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## Springtime enhancement of upper tropospheric aerosol at 45S

### Abstract

Monthly sonde data for Lauder in Central Otago, New Zealand show profiles of aerosol backscatter from the surface to over 30 km altitude. The tropospheric data vary by season, with greater aerosol backscatter throughout the free troposphere in springtime. Aerosol mixing ratios in layers in the upper troposphere at these times are often much higher than anywhere else above the boundary layer, suggesting that they arise from horizontal transport. Ozone measurements from the sonde show correspondence in vertical structure to the backscatter data and also seasonal enhancement. The latter correlates with aerosol, but competing causes of ozone enhancement make the correspondence indistinct. High concentrations of carbon monoxide are observed by Fourier transform spectroscopy in spring, and altitude profiles derived from line shape suggest that the peak in CO occurs in the same altitude range as the aerosol enhancement.

### Keywords

upper, enhancement, 45s, aerosol, tropospheric, springtime, GeoQuest

### Disciplines

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## Springtime enhancement of upper tropospheric aerosol at 45° S

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**Abstract.** Monthly sonde data for Lauder in Central Otago, New Zealand show profiles of aerosol backscatter from the surface to over 30 km altitude. The tropospheric data vary by season, with greater aerosol backscatter throughout the free troposphere in springtime. Aerosol mixing ratios in layers in the upper troposphere at these times are often much higher than anywhere else above the boundary layer, suggesting that they arise from horizontal transport. Ozone measurements from the sonde show correspondence in vertical structure to the backscatter data and also seasonal enhancement. The latter correlates with aerosol, but competing causes of ozone enhancement make the correspondence indistinct. High concentrations of carbon monoxide are observed by Fourier transform spectroscopy in spring, and altitude profiles derived from line shape suggest that the peak in CO occurs in the same altitude range as the aerosol enhancement.

### 1. Introduction

Springtime enhancement of aerosol in the South Pacific free troposphere is well documented [Kristament *et al.*, 1993; Kent *et al.*, 1995] even from the limited data for this region of the globe, but the origin and composition remain uncertain. Possible causes include variation in the downward flux of stratospheric aerosol, increased production or conversion of precursor gases, seasonally enhanced generation or lofting of surface-derived aerosols (sea salt or aeolian dust), or biomass burning. In the southern hemisphere industrial sources for aerosol are of minor importance.

Kent *et al.* [1995] separate SAGE II data for the upper troposphere in the aftermath of the El Chichón eruption into two components. The 'volcanic' component follows the approximately exponential decay in stratospheric aerosol concentrations, leaving a residual 'baseline' attributable to other sources. By including earlier SAGE I and later SAGE II data Kent *et al.* [1998a] were able to represent the two recent episodes of massive global stratospheric aerosol enhancement and three contrasting quiescent periods. In this expanded dataset the seasonal mean aerosol concentrations in the upper troposphere and the stratosphere are correlated, and the correlation is a measure of the volcanic component in the former. Both seem to have a substantial seasonal variation, with southern hemispheric maxima in spring at different latitudes.

Kent *et al.* [1998a] note the lack of any major enhancement in upper tropospheric aerosol even where surface dust is very visible in AVHRR images [e.g., Husar *et al.*, 1997], so it is unlikely to be significant in the southern hemisphere where AVHRR sees little effect from desert regions.

Though dominant in the marine boundary layer, sea salt is not generally a major component of free tropospheric aerosol numerically, nor by mass despite the larger diameter of the mode. Size determinations for the springtime enhancements are similar to those for other seasons [Kristament *et al.*, 1993; Kent *et al.*, 1995], suggesting that sea salt is not a probable explanation.

Composition is a key criterion for aerosol origin, but determinations are rare for the free troposphere. Though sulphate, partially ammoniated, predominates [Clarke, 1993], many other species are common and most chemical elements are present in at least trace amounts [Murphy *et al.*, 1998]. Kristament *et al.* [1993] note that the K:Ca ratio in fine particles on filter samples is enhanced, matching the indicator of biomass-burning origin given by authors cited therein. More measurements of aerosol composition by latitude, altitude, and season are certainly needed.

The concentration of ozone and other trace species relative to aerosol is an important diagnostic of air mass history. Gravitational settling of stratospheric aerosol into the upper troposphere would not involve any enhancement of ozone, but air mass exchange across the tropopause would. Increased stratospheric-tropospheric exchange is expected in springtime from tropopause folding events and breaking gravity waves. A biomass-burning source for tropospheric aerosol might also be expected to correlate with enhanced ozone. Carbon monoxide and various hydrocarbons are more specific as tracers of air mass origin.

By relating lidar data from the space shuttle-borne LITE campaign in September 1994 to SAGE II data, Kent *et al.* [1998b] identify layers of enhanced aerosol in plumes downwind of major biomass-burning regions in the tropics and the sub-tropical southern hemisphere. These authors also observe that the seasonal cycle in upper tropospheric aerosol is similar to published distributions for ozone and carbon monoxide.

In this paper and one which follows we demonstrate that the seasonal cycle of aerosol enhancement in the free troposphere over Lauder closely matches a similar time series of carbon monoxide, but the correlation with ozone is less distinct.

### 2. Data

Backscatter sondes [Rosen and Kjome, 1991] were launched quasi-monthly from Lauder (45° S, 169.6° E) in Central Otago, New Zealand, from 1992 to 1998. The balloon-borne instrument uses a collimated xenon flash lamp and dual wavelength sensors to detect local molecular and aerosol backscatter in two broad wavelength bands around 940 and 490 nm. Because of the instrument geometry the measurement is not of true

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backscatter ( $180^\circ$ ) but is instead characterised by a distribution centered on  $173^\circ$  from forward. On the same payload an electrochemical cell measures ozone concentration, and a meteorological radiosonde measures temperature, pressure, and tropospheric humidity and transmits the data stream.

The backscattersonde calibration is by comparison with four standard backscattersondes maintained at Laramie which are in turn compared with an absolutely calibrated backscatter nephelometer. Repeatability of the relative calibration is about  $\pm 1\%$ , and the absolute calibration is better than  $\pm 3\%$ . There is also uncertainty of  $\pm 1\%$  in Rayleigh backscatter for air density calculated from the pressure and temperature measurements.

Like lidar systems and SAGE, the backscattersonde has mostly been used to study the stratospheric aerosol system, but unlike both of them it performs well throughout the troposphere [Rosen *et al.*, 1997]. Sondes are launched after sunset in relatively clear (less than 50% cloud cover) and calm (wind gusts less than  $\text{ms}^{-1}$ ) conditions, creating a bias in tropospheric sampling conditions which is common to other balloon-borne systems and to lidar measurements. The cloud cover limitations for good satellite-based measurement of tropospheric aerosol are generally more stringent, so that the same 'fair weather' bias will also affect SAGE data.

In total, 76 backscattersondes have been launched from Lauder. Despite the inevitable risk of mishap with balloon launches and telemetry, all flights have returned usable data. As a stratospheric data series, the backscattersonde measurements cover a very wide range of concentrations. In longer time series, such as ruby lidar data from Mauna Loa, the period since 1992 includes the highest values of peak and integrated stratospheric aerosol backscatter, and the recent values are amongst the lowest values. The former observation concurs with global volcanic records that show the Pinatubo eruption to be the largest in the past century, and the latter is an important constraint on our understanding of stratospheric aerosol

processes. For the present study the wide range of stratospheric aerosol amount is valuable for quantifying exchange into the troposphere.

### 3. Analysis

Clouds are the major contaminant in the data series, and for the data presented here clouds were identified by several means. Detector saturation is always attributable to water droplets or ice crystals, whether in fog, cloud, or condensing smoke, and all such instances were removed. High values of 'backscatter color ratio' (aerosol backscatter over Rayleigh backscatter at 940 nm relative to 490 nm [Rosen *et al.*, 1997]) are indicative of large particles, which are 'white' scatterers at visible wavelengths. The color ratio is 1 for molecular scatterers and around 14 for the largest particles; in this analysis those instances of high red backscatter for which the color ratio was greater than 10 were treated as cloud. It is possible that just above or below cloud there could be enhanced aerosol which will be rejected as cloud by the above criteria, but any such bias in the data should be slight.

Figure 1 shows contour plots of backscatter ratio obtained by averaging backscattersonde data in 500 m altitude bins. The most obvious feature is the now familiar decay in stratospheric aerosol in the aftermath of the Mount Pinatubo eruption. In the troposphere, the seasonal cycle is readily apparent in the 940 nm data, and the structure appears similar to the plots of Kent *et al.* [1998a]. High aerosol concentrations above the boundary layer occur around September (austral spring) in at least one profile every year, though the 1998 spring showed only moderate enhancement. With high mixing ratios in the stratosphere at the start of the series and in the boundary layer throughout, the possibility of local downward or upward mixing must be considered.

The tropopause altitude for each flight is shown in the white trace. It is typically 9-12 km for most of the year, with excur-

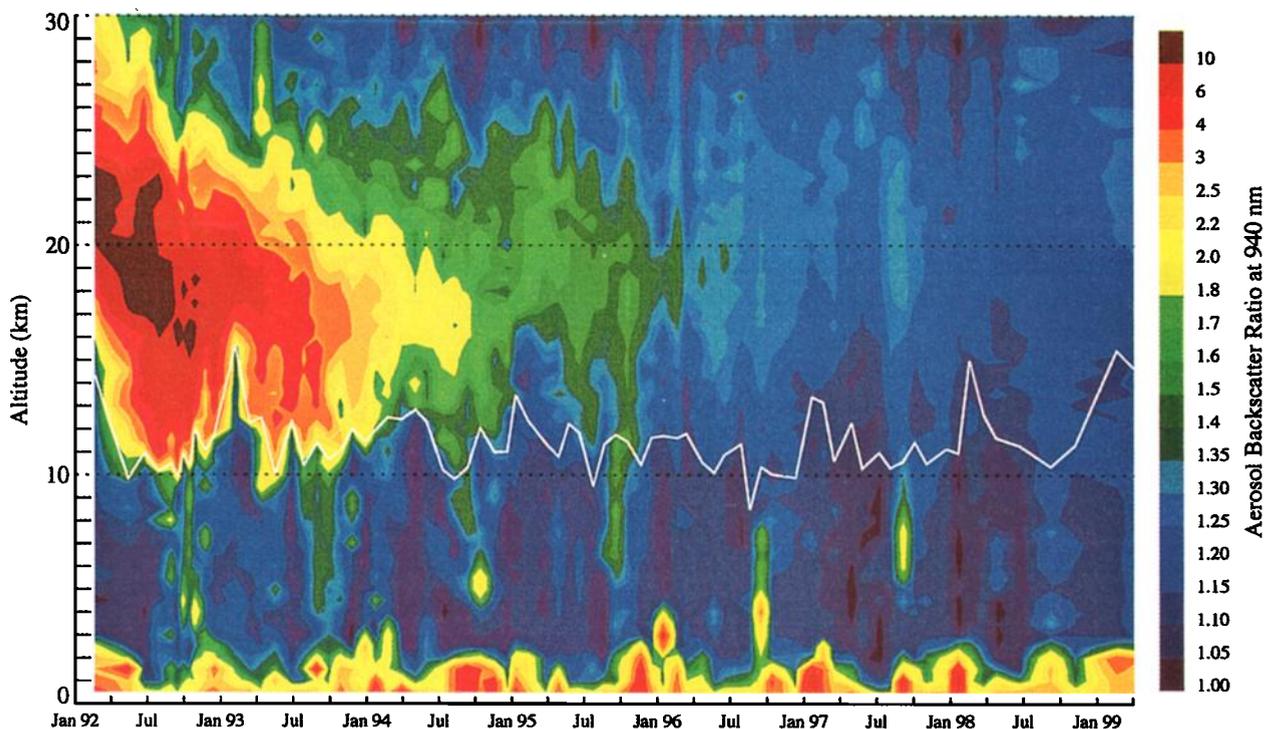
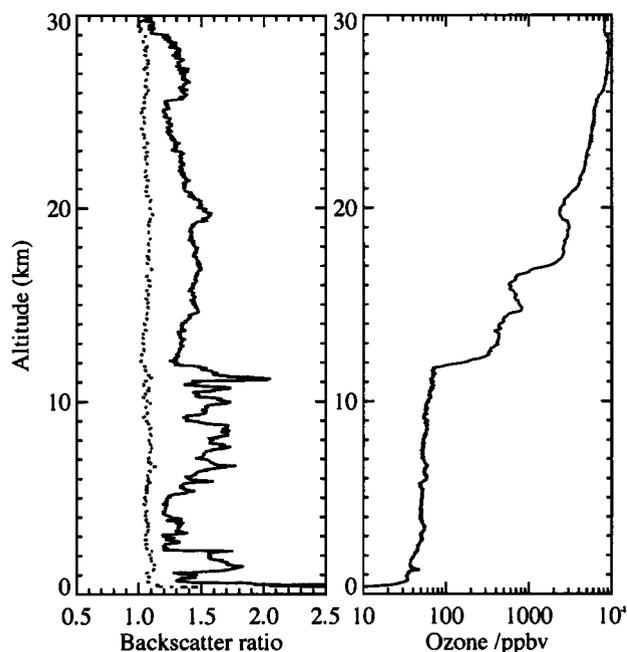


Figure 1. Backscatter ratio at 940 nm from monthly backscattersonde data. The tropopause height is shown in white.



**Figure 2.** Profiles of backscatter at 940 nm (solid) and 490 nm (dotted) and ozone mixing ratio from the sonde flight of September 19, 1995.

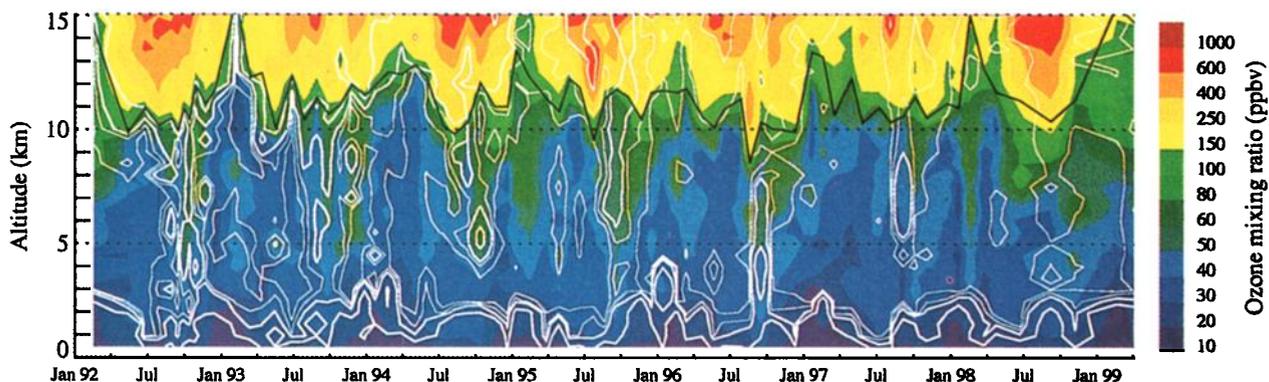
sions up to 15 km in the summer months. At the same time of year there is a major loss of aerosol from the 12–15 km altitude range and a concomitant reduction in the total column. It is apparent that lower stratospheric aerosol has passed into the troposphere whence it is removed by cloud and precipitation processes. The monthly time scale of sampling in this data set is much longer than aerosol lifetimes in the presence of cloud. It is also much longer than that of passing weather systems with which tropopause altitude varies throughout the range for that season (9–15 km in summer, 9–12 at other times). Monthly sampling thus cannot resolve any detail of aerosol or ozone removal from the troposphere, but clearly the large upward shifts in the tropopause in summer are associated with aerosol exchange to the troposphere. Even though exchange across the tropopause seems to be greatest in summer, it does not result in enhanced upper tropospheric aerosol at that time.

Figure 2 shows the aerosol backscatter profiles for flight 42 in September 1995. Enhanced aerosol concentration is seen

between 5 and 12 km altitude, with a sharp peak at 11 km, and there is substantial layer structure. Such layers are a common occurrence, as noted by *Newell et al.* [1999] who ascribe to them origins in the stratosphere or the boundary layer according to relative concentrations of ozone and water vapor. Note for flight 42 that the free-tropospheric maximum in backscatter ratio is also greater than its value anywhere in the stratosphere, so it cannot result from downward mixing within the column. The same constraint applies to all of the profiles in the latter half of the data series, but in the first three years mixing across the tropopause may contribute to the maxima in the free troposphere.

One mechanism for transport from the stratosphere which could concentrate aerosol to higher mixing ratios involves the polar vortex. Aerosols nucleate or are entrained into polar stratospheric clouds which sediment through the descending vortex and may subsequently evaporate to core aerosol. *Thomason and Poole* [1993] postulate that “this mechanism may, in fact, dominate the upper tropospheric loading in the southern hemisphere.” Their figure 3 series of vertical profiles by season shows very low aerosol concentrations in the mid stratosphere within the springtime Antarctic vortex, and also that lower stratospheric mixing ratios in spring exceed corresponding autumn values. Nevertheless the “weak maximum...near the 325 K potential temperature level” is consistently less than those in the broad mid-stratospheric peak, even before any wet removal in the transport of tropospheric aerosol to lower latitudes. While it does provide a route for the “maximum downward transfer of volcanic material into the upper troposphere...in local spring...at 70° S or greater in the southern hemisphere” observed by *Kent et al.* [1998a], this ‘vortex pump’ does not seem to give rise to profiles like that in figure 2 here. Nor does it account for the continuation of tropospheric seasonal maxima into the era of low stratospheric aerosol amounts.

Aerosol exchange from the planetary boundary layer should be greatest in summer, when there is more convection and total boundary-layer aerosol is greatest. The latter point is apparent in figure 1, but there are also boundary layer aerosol enhancements in springtime in most years of the record. In 1997 and 1998 they are coincident with the mid-tropospheric increase, and in 1995 and 1996 they seem to be connected even though the maxima do not occur at the same time. Grass fires are a frequent occurrence in Central Otago in early spring, and they are a likely explanation for boundary-layer aerosol enhancement then, but the smoke plumes do not extend to altitudes above 2–3 km.



**Figure 3.** Ozone mixing ratio from the monthly backscattersonde series. The tropopause height is shown in black. Contours of backscatter ratio at 940 nm are overlaid in white.

Ozone is enhanced in the upper troposphere over Lauder in springtime, as shown from analysis of the full ozonesonde time-series [Bodeker *et al.*, 1998]. For the subset of ozonesondes which are part of the backscattersonde package, the seasonality is apparent in figure 3, which shows in color the contours for ozone mixing ratio. Relevant contours of backscatter data are overlaid in black. Some correlation is visible but many instances of ozone increase are not associated with aerosol, especially in the summer months when tropopause height is most variable. Biomass burning can produce ozone in the troposphere through precursors including  $\text{NO}_x$ , CO, and hydrocarbons, but in different relative concentrations the same species can cause net ozone destruction [Isaksen *et al.*, 1992]. Tropospheric ozone is also increased by downward mixing from the stratosphere, and this is the probable origin for the higher ozone concentrations which are not correlated with aerosol.

Carbon monoxide is a more direct indicator of biomass burning, especially for the southern hemisphere where industrial sources are weaker. Since early 1994 a Fourier Transform Spectrometer has measured infrared spectral absorption of direct solar radiation at Lauder. From these measurements are derived the total column amounts of numerous trace gas species, and for some strong absorbers the line shape can be used to derive approximate profiles of the gas. A companion paper [Jones *et al.*, in press, 2001] compares CO data for Lauder with aerosol backscatter as described here and demonstrates that the two series correlate strongly. The CO mixing ratio in the stratosphere is consistently lower than in the troposphere, so it cannot be a source for relative enhancement as it is for ozone.

Monthly sampling at one location cannot begin to reconstruct hemispheric behavior or support an inventory of sources and sinks, but it can still constrain such calculations. In the backscattersonde data peaks in aerosol concentration are narrowly confined within springtime and often to a limited range of altitudes. These peaks can be very large, and it would seem that the broad seasonal peak apparent in zonal and seasonal averages may arise from such localised high concentrations. Significantly, the mixing ratio can be much higher than anywhere in the stratosphere, ruling out the latter as a source. In other profiles during the period of seasonal enhancement aerosol concentrations are much closer to the averages seen by SAGE, perhaps because the temporal and spatial distribution has been smoothed by turbulent diffusion during transport.

#### 4. Conclusions

The seasonal peak in upper tropospheric aerosol over Lauder is consistent with satellite observations and airborne studies. The lack of correlation with stratospheric and boundary layer aerosol and the high mixing ratios at the peak sug-

gest horizontal transport as the source. Correlations with ozone and especially carbon monoxide point to biomass burning origins.

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