be near 2.2 in the troposphere. We note that the aerosol lifetime required to maintain the ^{10}Be/^{7}Be ratio at 2.2 when Q = 0 decreases if Pr increases; for Pr = 0.8, 1.0, and 1.2, the necessary lifetimes would be 135, 92, and 64 days, respectively. Even for Pr = 1.2, these aerosol lifetimes seem too long. However, it is apparent that if Pr is shown to be greater than 0.6, the stratospheric influence estimated in all calculations presented below will have to be reduced.

Application of the Model to the Alert Results

If the residence time of Be-bearing aerosol in the Arctic troposphere were between 30 and 40 days and if R were truly constant at 2.2, then Q_7/P_7 would fall in the range 0.24-0.29 (Figure 3). This is in very good agreement with the global average of 0.26 calculated from a three-dimensional global climate model [*Brost et al.*, 1991]. However, the measured values of R at Alert showed an apparent seasonal pattern, increasing from about 2.0 in fall and winter to about 3.0 in July and August (Figure 2). We have averaged the observations from Alert by month to estimate changes in stratospheric injections of Be through the year. For constant aerosol lifetime, the higher R values in midsummer relative to those in winter indicate a twofold increase in the stratospheric fraction of tropospheric ^{7}Be (Figure 4a).

The earlier discussion of ^{210}Pb concentrations and the Arctic haze phenomenon indicates that there is also pronounced seasonality of aerosol scavenging in the Arctic troposphere, with maximum lifetimes in winter and minima in summer. Inclusion of an idealized seasonal pattern of aerosol lifetime consistent with this scenario accentuates the winter to summer increase in the stratospheric influence on tropospheric ^{7}Be (Figure 4b).

These calculations all use the equilibrium form of (6), since we were uncertain how to prescribe a time history of complete Be washout which would allow including the time-dependent terms.

However, equilibrium values of Q_7/P_7 are approached rapidly from higher values, suggesting that our approach yields conservative estimates of stratospheric influence. Regardless of the details, the higher values of R during midsummer indicate increased injections of stratospheric Be into the troposphere during the period when surface concentrations of both isotopes were at their lowest (Figure 2). Similarly, the spring peak in Be concentrations at Alert occurs when the ^{10}Be/^{7}Be ratio suggests that stratospheric influence is just beginning to increase from the smallest fraction observed during the year (Figures 2 and 4). These findings reflect the various factors that determine ^{7}Be concentrations at ground level [e.g., *Feely et al.*, 1988; *Dibb et al.*, 1992b]. At Alert the spring concentration peak of both Be isotopes appears to reflect increased vertical mixing of free tropospheric air down to the surface [e.g., *Putnins*, 1970]. During the summer, enhanced aerosol scavenging in the pervasive stratus deck over the Arctic basin results in low concentrations of aerosol-associated species in the boundary layer despite higher concentrations aloft [e.g., *Talbot et al.*, 1992].

The concentration of stratospherically derived ^{7}Be reaching the surface at Alert can be estimated from

$$S_{7,S} = (Q_7/(Q_7 + P_7))C_{7,S}, \qquad (7)$$

where the subscript S refers to surface observations. Values of $Q_7/(Q_7 + P_7)$ for all three aerosol lifetime scenarios explored in Figure 4 were multiplied by monthly average ^{7}Be concentrations. A peak in stratospheric ^{7}Be reaching the surface is clearly evident in March-April (Figure 5), but this peak largely reflects vertical mixing in the troposphere rather than stratospheric injections into the troposphere which peak in July-August (Figure 4). It should be noted that (7) and the observed ^{7}Be concentrations at Alert would predict a spring maximum in stratospheric ^{7}Be at the surface even if R was held constant at the mean value of 2.2 (fixing Q_7/P_7 in the 0.24-0.29 range). In fact, the relative amplitude of the peak would increase, as summertime estimates would be even lower than those in Figure 5.

It should be noted that all of the conclusions regarding seasonality of stratospheric injections into the stratosphere and stratospheric Be reaching the surface remain valid even if Pr is >0.6. The magnitude of stratospheric influence will be reduced but not eliminated even if Pr is as high as 1.2 (which we feel is probably too high, as outlined above).

Ozone at Alert and the Impact of Stratospheric Ozone

The estimated concentrations of stratospheric ^{7}Be at Alert also yield preliminary estimates of the volume fraction of stratospheric air F_{sa} at the surface:

$$F_{sa} = S_{7,S}/SS, \qquad (8)$$

(Figure 5). We assume that the Arctic stratospheric source (SS) has a constant ^{7}Be concentration equal to the mean of the six AGASP samples (32.7 x 10^4 atoms m^{-3} STP) (Table 1). We have multiplied the Be-based estimates of stratospheric air fraction by the monthly mean O$_3$ concentration just above the tropopause as determined from weekly soundings at Alert and we have compare the results of this calculation to observed surface O$_3$ concentrations (Figure 6).

The Be-derived estimates of stratospheric air reaching the surface will be relevant to O$_3$ only if the tropospheric lifetime of O$_3$ injected from the stratosphere is comparable to that of the Be-
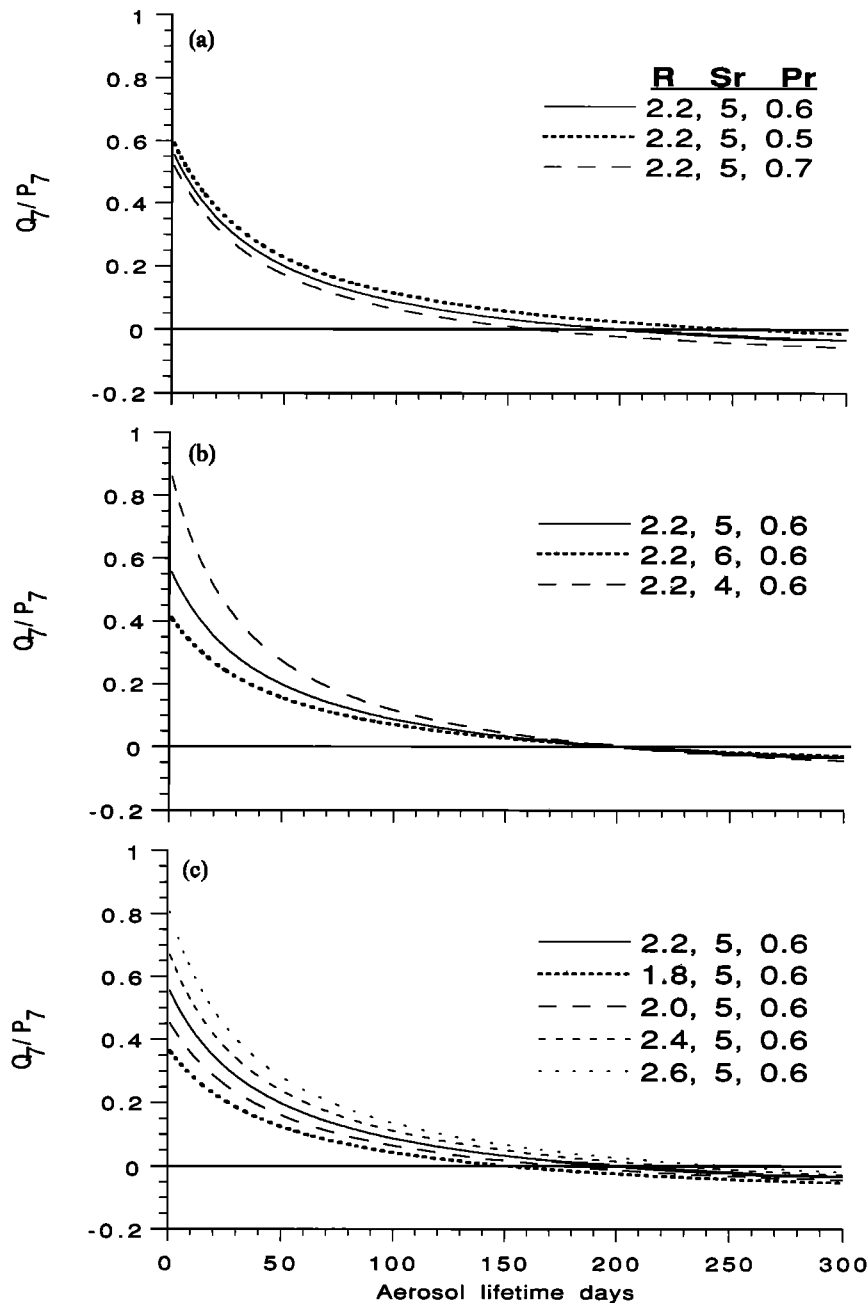
Figure 3. Values of the ratio of stratospherically injected ^{7}Be/tropospherically produced ^{7}Be (Q_7/P_7) in the troposphere as a function of aerosol residence time at steady state. (a Ratio of ^{10}Be/^{7}Be in the troposphere and stratospheric source held constant while the production ratio is varied; (b effect of varying the ratio in the stratospheric source; (c effect of different ratios in the troposphere.

bearing aerosols. *Jacob et al.* [1992] calculated a mean summertime lifetime of 46 days for O_3 in the Arctic troposphere, which is similar to the aerosol lifetimes assumed in the calculations behind Figures 4 and 5. Our estimates of stratospheric O_3 accounting for 10-15 % of the total at the surface during spring may thus be reasonable (Figure 6). During the dark Arctic winter, O_3 lifetimes are likely to be longer, suggesting that our estimated stratospheric contributions of <1 part per billion by volume (ppbv) are too low, but the lifetime of O_3 would have to be very long for the Arctic stratospheric contribution to exceed the 4 ppbv estimated in the spring. On the other hand, meridional transport of tropospheric air masses from

midlatitudes into the Arctic increases in the winter. Larger volumes of stratospheric air with low (<5) ^{10}Be/^{7}Be ratios (see midlatitude samples in Table 1) would be required to maintain R at 2.2 (Figure 3b), which might further increase the stratospheric fraction of O_3 at Alert during the winter. However, transfer of stratospheric air into the midlatitude troposphere is frequent in spring [e.g., *Staley*, 1982; *Dutkiewicz and Husain*, 1985] but may not be so common in winter when rapid transport into the Arctic occurs.

On an annual basis it appears that O_3 injected into the Arctic troposphere from the overlying stratosphere can account for only a small fraction of the total seen at Alert (and this fraction will be
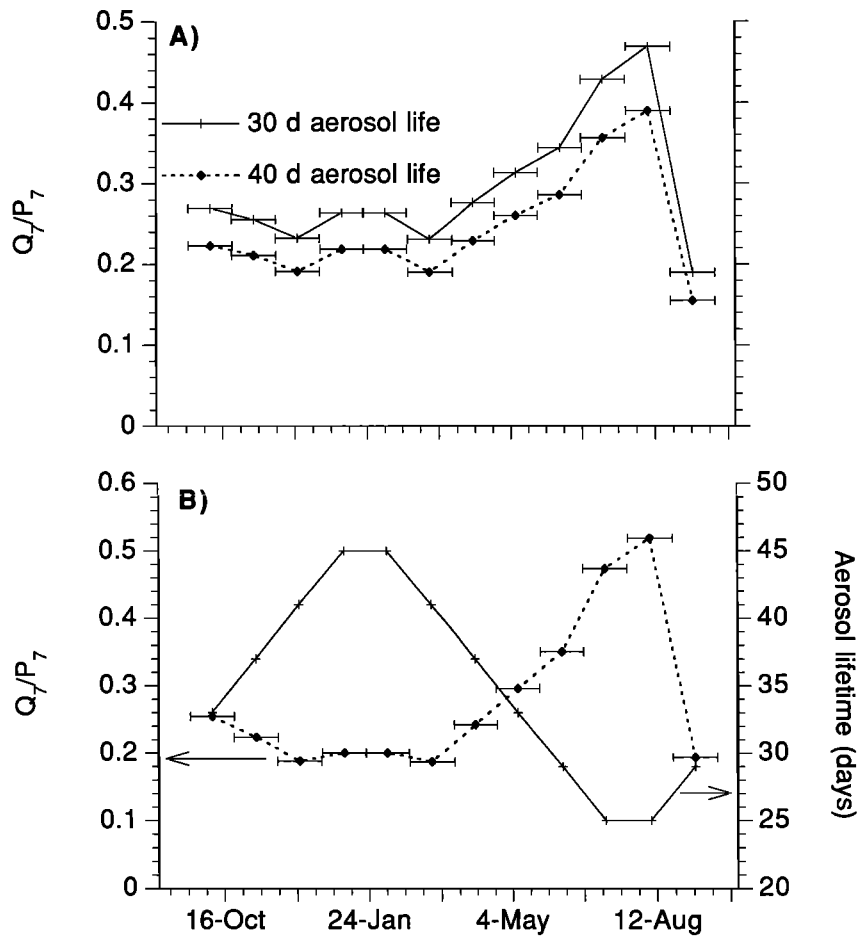
Figure 4. Values of Q_7/P_7 calculated from the monthly average value of $^{10}Be/^7Be$ at Alert. (a Aerosol lifetime assumed to be constant at 30 or 40 days; (b idealized seasonal pattern of aerosol lifetime assumed.

even smaller if new information about Pr indicates that the Be-based estimates should be revised downward). These estimates are in accord with a recent modeling study which found stratospheric O_3 to account for <5% of the zonal mean surface O_3 concentration above 70°N throughout the year (*Follows and Austin*, 1992).

Conclusion

Two years of sampling at Alert have demonstrated that ^7Be and ^{210}Pb concentrations in surface level air follow pronounced seasonal patterns that appear to have an impact on a wide region of the western Arctic and are quite predictable from year to year.
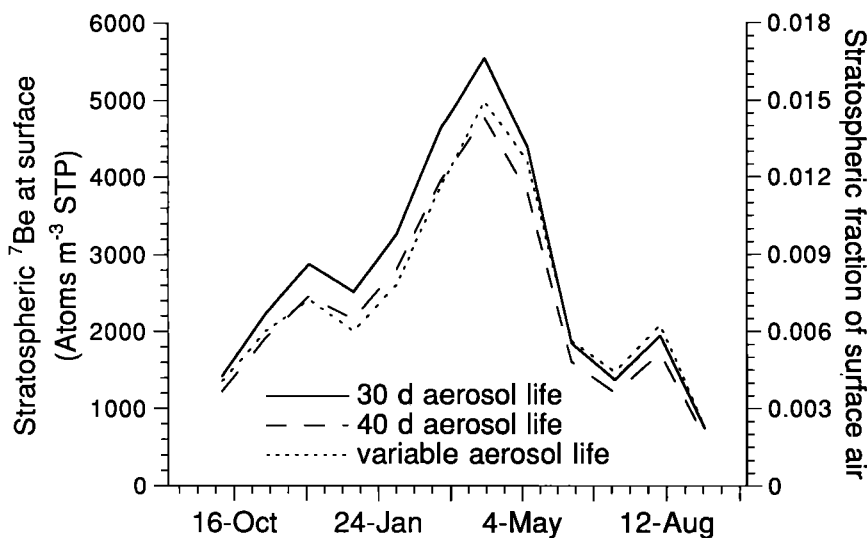
Figure 5. Estimates of the concentration of stratospheric ^7Be reaching the surface at Alert. The right axis converts ^7Be concentrations to the volume of stratospheric air required to deliver this much ^7Be, assuming a constant concentration of 32.7 x 10^4 atoms m^{-3} STP in the stratospheric source.
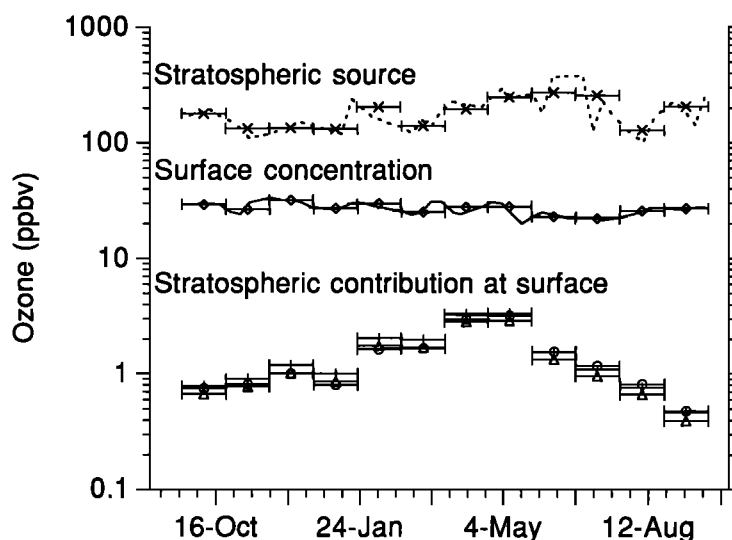
Figure 6. Estimated stratospheric ozone at the surface compared to observations at Alert. The estimates are the product of the stratospheric air fraction from Figure 5 and the monthly average O_3 concentrations at the base of the stratosphere over Alert (shown as the top curve here). Surface and stratospheric O_3 concentrations are shown for all soundings, with monthly averages represented by the horizontal bars. Estimates of stratospheric are shown as crosses O3 for a 30 day aerosol lifetime, as triangles for a 40 day lifetime, and as circles for a variable lifetime.

Similar findings have long been established for sulfate and other pollutants in the context of Arctic haze studies. This may indicate that the conclusions regarding stratospheric influence on surface air chemistry at Alert are applicable for the same large-scale region.

Concurrent ^{10}Be measurements during the first year of this study indicate that stratospheric air plays an important role in the budgets of both Be isotopes in the Arctic troposphere throughout the year. Small seasonal variations of the $^{10}Be/^7Be$ ratio indicate a twofold increase in stratospheric injections of the Be isotopes into the Arctic troposphere during late summer. The spring peak in the concentrations of both Be isotopes at the surface at Alert does represent the maximum stratospheric influence on surface level air, but it reflects more vigorous downward mixing of free tropospheric air rather than increased stratosphere-troposphere exchange during this season.

A simple mixing model based on the Be isotopes also allows first-order estimates of the portion of surface O_3 at Alert that could have come from the Arctic stratosphere. The maximum stratospheric influence on surface O_3 is predicted for the spring, but it accounts for only 10-15% of the surface concentrations. Lack of information about O_3 lifetime during the Arctic winter makes estimates of stratospheric contributions during this season highly uncertain, but the estimates are so low that it is unlikely that such contributions could equal those in spring.

At present, the sparse data on spatial and temporal distributions of $^{10}Be/^7Be$ ratios in the stratosphere represent the major limitation to quantifying stratosphere-troposphere exchange with this isotope pair. Improving this data base would also provide insights into vertical and horizontal mixing within the stratosphere. We are currently exploring the feasibility of quantifying ^{10}Be concentrations on some of the aerosol filters collected in the stratosphere during various high-altitude sampling programs in the 1960s.

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L. A. Barrie, Atmospheric Environment Service, Downsview, Ontario, Canada.

M. W. Caffee, R. C. Finkel, and J. R. Southon, Lawrence Livermore National Laboratory, Livermore, CA 94550.

J. E. Dibb and L. D. Meeker, Glacier Research Group, Institute for the Study of Earth, Oceans and Space, University of New Hampshire, Durham, NH 03824-3525. (e-mail: jack_dibb@grg.unh.edu)