Stratospheric Influence on the Northern North American Free Troposphere during TOPSE: ^{7}Be as a Stratospheric Tracer

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Abstract.

We use ^{7}Be, with HNO_3 and O_3, to identify air masses sampled during TOPSE that retained clear evidence of stratospheric influence. A total of 43 such air masses, spread

fairly evenly across the February to May duration, and 38°N - 86°N latitude range, were encountered. South of 55°N nearly all clear stratospheric influence was restricted to altitudes above 6 km, at higher latitudes the impact was felt through the depth of the troposphere. Approximately 12% of all TOPSE sampling time at altitudes above 2 km was spent in stratospherically impacted air, above 6 km this increased to more than half of the time. Because it is not certain how much of this stratospherically influenced air irreversibly injected mass (and chemical compounds) into the troposphere, we estimate the stratospheric fraction of O_3 in high latitude TOPSE samples based on a linear relationship to ^7Be and compare it to insitu O_3. This analysis indicates that the stratospheric source can account for a dominant fraction (> 85%) of insitu O_3 throughout TOPSE, but that the stratospheric contribution was nearly constant through the 4 month campaign. In February and March the ^7Be based estimates of stratospheric O_3 account for 10 - 15 % more O_3 than was measured, but by April and May there is up to about 10 % more O_3 than expected from the stratospheric source. This trend suggests that a seasonal transition from O_3 depletion to photochemical production in the high latitude North American troposphere is the major cause of the spring time increase in O_3.

Introduction.

The overall goal of the Tropospheric Ozone Production about the Spring Equinox (TOPSE) airborne experiment was to investigate the chemical and dynamic evolution of the troposphere over northern North American during the winter/spring transition. Particular emphasis was placed on understanding the evolution of the spring time ozone maximum in the middle troposphere. A central issue in this regard was to determine the

relative contributions of stratosphere-to-troposphere exchange and photochemical production of O_3 to the spring peak.

Previous investigations in Europe point toward a significant photochemical component in the spring O_3 peak [eg., *Penkett and Brice*, 1986; *Penkett et al.*, 1993; *Solberg et al.*, 1996; *Simmonds et al.*, 1997]. However, *Bazhanov and Rodhe* [1997] point out that at Areskutan, Sweden the contribution of stratospheric O_3 peaked during the spring tropospheric O_3 maximum and was as important as photochemical production to the tropospheric budget. In North America the composition of the high latitude troposphere during spring has not been well constrained to date, though the NASA GTE ABLE 3 (A + B) campaigns extensively characterized the Arctic and sub-Arctic during summer. Stratospheric injection was shown to be a significant source of O_3, NO_y and ^7Be [*Browell et al.*, 1992, 1994; *Dibb et al.*, 1992; *Gregory et al.*, 1992; *Sandholm et al.*, 1992; *Wofsy et al.*, 1992; *Anderson et al.*, 1994; *Bachmeier et al.*, 1994], though photochemistry was also very active. During the ABLE 3B campaign in summer 1990, *Mauzerall et al.* [1996] calculated that photochemical production of O_3 was the dominant source term despite clear stratospheric influence.

During winter photochemistry is reduced (or shut down at Arctic latitudes), while stratospheric injections still occur [*Shapiro et al.*, 1984; *Raatz et al.*, 1985; *Oltmans et al.*, 1989]. Presumably the stratospheric source is dominant in the dark, even if the magnitude of stratosphere-to-troposphere exchange is not at its annual peak. *Dibb et al.* [1994] reported year-round measurements of the $^{10}Be/^7Be$ ratio at Alert that indicate there must be significant stratosphere/troposphere exchange in the high Arctic throughout the

year, but that the peak injection of stratospheric Be isotopes occurred in summer, rather than during the spring peak in tropospheric O_3.

TOPSE was designed to probe the composition and chemical evolution of the North American troposphere between early February and late May, over the latitude range from Boulder (38°N) to 86°N. A series of flights using the NCAR C-130 was conducted at intervals through the winter/spring transition. *Atlas et al.* [this issue] describe the instrument payload and the detailed objectives of each flight conducted on the seven trips. In this paper we identify stratospheric influence on the composition of air masses sampled during TOPSE through enhancements in ^7Be, HNO_3 and O_3. In addition, we attempt to constrain the contribution of stratospheric O_3 to the O_3 budget in the high latitude portion of the TOPSE study region by using ^7Be as a tracer of stratospheric injections.

Methods.

Beryllium-7 activities were determined by non-destructive gamma spectroscopy in the laboratory in NH, as described previously [*Dibb et al.* 1996, 1997]. Aerosol particles were collected onto Whatman GF/A glass fiber filters. We employed the NCAR Small Community Aerosol Inlet (SCAI) which incorporates a curved leading edge nozzle, isoaxially mounted inside a cylindrical shroud, much like the inlet system that we have developed for use on the NASA DC-8 [*Dibb et al.*, 1996, in press]. Our experience has shown that such an inlet reduces particle losses compared to systems that do not include the shroud (which helps to straighten streamlines), and especially compared to knife edged nozzles. We fabricated a replacement nozzle for the SCAI with a larger diameter opening in order to achieve higher sampling flow rates while keeping the

velocity of air entering the nozzle within 10% of isokinetic rates. We expect that the submicron aerosol particles which carry essentially all ^{7}Be were transmitted to our filters with high efficiency by this system. (Limited comparisons between bulk $SO_4^{=}$ collected through the SCAI and fine (<2 micron) $SO_4^{=}$ collected with our mist chamber sampler during TOPSE [*Scheuer et al.*, this issue] suggest that the passing efficiency of the SCAI remains relatively high for particles with diameters up to at least 3 microns.) Filter samples were collected during all constant altitude flight legs throughout TOPSE, with nominal integration times of 15 – 20 minutes. Unanticipated changes in flight altitude resulted in some much shorter samples (minimum 3 minutes), while problems with our mist chamber sampling system occasionally distracted the operators and lead to longer samples (maximum 52 minutes), but the average filter exposure interval was 18.6 minutes (median 17.2 minutes). In total, 302 samples were collected for ^{7}Be.

Nitric acid was determined in near real-time onboard the LC-130, using the mist chamber sampling technique and ion chromatographic analysis [*Talbot et al.*, 1999]. This system is being continually refined to provide faster sample throughput and lower detection limits. The TOPSE configuration is described by *Scheuer et al.* [this issue]. The system was largely automated and collected a sample every 2.5 minutes, except when standard additions of HNO_3 and SO_2 were conducted to monitor passing efficiency of the inlet.

Ozone was determined with the NCAR ACD fast chemiluminescence instrument [*Ridley et al.*, this issue]. Data were reported at 1 Hz but we exclusively use averaged values in this paper, with one merge based on the mistchamber sampling times and a second using the filter sampling intervals. All raw data, and a selection of merged

products, including the one based on mist chamber sampling times used here, are archived at and available from NCAR ACD. The merge based on filter sampling times was generated at UNH and is available from the first author (this merge only includes measured ^{7}Be and ^{210}Pb activities plus average mixing ratios of HNO_3 and O_3).

Results.

TOPSE comprised 42 flights, including 4 instrument test flights in the vicinity of Boulder, followed by 7 trips northward at approximately biweekly intervals [*Atlas et al.*, this issue]. Each of the experimental trips included 4 – 7 flights, all included a transect from Boulder to the southern Hudson Bay near Churchill, Manitoba, with five trips extending to Thule, Greenland and points north. Most instruments were operational for the final test flight, hence the TOPSE data set includes results for flights 4 - 42.

We were able to quantify ^{7}Be activities on 244 out of the 302 samples collected during TOPSE (the remainder were below detection limits). As expected, highest activities were encountered at the highest altitudes and activities generally decreased toward the surface (Figure 1). Similar altitude trends in the mixing ratios of HNO_3 and O_3 (Figure 1) suggest that a common, stratospheric, source made a significant contribution to the tropospheric burdens of these gases during the TOPSE study period. However, the nature of the TOPSE experiment, with flights extending over almost 4 months and covering approximately 50^{o} of latitude, limits the utility of aggregated overviews like the altitude profiles in Figure 1. In order to identify stratospheric

influence during TOPSE we searched each flight for intervals where ^{7}Be, HNO_3 and O_3 increased concurrently.

The most obvious, and longest, example of an encounter with stratospherically influenced air occurred on TOPSE flight 16 (Figure 2). [This flight, conducted on 7 March, was entirely over Hudson Bay, originating from, and returning to, Churchill.] Between about 14:30 and 16:00 five consecutive filter samples with ^{7}Be activities > 1500 fCi m^{-3} were collected. Throughout this interval the mixing ratios of HNO_3 and O_3 were elevated compared to the rest of the flight. Considerable correlated structure is apparent in the gas mixing ratios through this interval (at the 2.5 minute resolution of the mist chamber samples), likely reflecting small scale spatial heterogeneity where air masses with stronger stratospheric influence inter-fingered with less impacted upper tropospheric air masses. For our purposes this high frequency variability is ignored and the entire interval is considered to be stratospherically influenced.

Flight 16 also illustrates the subjective nature of our categorization. Immediately prior to the identified interval of stratospheric influence O_3 increased from 60 to 110 ppb, and ^{7}Be activity was relatively high at 850 fCi m^{-3} (Figure 2). However, HNO_3 did not consistently mirror the increase in O_3, so this short interval was not classified as being stratospherically influenced. As a result, our compilation of encounters with stratospherically influenced air masses along TOPSE flight tracks should be viewed as conservative, with subtly influenced air masses likely more abundant than the clear intervals we identified.

Flight 21 presents another example of a clear encounter with stratospherically influenced air, as well as intervals with enhancements of one or two, but not all three, of

our stratospheric tracers. This flight was conducted out of Thule on 23 March and targeted boundary layer air above the Arctic ice pack with depleted O_3 [*Ridley et al.*, this issue]. Sharp increases in all three tracers were observed along the short level leg near 7 km during the return to Thule (Figure 3). About one hour later, ^{7}Be activity more than doubled in the final sample collected near 6 km but the mixing ratios of O_3 and HNO_3 decreased slightly. While it is likely that a large fraction of the ^{7}Be in this air originated in the stratosphere, the interval is not considered to be stratospherically influenced by our definition, again pointing out that our criteria highlight unambiguous stratospheric influence.

This analysis identified 43 intervals when the C-130 flew through stratospherically influenced air (Figure 4). It should be noted that, while we required elevated ^{7}Be activity to define stratospheric influence, the timing of encounters was based on parallel increases/decreases of O_3 and HNO_3 at the temporal resolution of the mist chamber sampling. On average, encounters with stratospherically influenced air masses lasted 28.5 minutes, but ranged from as short as 7 minutes up to the 80 minute encounter on Flight 16 (Figure 2). Stratospherically influenced air masses were identified on 23 of the 39 TOPSE science flights and occurred with similar frequency at all latitude bands of the study domain (Figure 4, top panel). There is also no obvious seasonal trend, with similar numbers of encounters on each of the 7 trips from February through May (Figure 4, mid panel). Most stratospherically influenced parcels were encountered near and above 6 km, in fact, we defined just over half of all flight hours above 6 km to be in stratospherically impacted air (Figure 4, bottom panel). South of 50^{o}N we only identified

a single encounter with stratospherically influenced air below 6 km, while at higher latitudes encounters occurred throughout the lower and middle troposphere.

As an additional check on our identification of stratospheric influence, we examined the fractional contribution of HNO_3 to NO_y during the intervals with elevated ^7Be, HNO_3 and O_3. During these intervals HNO_3 was the dominant N oxide, accounting for 50 – 80% of measured NO_y. This was not generally the case during TOPSE, with PAN and PPN usually the dominant N oxides [*Flocke et al.*, this issue], but is consistent with the overwhelming dominance of HNO_3 in the stratospheric N budget.

Discussion.

Enhancements in ^7Be, HNO_3 and O_3 clearly reveal that stratospheric influence was frequent during TOPSE, but do not provide much quantitative insight into the impact of stratospheric injections on the tropospheric budgets of reactive compounds. A big problem is that we can not determine, from available data, what fraction of the stratospheric tracers we measured along the flight track were ultimately injected into the troposphere. How much of the apparent stratospheric influence was irreversible stratosphere-to-troposphere exchange, as opposed to encounters with tropopause folds where much of the stratospheric air would end up returning above the tropopause? Several TOPSE modeling teams address this question (with different tools and approaches), and have not yet reached consensus (e.g., *Emmons et al.*, this issue; *Wang et al.*, this issue).

If we assume that all ^7Be measured during TOPSE originated in the stratosphere, it is possible to obtain semi-quantitative estimates of the stratospheric O_3 that would have

accompanied the ^{7}Be when it was injected into the stratosphere. We will focus on the higher latitudes where our analysis indicates stratospherically impacted air masses were encountered throughout the troposphere (Figure 4). (Note that this geographic restrictions also supports our neglect of ^{7}Be produced in the troposphere, since stratospheric production accounts for 85% of the total above 55^{o}N and 90% above 65^{o}N compared to 70% on a global basis [*Lal et al.*, 1958].) Rather than select an arbitrary latitude boundary, we include all flights except the transits between Boulder and Churchill in our high latitude subset (note that this eliminates 21 flights, since the northbound transits included a refueling stop in Winnipeg, resulting in 2 flights each trip enroute to Churchill). We collected 165 filter samples on the 18 high latitude flights, and were able to quantify ^{7}Be activities in 137 of them.

Ozone (averaged to filter collection times) was reasonably well correlated with ^{7}Be in the high latitude subset of TOPSE measurements (Figure 5). We tested several additional data filters to restrict the regression analysis to samples with strongest stratospheric influence. Selecting only samples with ^{7}Be activity > 500 fCi m^{-3} causes only small changes in the slope and intercept (Figure 5). Similar minor changes result when the regression is restricted to samples collected above 5 or 6 km (not shown), where air masses with obvious enhancements of all three stratospheric tracers were most frequently encountered (Figure 4). Regardless of filtering, linear fits to the TOPSE high latitude data set consistently yields estimates in the range 22 – 25 ppb O_3/pCi ^{7}Be. This value is only slightly higher than the 15 – 20 ppb/pCi range in (estimated stratospheric O_3)/^{7}Be reaching the surface at Alert in spring 1991 [*Dibb et al.*, 1994]. The TOPSE ratio is also similar to O_3/^{7}Be relationships we have observed in NASA airborne campaigns on

the DC-8 (maximum sampling altitude up to 12 km) over the north Pacific (in spring) and north Atlantic (in fall), but is more than 2-fold lower than observed over the central US in spring, and in several investigations that penetrated tropopause folds (Table 1).

The linear relationships between O_3 and ^7Be (Figure 5) were used with measured ^7Be to estimate stratospheric O_3 for each high latitude filter sample interval. Subtracting this estimate from observed O_3 yields a residual O_3 term that may provide an indication of O_3 produced (or destroyed) in the troposphere. Plotting residual O_3 against altitude suggests that most of the air masses sampled below 4 km had less O_3 than would be expected on the basis of observed ^7Be (Figure 6). At higher altitudes residual O_3 is roughly balanced between negative and positive values, mostly bounded by +/- 20 ppb. Plotting residual O_3 as a time series shows a small, but steady, increase over the length of the TOPSE campaign (Figure 7). On the first 3 trips nearly all sample periods had O_3 deficits and by the final 2 trips most intervals had higher O_3 than was predicted on the basis of ^7Be. The switch from an average deficit of about 15 ppb to an apparent excess of about 10 ppb O_3 accounts for much of the observed 34 ppb increase in average O_3 between the first and last trips (Table 2). It should also be noted that the average estimated stratospheric O_3 ranged only from 61 to 72 ppb (with the exception of trip, 3 which included Flight 16 discussed earlier) with little or no trend over the four months. This is equivalent to stating that the average ^7Be activity, inferred to reflect stratosphere-to-troposphere exchange, varied little over the length of the TOPSE campaign. *Browell et al.* [this issue] also point out the lack of seasonal variation in ^7Be and argue that the observed increase in O_3 thus can not be due to increased injection of stratospheric O_3. The analysis described above is consistent with this view, but it also suggests that

throughout TOPSE most of the O_3 in the high latitude troposphere could have been derived from the stratosphere. On average, our inferred stratospheric source provided more O_3 than was observed through the first 4 trips and even by the last trip the residual (non-stratospheric) O_3 only comprises about 15% of the total (Table 2).

The trend in residual O_3 (Figure 7) is consistent with a seasonal change from net O_3 destruction to net production in the Arctic over the TOPSE study period. However, the use of $O_3/^7Be$ to estimate stratospheric O_3 entails several major assumptions that may limit the utility of the analysis. As noted above, we assume that all of the ^7Be originated in the stratosphere. By pointing out that very little ^7Be is produced in the Arctic troposphere, we implicitly assume that large-scale transport of ^7Be is mainly vertical (downward from the Arctic stratosphere). *Feely et al.* [1988] argue that meridional transport (within the troposphere) of ^7Be from mid-latitudes may contribute to the spring time peak at Barrow. If this was important during TOPSE we would presumably over estimate stratospheric O_3 by the fraction of ^7Be that had been produced in the troposphere further south and was then advected into the study area. The spring peak in surface level ^7Be at Barrow (and several other Arctic stations [*Dibb et al.*, 1994]) occurs in March and April, thus our estimates of residual O_3 during trips 3 - 5 could be artificially low. On the other hand, it must be recalled that we saw no evidence for a spring-time peak in ^7Be above the surface during TOPSE, nor did ^7Be peak in spring at 2.5 km altitude on the Greenland ice sheet during the DGASP campaign [*Dibb and Jaffrezo*, 1993]. High values of the $^{10}Be/^7Be$ ratio throughout the year at Alert demand steady injection of stratospheric air into the Arctic troposphere [*Dibb et al.*, 1994]. It is hard to envision how pronounced seasonality in advection of tropospheric (lower activity) ^7Be from mid-

latitudes could combine with steady injection from the stratosphere to yield nearly constant average activities in the Arctic troposphere over the four months of TOPSE.

Our analysis also implicitly assumes that ^{7}Be injected from the stratosphere is effectively an inert tracer (i.e., we attribute changes in the $O_3/^{7}$Be ratio to processes changing the abundance of O_3). This is obviously an oversimplification, since radioactive decay and removal by wet deposition will reduce ^{7}Be activities without impacting O_3. Neglecting loss by radioactive decay can be justified on several grounds, most importantly because it does not vary over time, hence would not contribute to the seasonal trend in residual O_3. Seasonal variation in the efficiency of aerosol removal may be a more significant problem. Extremely long aerosol lifetimes are known to be an important component of the winter/spring Arctic Haze phenomenon, with more efficient scavenging in the pervasive stratus decks a major factor causing very clean air in the Arctic boundary layer during summer [*Dibb et al.*, 1994 and references therein]. An increasing trend in the rate of aerosol (and ^{7}Be) removal through the TOPSE period would cause us to estimate increasing values of residual O_3, since our estimates of stratospheric O_3 would be biased low by an increasing amount. Note that increasing the removal of ^{7}Be could be balanced by an upward trend in its injection from the stratosphere, explaining the nearly constant ^{7}Be activity that we observed across the seven TOPSE trips. We do not believe that seasonality in aerosol removal is seriously impacting our analysis, largely from consideration of the dynamics in the Arctic troposphere. Throughout the TOPSE period the cold surface maintained a very stable atmosphere, suppressing convection and limiting the opportunity for precipitation scavenging to preferentially remove ^{7}Be (relative to O_3). Under these conditions,

removal of ^{7}Be would be expected to be most efficient from the lower layers of the atmosphere, implying that we would under estimate stratospheric O_3 (overestimate residual O_3) most seriously close to the surface. Below 4 km our residual O_3 estimates are nearly all negative already (Figure 6), suggesting that any bias due to removal of ^{7}Be is not very large.

Another limitation of our analysis arises from the scatter in the O_3/^{7}Be regressions (Figure 5). We do not fully understand the sources of this variability, and recognize that it introduces uncertainty into the magnitude of residual O_3 that we estimate. However, it is unlikely that statistical noise would result in such a clear seasonal trend in the estimated residual O_3 (Figure 7). We therefore conclude that the increasing trend in residual O_3 must reflect loss of tropospheric O_3 throughout most of the TOPSE high latitude study region in February and March, changing to photochemical production during April and May.

Conclusions.

Stratospheric influence on the composition of the troposphere was frequently evident during TOPSE. Simultaneous enhancements in ^{7}Be, HNO_3 and O_3 can be used to identify air masses with a clear stratospheric component, but may not reveal more diffuse impacts and do not allow quantification of irreversible stratosphere-to-troposphere exchange. We presented estimates of the stratospheric contribution to observed O_3 at high latitudes based on linear regression between O_3 and ^{7}Be. This simple model suggests that most (> 85%) of the O_3 between the surface and the 8 km maximum sampling altitude originated in the stratosphere throughout the February – May TOPSE

campaign. However, our estimates of average mixing ratios of stratospheric O_3 were essentially constant at 65 ppb, while observed average mixing ratios increased from 48 to 82 ppb between early February and late May. Stratospheric injections of O_3 thus appear to be the dominant source of O_3 in the troposphere at high northern latitudes over this time period, but do not cause the spring time peak. Our analysis indicates that this region changes from one of net O_3 destruction in February and March to net production in April and May, with this change in photochemical tendency driving the observed seasonal increase in O_3 mixing ratios.

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Table 1. Comparison of ^{7}Be and O_3 relationship during TOPSE to other recent airborne campaigns.

Region	ppb O_3/pCi ^{7}Be	Campaign	Reference
North American high latitudes	25	TOPSE	*this work*
N. Am. high lat., ^{7}Be > 0.5 pCi m^{-3}	23	TOPSE	*this work*
Tropopause folds, western US (lower stratosphere)	67	STEP	*Kritz et al.*, 1991
Tropopause fold, Sea of Japan (upper troposphere)	59	PEM West B	*Dibb et al.*, 1997[1]
North Pacific (upper troposphere)	31	PEM West B	*Dibb et al.*, 1997[1]
North Atlantic (upper troposphere/ lower stratosphere)	25	SONEX	*Dibb*, unpublished[2]
Central US (upper troposphere/ lower stratosphere)	53	SUCCESS	*Dibb*, unpublished[2]

1 These data, and all ^{7}Be measurements we have made on GTE campaigns, are archived at the NASA Langley Distributed Active Archive Center.

2 SONEX and SUCCESS data archives are maintained at NASA Ames.

Table 2. Summary of the relationship between observed O_3 and stratospheric O_3 estimated from measured ^7Be at high northern latitudes during TOPSE.

Trip	1	2	3	4	5	6	7
Average O_3 (ppb)	47.6	57.8	85.7	59.0	69.0	78.5	81.9
Residual O_3 (all Be)[1]	-13.1	-9.3	-9.9	-5.7	3.9	8.0	11.5
Residual O_3 (Hi Be)[1]	-15.5	-11.2	-9.6	-7.8	1.8	6.3	9.8

1 Residual O_3 is simply observed O_3 minus the estimated stratospheric O_3, using the 2 different linear regressions of O_3 on ^7Be shown in Figure 5. This table presents the average difference, by trip, for all points with both ^7Be and O_3 observations.

Figure Captions.

Figure 1. Altitude distributions of ^{7}Be, HNO_3 and O_3 during TOPSE. Nitric acid and O_3 mixing ratios have been averaged over the aerosol collection interval.

Figure 2. Time series of ^{7}Be, HNO_3 and O_3 on TOPSE flight 16 presented to illustrate how increases in all three species during the shaded interval were used to identify air masses with stratospheric influence.

Figure 3. As in Figure 2, but for TOPSE flight 21.

Figure 4. Graphical summary of the distribution of stratospherically influenced air masses identified from enhancements of ^{7}Be, HNO_3 and O_3. Top panel simply plots occurrences in altitude – latitude space. Middle panel plots the length of each encounter against flight number. Flight 4 was the final test flight, flights 5 – 8 constitute trip 1 (4 – 9 February), flights 9-13 trip 2 (21 – 27 February), flights 14 – 17 trip 3 (5 – 8 March), flights 18 – 23 trip 4 (19 – 26 March), flights 24 – 30 trip 5 (2 – 11 April), flights 31 – 36 trip 6 (23 – 30 April), and flights 37 – 42 trip 7 (15 – 23 May). The bottom panel shows the ratio (time in stratospherically impacted air)/(total sampling time) as a function of altitude, for 3 broad latitude bands and also for all samples.

Figure 5. Scatter plot of O_3 (averaged to ^{7}Be integration time) against ^{7}Be at high latitudes during TOPSE (see text for discussion of the high latitude subset of data). The solid line is the least-squares fit through all data pairs, while the dotted line is the fit to all pairs in which the ^{7}Be activity was > 500 fCi m^{-3} (the solid points).

Figure 6. Estimated residual O_3 (= measured O_3 – stratospheric O_3) as a function of latitude. The upper panel uses the fit to all data points in Figure 5 to estimate

stratospheric O_3 from measured ^{7}Be, the lower panel uses the fit based on high ^{7}Be activity.

Figure 7. As in Figure 6 except that estimated residual O_3 is plotted against the time of sample collection. The clusters of points reflect the seven different trips described in Figure 4.