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Abstract

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Keywords

biomass, evidence, 45, correlation, carbon, aerosol, burning, emissions, monoxide, GeoQuest

Disciplines

Medicine and Health Sciences | Social and Behavioral Sciences

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Correlation of aerosol and carbon monoxide at 45°S: Evidence of biomass burning emissions

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Abstract. Altitude profiles of Carbon Monoxide (CO) and aerosols have been compared from the Network for Stratospheric Change (NDSC) mid-latitude southern hemisphere site at Lauder, New Zealand. The CO mixing ratio profile was derived from infrared spectra recorded with a very high resolution Fourier Transform interferometer using three lines of the (1-0) band between 2057 and 2160 cm^{-1}. The aerosol surface area was derived from balloon-borne backscatter radiation at 940 nm. Both datasets show significant enhancements occurring over the observation site in the austral spring. When displayed together their combined effect illustrates the close correlation between CO and aerosols. Peak concentrations are consistently recorded between September and October over a five year time frame (1994-1999), with the enhancements typically occurring at heights of between 3 to 8 km. The temporal and spatial correlation between the aerosol plumes and enhanced CO concentrations are interpreted in terms of the effect of long range transport of biomass burning plumes in combination with the El Nino-Southern Oscillation (ENSO) cycles influence on southern hemisphere climate dynamics.

Introduction

Several key species which are used as indicators of tropospheric pollution and transport have been measured for a number of years at the NDSC, [*Kurylo,* 1991] station at Lauder, New Zealand (latitude 45.045°S, longitude 169.684°E, altitude 0.37 km). Among the techniques used on site is a very high resolution Fourier Transform Spectrometer (FTS) with an unapodised resolution of 0.0035 cm^{-1} (Bruker 120M) that has been used for the abundance determination of carbon monoxide (CO) and ethane (C_2H_6). Further, balloon-borne backscattersondes *Liley et al,* [2000] have provided a record of aerosol concentrations, another tropospheric key indicator. In this paper a series of measurements of CO from the FTS instrument is used to help interpret aerosol observations over the time period of 1994 through to the end of 1998.

The species CO has been chosen from a number of possible pollution indicators to compare with the aerosol data. The reason for this is in part to simplify the analysis, but also to reflects the importance of CO has as a factor in determining the concentration of other atmospheric oxidants [*Thompson et al.,* 1992]. The molecule has similarly been chosen by a number of intensive field and space-based campaigns, as well as having an extensive history of ground-based infrared solar spectroscopic measurements [see *Pougatchev et al,* 1999, and the references therein]. On the other hand there are still very few measurements in the free troposphere, [*Notholt et al.,* 2000], so that there is some geophysical interest in the CO measurements themselves.

Enhanced values of aerosols in the South Pacific free troposphere have been less studied than those from equivalent latitudes in the Northern Hemisphere [*Kent et al.,* 1998]. It is known that maximum aerosol concentrations in the mid to upper troposphere occur in the springtime [*Liley et al.,* 2000] but the exact origin and composition is not completely understood. Unlike CO and hydrocarbons, whose origins during tropospheric pollution episodes in the southern hemisphere are closely linked with biomass burning sources, the complex nature of aerosol sources (e.g., forest fires, volcanic eruptions, desert dust, industrial emissions) make it more difficult to associate observed aerosol plumes with a particular known source.

The purpose of this paper is to present co-located measurements, spanning 4 years, of aerosols recorded over Lauder from a balloon-borne sensor, in conjunction with CO concentration profiles derived from high resolution infrared spectra. To our knowledge this is a unique data set which illustrates the novel use of the combination of two quite different techniques that are complementary, which taken together add significant new information to the question of the nature and origin of tropospheric pollution plumes observed at the site of measurement.

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Method

The CO results have been reported in some detail in a previous paper by *Rinsland et al.*, [1998], but due to some minor differences in the spectral intervals used some explanation will be given here. The aerosol data set is described elsewhere and will therefore not be repeated here, [*Liley et al.*, 2000].

In the paper by *Rinsland et al.*, [1998], the Lauder CO data base is described in detail, along with the analysis method, the forward and inversion models (SFIT2 algorithm), instrument model considerations, as well as a discussion on sources of error. However in this current study, the selection of the spectroscopic microwindows is different for the following reason. In the earlier study, the purpose of the paper was to compare the column amounts between two gases, CO and C_2H_6, noting their mutual daily, seasonal and long trends. Because CO and C_2H_6 have very similar tropospheric lifetimes, and have a common principle loss via OH, it is important to sample the vertical structure of the atmosphere in a consistent manner [*Rinsland et al.*, 1998]. To achieve this, the spectral intervals used were constrained by the nature of the relatively weak absorption features available for C_2H_6 in ground-based spectra. The retrieval method uses the shape of the pressure-broadened lines to adjust the apriori mixing ratio of the gas in question. For a molecule like C_2H_6, with a vertical distribution that rapidly decreases with altitude in the troposphere, and weak spectral features, the resultant vertical sampling is weighted to the mid-troposphere. On the other hand, CO has a wide range of available absorption lines, so that in the current study we chose to use a combination of both weak and strong lines, resulting in a uniform sampling of the troposphere.

The CO absorption lines chosen for this study are part of the strong 1-0 band at 4.7 μm. This follows the study of *Pougatchev et al.*, [1995], except that the P(9) $^{13}C^{16}O$ was excluded due to a strong blend with a nearby O_3 line. The HITRAN96 linelist [*Rothman et al.*, 1998] was used for all spectroscopic lines. For all spectra, these three CO lines were fitted simultaneously. Figure 1 illustrates the mixing ratio averaging kernels [*Rodgers*, 1990] which provide a direct assessment of the theoretical altitude sensitivity of the measurements in the absence of measurement and forward model parameter errors. The signal to noise ratio of the spectra, defined here as the depth of the CO absorption divided by the random noise (as determined at the center of black lines), was about 400. In the retrieval process itself, the fitted signal to noise was set to a conservative 150, avoiding any potential retrieval instabilities, while still making use of most of the vertical profile information in the spectra. The *a priori* CO mixing ratio profile is the same profile used in *Rinsland et al.* [1998] and *Pougatchev et al.* [1998], and is plotted in figure 3 of the latter paper. The error budgets discussed in both *Rinsland et al.* [1998], and *Pougatchev et al.* [1998] are relevant to this study, except that due to the finer grid reported here (2 km) compared with the earlier studies (0-12 km column), the independent errors are approximately 2.5 times larger. Therefore both the random error and systematic errors are estimated to be ±5% for a single measurement.

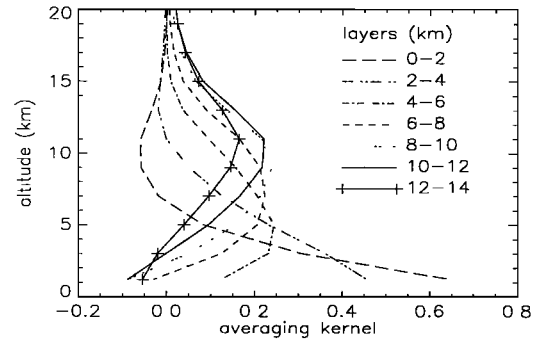

Figure 1. CO volume mixing ratio (VMR) averaging kernels computed for the individual 2 km thick layers from the ground to the tropopause. The finite width of the kernels gives a vertical resolution of approximately 5 km in the troposphere.

Results

The carbon monoxide and aerosol concentrations are plotted in figure 2. The CO data are shown as color coded mixing ratio (ppbv), while the aerosol data are overlaid as line contours of backscatter ratio. Neither data set was uniformly sampled in time, because of realistic constraints imposed by local weather conditions during measurement (low wind speed and no precipitation for balloon launches, sunlight for FTS measurements) and occasional instrument downtime. Vertical detail is also quite different in the two data sets. For the temporally more irregular CO data, Delaunay triangulation is combined with Akima's quintic polynomial interpolation onto a fine grid. The vertical sampling for CO is at 2 km intervals which reflects the internal forward model grid. This grid is on a relatively fine scale so that atmospheric radiative transfer calculations are accurately represented. From figure 1, the vertical resolution is about 5 km in the troposphere. Approximate monthly sampling in the aerosol data allows them to be contoured directly by line-following, but high vertical resolution emphasizes detail that is inconsistent with the monthly time step. As described in *Liley et al.* [2000], the aerosol data are smoothed to 0.5 km resolution, but they should not be interpreted as monthly means.

Presenting these ground-based infrared measurements on a vertical 2 km grid is also unique compared with previous ground-based CO results because historically they have been in the form of total column amounts. The exception is the paper by *Rinsland et al.* [1998], which published a single column amount for the troposphere. Despite the limited vertical resolution of the CO data, it is possible to infer information on scales of order of the grid spacing of 2km, like the center of mass of plumes. It is not possible however, to detect any details in the CO mixing ratio concentration on this fine scale.

The data as presented show two different geophysical features. First, both the CO and aerosol data sets ex-

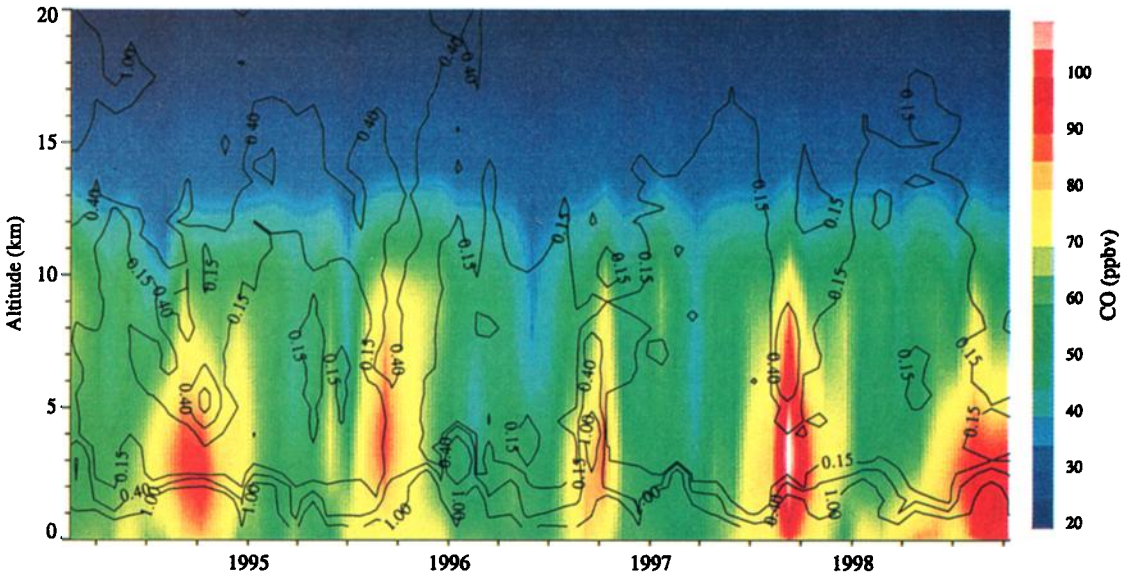

Figure 2. Profiles of CO mixing ratio plotted as color coded contours (ppbv), while aerosol is computed onto constant backscatter ratio surfaces. The aerosol contours are at backscatter ratios of 0.15, 0.25, 0.4, and 1.0 at 940 nm.

hibit a spring-time maximum, peaking in the mid troposphere (3-5 km). Second, the CO data set clearly indicates year to year variability, but this is not necessarily correlated with the aerosol data set to the same extent as the spring-time maximums. The measurement frequency of the CO data set is of the order of twice weekly, while the aerosol record is on a monthly basis. Keeping in mind this sampling difference, and the fact that unlike CO, aerosols are not necessarily a conserved quantity on the order of weeks to months, the level of correspondence between the two data sets is quite remarkable.

In the spring of 1994 (figure 2), the enhanced CO layer extended from the ground to approximately 5 km, with peak values at around 3 km. In the following year, 1995, the enhancement begins at about 2 km and extends to about 8 km, with a peak around 4 km. During the spring of 1997, the enhancement extends from the ground to 10 km, with the highest recorded enhancements in CO mixing ratio of > 110 ppbv centered at 3 km. The most recent year for which data is available, 1998, is slightly different again, with high values from the ground to only 5 km, but extending over a much longer time. The spring of 1998 and summer of 1999 were unusually hot and dry for New Zealand. The high values close to the ground could indicate, in this case, a local source (farmers' periodically burn-off vegetation to encourage spring growth, a practice that was noted in the Lauder meteorological records at the end of 1998). The aerosol data clearly correlate extremely well with the spring-time CO maxima. Comparing season to season however, the aerosol data tend to peak higher at about 5 to 6 km (1994, 1995, and 1997), while the highest recorded values in the mid-troposphere appeared in 1996, with a backscatter ratio > 1.0. In that particular year, the altitude of the aerosol peak is consistent with the maximum in CO concentration.

Discussion

The obvious temporal and to a lesser extent spatial correlation between the aerosol plumes and enhanced CO concentrations in figure 2 can be understood in terms of two effects operating on different spatial and temporal scales.

On time scales of order a year, the CO and aerosol are influenced by biomass burning plumes. During the NASA Global Tropospheric Experiment (GTE) missions which included Pacific Exploratory Mission (PEM) Tropics A [*PEM-Tropics A Science Team,* 1999; *Hoell,* 1999; *Singh et al.,* 2000] (September-October 1996)the measurement of a number of trace chemicals were reported, including CO, which were used as indicators of biomass burning activities. A major finding of the PEM-Tropics A mission was the pervasiveness of biomass burning plumes and their impact on trace gases throughout the southern pacific region [*Hoell,* 1999]. In particular a number of plumes, during Austral spring of 1996, were observed east of New Zealand in the middle troposphere. *Singh et al.* [2000] cite model studies [*Chatfield et al.,* 1999] suggesting that the observed plumes were the result of long-range transport of air originating near the southern tip of Africa and also Brazil. Reported calculations [*Pougatchev et al.,* 1999] performed by R.B. Chatfield specific to the Lauder situation used the three-dimensional Global Regional Atmospheric Chemistry Event Simulator (GRACES). The results indicated that during the months of September and October, Lauder can be within an air mass that is either influenced by biomass burning or originating from higher latitudes that is relatively clean. Also, air south of the South Pacific Convergence Zone had a chemical signature that was dominated by combustion products associated with biomass burning [*Gregory et al.,* 1999]. Most impor-

tantly, the five years of Lauder data confirms the conjecture [*Hoell,* 1999] that the effects of biomass burning is an annual process.

Dibb et al. [1999] found that the mixing ratio of aerosol associated soluble ions were low in air influenced by biomass burning (on the basis of other tracers like CO), suggesting that scavenging by precipitation had taken place. It is interesting to note that the plumes identified by *Singh et al.* [2000] were imbedded in dry to very dry air (less than 1000 ppm and 200 ppm water vapor respectively), consistent with subsiding long-range trajectories. At lower altitudes, large decreases in the concentrations of CO and O_3 to background levels of 50 and 30 ppb respectively were associated with more clouds and a moist boundary layer. Based on these reports, it would seen reasonable that while enhanced levels of both CO and aerosols should be recorded during the austral-spring period, the fact that they do not always correlate between measurements that are at best sampled monthly is not surprising.

The second effect, on inter-annual time scales, is the ENSO cycle. The ENSO index is defined by the Center for Ocean Atmospheric Prediction Studies (http://www.coaps.fsu.edu/~legler/jma_index1.html) as a 5-month running mean of spatially averaged Sea Surface Temperatures (SST) anomalies over the tropical Pacific: 4°S - 4°N, 150°W - 90°W. This index identifies the years of 1994, 1995, and 1996 as neutral (index values between $\pm 0.5^{\circ}$C), year 1997 as a warm phase ($> 0.5^{\circ}$C), and 1998 cold ($< -0.5^{\circ}$C).

The inter-annual behavior in the CO mixing ratio is consistent with moderate biomass burning activities in ENSO neutral years (1994-1996), large effects in ENSO warm years (1997), while the ENSO cool year (1998) has possible effects of local sources as mentioned above. The paper by *Kent et al.* [1998] examines the behavior of aerosols in the upper troposphere during the spring season in the southern hemisphere. Plate 8 of *Kent et al.* [1998], illustrates the difference in the Southern Hemisphere, centered at 6.5 km, for ENSO warm and cool years. The figure indicates significantly higher aerosol concentrations during the warm phase, with high extinction aerosols extending from South America at 10°S to 20°S around the globe, with enhanced aerosols to the south of Australia and north of New Zealand originating from this same plume. The Lauder aerosol record is not completely consistent with this picture, but this is most likely due to the combined effects of the infrequent nature of balloon flights coupled with aerosol scavenging processes.

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