

**Carbon Monoxide from Biomass Burning in the Tropics
and its Impact on the Tropospheric Ozone**

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Abstract:

Carbon monoxide (CO) measurements from the Measurements Of Pollution In The Troposphere (MOPITT) experiment are used to characterize the effects of biomass burning at several ozonesonde stations in the tropics, namely Reunion, Irene, Natal, Ascension, San Cristobal and Paramaribo. Distinct seasonal patterns of CO at each station indicate the strong influence of the African and South American biomass burning. While all stations show enhanced CO columns during September-November (SON), corresponding to the peak in austral burning, the effects of sub Saharan burning can be seen at Natal and Ascension and that of the northern Amazonian fires at San Cristobal. The CO variations are generally similar to the variations of aerosol optical depth retrieved from TOMS/NIMBUS-7 at most stations with notable differences at Irene and Reunion, indicating different sources for the aerosols. At Natal and Ascension, the CO peak lags the aerosol peak by 1-2 months consistent with the effects of deep convection. Tropospheric ozone from SHADOZ ozonesonde measurements at all stations show elevated levels corresponding to the CO enhancements in SON months. However there are several instances of ozone enhancements unaccompanied by any CO increase. This

might indicate that sources other than biomass burning such as stratospheric tropospheric exchange (STE) or lightning related NO_x may be operative. At San Cristobal strong CO enhancements during March-April are accompanied by only moderate increase of ozone, possibly due to low altitude transport from the source regions.

1. Introduction:

Some of the most interesting geographical areas from the point of view of tropospheric chemistry are the biomass burning zones of Africa and South America with their potential impacts on global climate (Crutzen and Andreae, 1990, Levine, 1996). Intense biomass burning takes place in the sub Saharan Africa (northern hemisphere) between December and February and in Southern Africa and Brazil between July and November (southern hemisphere). Crutzen and Carmichael (1993) state that biomass burning is believed to be the most important source of CO and NO_x in the tropics. These and additional products of the burning (PAN, NMHC, etc) are precursors of ozone. These precursors combined with the abundant sunlight and water vapor in the lower troposphere and convective venting in the upper troposphere make for a most interesting and complex atmospheric environment from the point of view of tropospheric ozone chemistry.

Tropospheric ozone columns estimated by residual techniques from satellite measurements as well as from seasonally averaged vertical profiles on cloud free days retrieved by SAGE II indicate the existence of a persistent ozone maximum in the South Atlantic area near zero degree longitude (the so called “wave 1”) (Fishman et al., 1992, Ziemke et al., 2001, Kar et al., 2002). This phenomenon has also been intensely studied from aircraft and ground campaigns as well as using models which indicate that biomass

burning in southern Africa and Brazil contribute significantly to this “wave 1”, particularly in SON months (e.g. Thompson et al., 1996, Diab et al., 1996, Jacob et al., 1996). However, the ozone distribution in the South Atlantic area is susceptible to a number of influences, and dynamics has been shown to be capable of producing “wave 1” even without any contribution from the biomass burning (Krishnamurty et al., 1993, 1996, Moxim and Levy, 2001). The ozonesondes from the SHADOZ program also indicate that the tropospheric ozone distributions have large variability with contributions from several sources (Thompson et al., 2003b).

It is therefore of crucial importance to delineate the various mechanisms contributing to the tropospheric ozone distribution in the tropics. While biomass burning is expected to significantly add to the tropospheric ozone burden, Thompson et al. (2001, 2003b) have noted that the ozone levels do not show a simple correlation with the biomass burning signal. They used the aerosol index derived from the TOMS backscatter measurements as a proxy for the biomass burning. A better measure of the latter would be concentrations of the ozone precursor gases like CO and NO₂. Jones et al. (2001) found similar spring time maximum in both aerosol and CO concentrations at Lauder, New Zealand, indicating long range transport of biomass burning plumes. CO measurements from aircraft campaigns have also been used to study their correlation with simultaneous measurements of ozone mixing ratios. Takegawa et al.(2003) found a positive correlation between the CO and ozone mixing ratios at low altitudes (< 3 km) for northern Australian biomass burning, thus indicating photochemical production of ozone from the precursors. With the availability of tropospheric CO measurements from the MOPITT instrument, it now becomes possible to assess the global effects of biomass burning in a more direct

way (Drummond, 1992). Edwards et al., (2003) have used CO data from MOPITT along with data from other satellites to study the effects of biomass burning in sub Saharan Africa during January, 2001. In this study we demonstrate the potential of the MOPITT CO measurements for qualitatively constraining the sources for the ozone variations in the tropics. We use ozone measurements from the SHADOZ program to study how they correlate with the coincident CO measurements at several tropical stations. Given the high spatial resolution of MOPITT, it is possible to obtain a sufficiently large number of CO profiles at these stations and thus study seasonal variations. These variations are compared with corresponding variations of TOMS/NIMBUS-7 aerosol optical depth climatologies to study how well the aerosol optical depth represents the biomass burning at these locations. Seasonal climatologies of CO and ozone are compared to identify the ozone variations which are associated with corresponding CO enhancements resulting from burning seasons in the northern and southern hemisphere.

2. Data

We use the CO data from the MOPITT instrument, which is aboard Terra spacecraft and was launched in December 1999. The spacecraft is in a sun synchronous polar orbit at an altitude of 705 km. MOPITT uses gas correlation radiometry at 4.7 μm and 2.3 μm to retrieve the CO mixing ratios (Drummond, 1992). It has a nadir view and a pixel resolution of 22x22 km and achieves a global coverage in about 3 days. The retrieval algorithm has been discussed in several publications (Pan et al., 1995, Edwards et al., 1999, Deeter et al., 2002,). We use validated version 3 data for the time period March 2000-May 2001 and provisional data for the period between August 2001-March 2003.

The difference between these two datasets is in the extent of validation that has been performed on the data as discussed in Emmons et al. (2003), with ‘validated’ being considered at a higher level of reliability than ‘provisional’. The CO vertical profile is reported at 7 pressure levels (surface, 850, 700, 500, 350, 250 and 150 hPa) with a precision of about 10%. The CO total columns have a similar precision of 10%. However the surface data is not used in this analysis. Analyses of averaging kernels have shown that retrieved volume mixing ratio has essentially two independent pieces of information (at 500-850 hPa and 150-350 hPa), resulting in a smooth profile. The ozone data are from the SHADOZ ozonesonde measurements, which report ozone mixing ratio with a precision of 5% (Thompson et al., 2003a, 2003b). For an independent measure of the biomass burning we have used aerosol optical depths as retrieved by the TOMS on board NIMBUS-7 using back scattered radiation in the near ultraviolet (Torres et al., 2002). These data are available for 1979-1992 and we have used them in a climatological sense.

3. Results

3.1. Seasonal distribution of CO in the tropics

Figure 1 shows the seasonally averaged distribution of the CO total column measurements in the tropics from MOPITT for the period March 2000 to March 2003. An intense plume of CO during December-February (DJF) is seen to extend from the east coast of Africa across the Atlantic. It is clearly associated with the savanna burning that takes place in the sub Saharan Africa at this time (Cahoon et al., 1992). The atmospheric lifetime of CO is about a month in the tropics, long enough to allow significant intercontinental transport [Staudt et al., 2001]. In March-May (MAM) period, savanna

fires decrease in intensity in the northern hemisphere due to precipitation. Significant fires occur in northern areas of South American continent during February-March and the CO plume from these fires is seen to move to the west, over the Andes and into the Pacific ocean areas. Around June-July, CO again starts increasing over Africa because of savanna burning in the south. The latter maximizes in SON period along with cerrado burning in Brazil. As can be seen, CO plumes cover the entire tropical belt between the west coast of South America and the east coast of Africa and beyond at this time, due to tropospheric air transported from Brazil over the Atlantic by high level westerly winds, and from Africa by easterlies in the lower and middle troposphere [Thompson et al, 1996, Jenkins et al., 1997].

The high level of CO from the biomass burning and its transport has interesting consequences for the tropospheric ozone distribution in the tropics, given the complex meteorology that prevails in this area (Krishnamurty et al., 1993). A network of ozonesonde stations under the SHADOZ program has been measuring the ozone vertical profiles in the southern tropics since 1998. We shall try to characterize the ozone distribution at 6 of these stations in terms of these CO measurements. The stations are marked in Figure 1. These are Ascension Island (7.98S, 14.42W), Natal, Brazil (5.42S, 35.38W), Irene, South Africa (25.25S, 28.22E), Reunion (21.06S, 55.48E), Paramaribo, Suriname (5.81N, 55.21W) and San Cristobal, Galapagos (0.92S, 89.60W). Estimates of tropospheric ozone columns have been made from satellites by residual techniques (Fishman et al., 1992, Kim et al., 1996, Ziemke et al., 2001). However there are differences between results from different techniques and the spatial resolution is also

limited. Therefore we have chosen to use the SHADOZ ozonesonde data for this work, particularly because the sonde data are available in the areas affected by biomass burning.

3.2. CO and aerosol variations at tropical stations

Figure 2 (left panel) shows an example of CO mixing ratio profiles retrieved by the MOPITT instrument near Natal in Brazil. All profiles within a radius of 500 km and within 24 hours of the given date were used to compute the mean profiles shown here. The mean profile on May 22, 2002 may be considered as the background CO profile, as biomass burning effects are minimal during this season (Figure 1). In comparison the mean profile on October 11, 2001 reflects the pollution from biomass burning reaching Natal from Brazil and Africa. As mentioned above, study of the averaging kernels and noise levels show that there are effectively two independent pieces of information in the vertical profiles retrieved by MOPITT. Despite the limited vertical resolution of MOPITT measurements, a general very significant enhancement of CO on October 11 can be clearly seen. On the right panel (Figure 2) the corresponding ozone mixing ratio profiles from the SHADOZ sondes launched from Natal are shown. The enhancement in the ozone mixing ratio in the lower and middle troposphere are consistent with the significant increase in the CO mixing ratios retrieved by MOPITT, particularly in the lower levels. The ozone profile shows a strong maximum (> 100 ppbv) in the upper troposphere which might indicate effects of dynamics, e.g. mixing of stratospheric air which would be consistent with the lower increment in the CO mixing ratios at these altitudes.

Figure 3 shows the climatology of seasonal variation of CO at the 6 selected SHADOZ stations. Data for the period March 2000-March 2003 were used to obtain this

climatology. The coincidence box is a circle of radius 500 km around each station. Each dot represents an average of all single measurements (typically there were about 200-300 profiles) for each day within the corresponding coincidence box for each station. Also plotted are corresponding monthly mean aerosol optical depths as estimated from the TOMS/NIMBUS-7 for the period 1979-1992. Once again a radius of 500 km around each station was taken for calculating the mean values. The TOMS aerosol optical depths as well as an aerosol index have been used as a proxy for biomass burning in various publications (Thompson et al., 2001, 2003b, Torres et al., 2002). Therefore, it should be interesting to compare the CO measurements with the aerosol optical depth. The stations form 3 distinct groups in terms of their CO variation. Irene and Reunion (panels a and b) are clearly similar with one strong CO maximum around October, consistent with biomass burning effects reaching this area from southern African regions (Randriambelo et al., 2000, Thompson et al., 1996). The aerosol optical depth also shows a strong enhancement at both stations around September-October, reaching a peak in September at Reunion (note the different scale for aerosol optical depth for Reunion). However, there seems to be no corresponding CO increase for the early part of the year when the aerosol optical depth is high at both the stations. Further, the aerosol optical depth at Irene continues to increase beyond the CO peak in October. The reason for these discrepancies is not clear, but Piketh et al. (1999) have pointed out that aeolian dust and industrial sulfur are the primary components of the aerosol load in the south African region throughout the year, rather than the aerosols from biomass burning. This may therefore be an example of two sources for the aerosols, inorganic around the turn of the year and organic (biomass burning) in the SON period.

At Natal and Ascension Island (panels c and d), a strong enhancement in CO is also seen during January-February as well as the peak in October and a clear minimum around May. The former is the signature of the sub Saharan burning during December-February (DJF) as may be seen from the CO total column maps (Figure 1) (Edwards et al., 2003). The aerosol optical depth at both these stations are by and large similar to the CO variations, except a phase difference of about one month for the SON peaks. This may be due to deep convection in the source regions, namely in South America in September-October which vents the biomass burning products (CO and ozone) in the upper troposphere followed by advection to Ascension and Natal (Diab et al., 1996). The signature of deep convection with the onset of the wet season can be seen in the decrease of aerosol optical depth, because the associated wet removal mechanisms can be efficient in removing accumulation mode aerosols (Andreae et al., 2001). This might explain the sharp drop in aerosol optical depth at Ascension around October and a similar drop at Natal although to a much lesser extent. Deep convection has been shown to play an important role in large scale transport of biomass burning products in Africa and South America (Thompson et al., 1996, Diab et al., 1996, Pickering et al., 1996, Jenkins et al., 1997, Andreae et al., 2001).

At San Cristobal (panel e) the aerosol and CO variations are similar with a maximum around March and a smaller maximum around September (note the change of scale for the aerosol optical depth). The former is likely related to fires that occur in the northern Amazonia around February-March while the secondary enhancement in September reflects the transport of biomass burning products from Brazil (Figure 1). At Paramaribo (panel f) CO is generally enhanced from January to March, receiving

pollution from northern African burning (DJF) as well as from fires in northern Amazonia in March-April. CO increases again starting September, when the burning peaks in both South America and Africa. However, the aerosols at Paramaribo do not seem to follow the CO enhancements from the austral burning having a strong peak in April followed by a steady decrease through the rest of the year. The reason for this is not clear, however, it is possible the aerosols are affected by local processes including precipitation patterns.

In general, the CO distribution at all the stations seem to be strongly influenced by the biomass burning in Africa and South America that reflects in distinct seasonal variations at each station. The aerosol optical depths obtained from TOMS measurements at most stations reflect the peak burning during September–October. However there are significant differences between the CO and aerosol variations particularly at Reunion, Irene and Paramaribo.

3.3. Comparison between seasonal variations of CO and ozone columns

Figure 4 shows the comparison between the seasonal climatologies of CO and ozone tropospheric column density at the SHADOZ stations. The ozone mixing ratio profiles between 900 hPa to 100 hPa have been used to compute the ozone columns in Dobson units. In some cases the data had to be interpolated to 100 hPa. As mentioned by Thompson et al. (2003b), there are large variations in ozone mixing ratios at each station indicating several contributing factors. At Reunion (panel b), ozone columns are highest in September-October, in phase with the CO peak, indicating the direct effect of biomass burning. During this period, the forest fires in South Eastern Africa and Madagascar maximize. High concentrations of ozone precursors from these fires are vented into the

upper troposphere by deep convection around this period and are then advected into the Indian ocean region by westerly winds (Randriambelo et al., 2000). However note that ozone columns at Reunion are also quite high during April-May period and Thompson et al., (2003b) attributed these to the central African burning. However this is not borne out by the CO measurements at Reunion, which do not show any concomitant enhancements during this period. A similar CO-ozone correlation can be seen at Irene (panel a) as well.

The climatological correlation between CO and ozone columns is significantly better at Natal and Ascension Island (panel c and d) with ozone columns reaching highest values in October in phase with the CO peaks and about 1-2 months later than the aerosol peak (indicating peak burning). As mentioned above, deep convection in South America and associated formation of ozone in the post convection cloud outflows primarily controls the distribution of ozone at Natal and Ascension during SON months (Thompson et al., 1996, Pickering et al., 1996, Diab et al., 1996). Ozone columns also show increased values during DJF months, but do not reach as high values as in SON. This is interesting because CO columns indicate similar strengths in both DJF and SON months. This is probably because ozone from the sub Saharan burning during DJF remains largely confined to lower troposphere (Martin et al., 2002, Edwards et al., 2003). At low altitudes the concentration of ozone is lower than would result from the photochemical production because of various ozone removal mechanisms including surface deposition (Delany et al., 1992). This combined with a lower lifetime leads to a lower overall ozone concentration during DJF than during SON. In other words the situation during DJF is one of ‘cook and mix’, wherein ozone is photochemically produced at lower altitudes and then vented to the free troposphere. This is likely to be less efficient compared to the

‘mix and cook’ mechanism where large amounts of the ozone precursors are quickly vented to the upper troposphere by deep convection followed by production of ozone in cloud outflows (Chatfield and Delany, 1990, Pickering et al., 1996). The latter is the dominant mechanism during SON.

At both San Cristobal (panel e) and Paramaribo (panel f) the CO and ozone columns were found to be uncorrelated. At San Cristobal the ozone columns are the lowest among all the stations, reaching highest values of about 25-30 DU during SON months. In particular, elevated levels of ozone columns are not seen in association with the high CO column values of March-April. At Paramaribo, the ozone columns are larger during SON (~35 DU) than at San Cristobal, but overall the correlation with CO columns is not significant.

To summarize, vertical columns of MOPITT CO and SHADOZ ozone sonde measurements show significant correlation over the entire year for four of the six stations, while in the cases of San Cristobal and Paramaribo there was essentially no correlation. We further discuss the ozone variations at these stations in the following section.

3.4. Time-altitude climatology of CO and ozone

In order to gain further insight we have plotted climatological CO and ozone time-altitude cross sections or ‘curtain’ plots at all the stations using all the available data between March 2000-March 2003 (Figure 5). Average of 5 day bins have been used to generate these ‘curtain’ plots for both CO and ozone. The white regions indicate the missing data from the ozonesondes. At all the stations, ozone mixing ratios show significant enhancement in SON, consistent with high CO levels from biomass burning. However, at both Reunion and Irene, high ozone mixing ratios in the middle and upper

troposphere could be seen during much of the year that are unrelated to CO enhancements from biomass burning. This enables us to delineate the effects of biomass burning clearly from other sources of tropospheric ozone at these stations. Recent studies have shown that Stratospheric-Tropospheric Exchange (STE) can take place near the edge of the tropical zones (Gouget et al., 1996, Folkins and Appenzeller, 1996, Baray et al., 1998, Zachariasse et al., 2000). In particular STE has been invoked in recent years at Reunion to explain observations of tropospheric ozone increases (Randriambelo et al., 1999, 2000, Baray et al., 2000). Another important source of upper tropospheric ozone could be lightning. Edwards et al. (2003) have shown that there was a strong plume of NO₂ over southern Africa in January 2001, which was related to strong lightning activity in this region. Similar NO₂ enhancements were also seen in this region almost year round in maps obtained from the GOME satellite (A. Richter, private communication) for the years 2000-2002.

At Ascension, the ozone response to CO from the two distinct biomass burning seasons can be distinguished. However Ascension is also subject to strong subsidence round the year (Krishnamurty et al., 1993, Thompson et al., 1996, Diab et al., 1996), which may be supplying ozone throughout the year. In particular ozone enhancements in the middle and upper troposphere are seen during JJA months. In contrast at Natal the ozone distribution seems to be primarily influenced by CO variations.

At San Cristobal the 'curtain' plot for CO shows the signatures of transported CO for several distinct biomass burning seasons. The ozone mixing ratios in March-April show a moderate enhancement in the middle troposphere (40-50 ppbv) corresponding to the strong CO enhancements. Oltmans et al. (2001) had earlier studied the ozone

distribution at San Cristobal and noted the lack of very low mixing ratios in the middle and upper troposphere during January-May, which they attributed to low convective activity at this site. The CO measurements from MOPITT indicate that the enhanced CO levels from burning in northern Amazonia may be the reason for this. However, note that the CO mixing ratios in March-April in the lower and middle troposphere at San Cristobal (≥ 150 ppbv) are of the same level as Ascension, Natal or Irene during the most intense southern burning (SON), and yet the corresponding ozone levels are much lower at San Cristobal. This might be related to the flow patterns in this area. Oltmans et al. (2001) found that in general, at 6 km some 75% of the back trajectories arriving at San Cristobal would have passed over the South American continent in the last 10 days, while at 13 km, the trajectories indicate stronger flow from the west. If the flow occurred at low altitudes, then the ozone increase at San Cristobal may be less evident either because of low ozone lifetime, or because of low ozone production at the source. The latter situation may be similar to Kalimantan in Indonesia in 1997 when low ozone concentration was observed in the lower troposphere despite sufficient NO_x obtained from Indonesian biomass burning (Tsutsumi et al., 1999, Kita et al., 2000). They attributed this to reduced solar flux levels because of high aerosol layer. In contrast the ozone increases in SON at San Cristobal in the middle and upper troposphere are much stronger despite the lower CO levels reaching San Cristobal. About half of all trajectories at 13 km during June-November originate from the east and north east (Oltmans et al., 2001), which would bring in the ozone formed in highly efficient post deep convection outflows (“mix and cook”) from Brazil. Biomass burning products from Brazil are transported to both east

and west during this period and can reach San Cristobal in a few days time (Oltmans et al., 2001).

Paramaribo in Suriname is near the middle of the latitudinal migration zone of the Inter Tropical Convergence Zone (ITCZ) and may often be north of ITCZ during the short dry period when biomass burning occurs in Northern Brazil, Venezuela and Guyana (February-March) period (Fortuin et al., 2000). Because of the dominant lower tropospheric trade winds from the northeast, Paramaribo is not expected to receive large amounts of biomass burning products from these fires. However, biomass burning products can be transported to the ITCZ from the burning zones, which can then be lofted to the upper troposphere and be transported towards Suriname. Andreae et al. (2001) found evidence of strong enhancements of CO and ozone in smoke plumes over Suriname, about 500 km upwind of the northern Amazonian fires. Fortuin et al. (2000) also found evidence of cross-equatorial transport of air in the middle troposphere level over Paramaribo. This might explain the stronger ozone levels over Paramaribo during March-April than over San Cristobal (Figure 5). During August to November, Paramaribo is south of the ITCZ and hence can receive biomass burning products by south-easterlies from Brazilian fires, which show up as strong enhancements in the middle and upper tropospheric ozone during SON months. However substantial ozone enhancements are also seen during other months often unaccompanied by any CO increase (Figure 5).

4. Conclusions:

We have used global CO satellite measurements by the MOPITT instrument to characterize the ozone distribution at several tropical ozonesonde stations. Distinct patterns of CO variation were seen at the various stations, which clearly indicate the strong influence of the biomass burning at these stations. Aerosol optical depths retrieved globally by the TOMS instrument have been earlier used as a proxy of the biomass burning at various places. Comparison of the aerosol optical depths with CO measurements helps delineate the aerosol variations due mostly to biomass burning. In particular, the early year aerosol enhancements seen at Reunion and Irene have no correspondence with CO—and thus may not be related to biomass burning. At Ascension and Natal, deep convection plays an important role in shaping the CO variations, which maximize after about 1 month of peak biomass burning indicated by the aerosol data.

The ozone measurements from SHADOZ program show a general enhancement in ozone columns during SON at all the stations in consistence with the CO levels measured by MOPITT. In addition at Ascension and Natal, there is evidence of a secondary increase in ozone correlated with the CO increase seen in DJF months from sub Saharan burning. On the other hand at San Cristobal, ozone increases in March-April were low despite high CO levels, presumably from the burning in northern Amazonia. There were also ozone enhancements at some stations notably at Reunion and Irene during DJF and Ascension and Paramaribo during JJA, which were accompanied by low CO levels, ruling out biomass burning as a source for these increments.

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Figure captions:

Figure 1: Seasonal MOPITT CO total column densities for December-January-February (DJF), March-April-May (MAM), June-July-August (JJA) and September-October-November (SON) and the sites of the six ozonesonde stations discussed (shown as filled red circles).

Figure 2.: Vertical profiles of CO (left) and ozone (right) mixing ratios from MOPITT and SHADOZ sonde measurements respectively for May 22, 2002 (grey) and October 11, 2001 (black) near Natal, Brazil.

Figure 3.: Climatology of MOPITT CO (red dots) total columns and TOMS/NIMBUS-7 monthly average aerosol optical depths (filled blue circles) for 6 tropical locations. Each red dot represents the average of all measurements within a radius of 500 km centered at each station. The data period refers to March 2000-March 2003 for MOPITT and 1979-1992 for TOMS/NIMBUS-7. The aerosol data have been plotted at the mid point of the months.

Figure 4.: Same as Figure 3 with tropospheric ozone columns (integrated between 900 hPa and 100 hPa in Dobson Unit) from SHADOZ sonde measurements as the blue dots. All the available sonde data between March 2000-March 2003 have been used.

Figure 5: Climatological (March 2000-March 2003) time-altitude cross sections for CO and ozone mixing ratios at the 6 tropical locations binned in 5 day cells. White areas correspond to missing sonde data.

Figure 1:

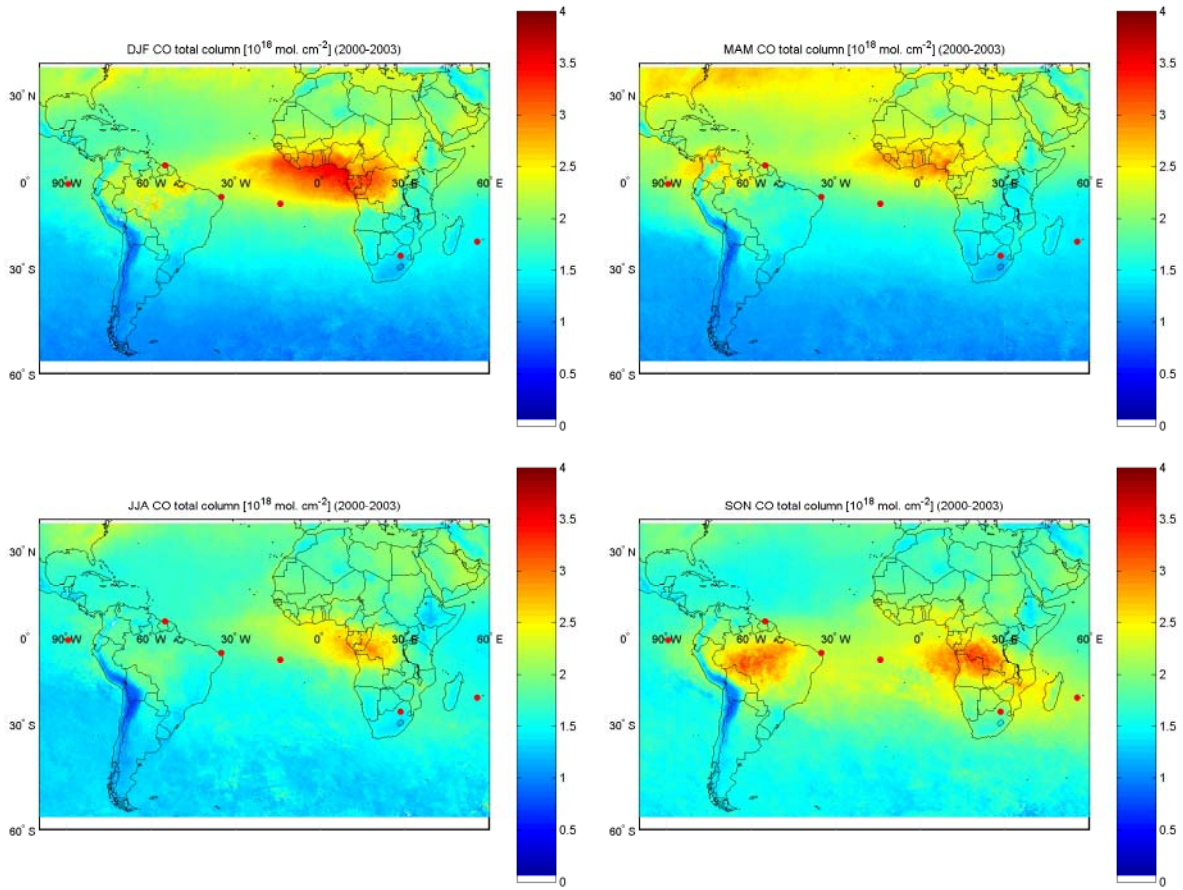


Figure 2:

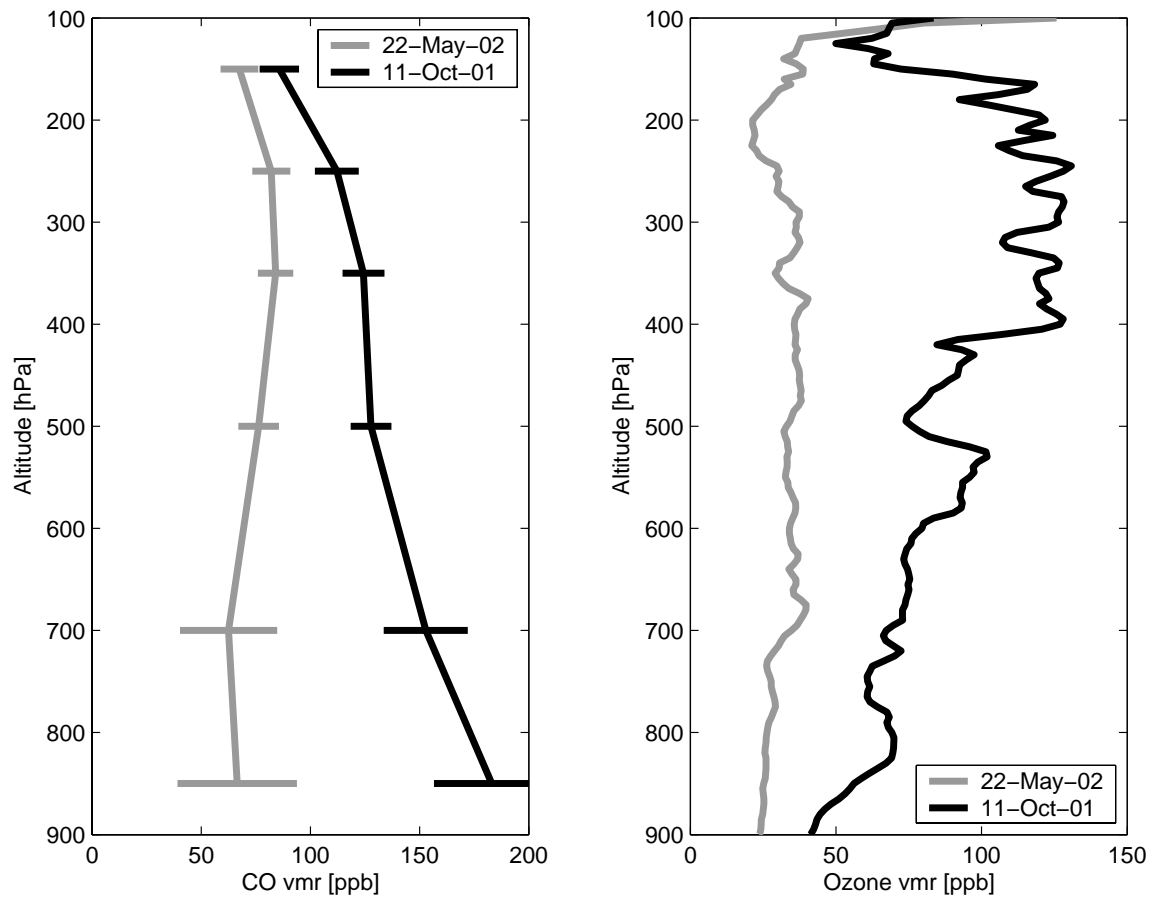


Figure 3:

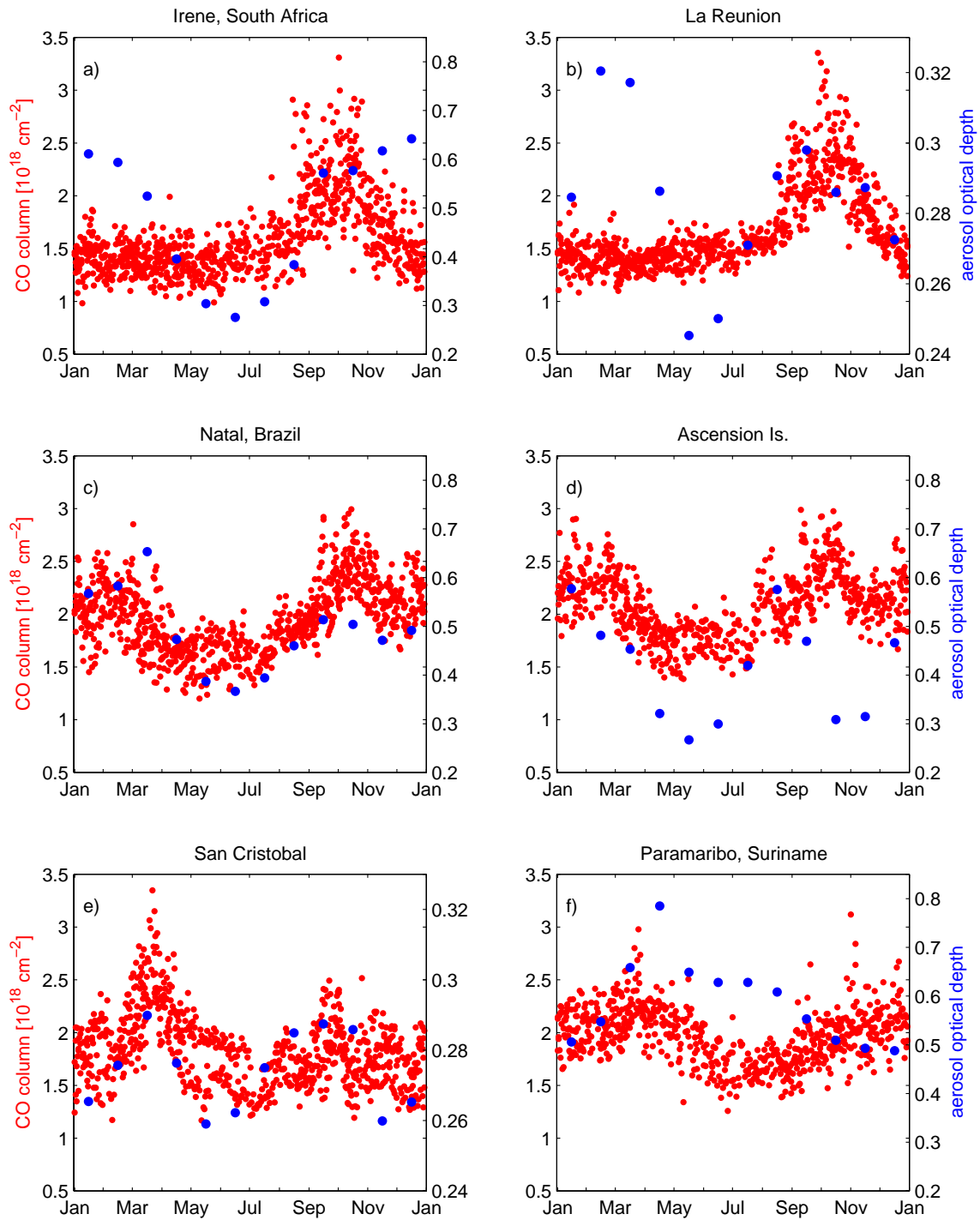


Figure 4:

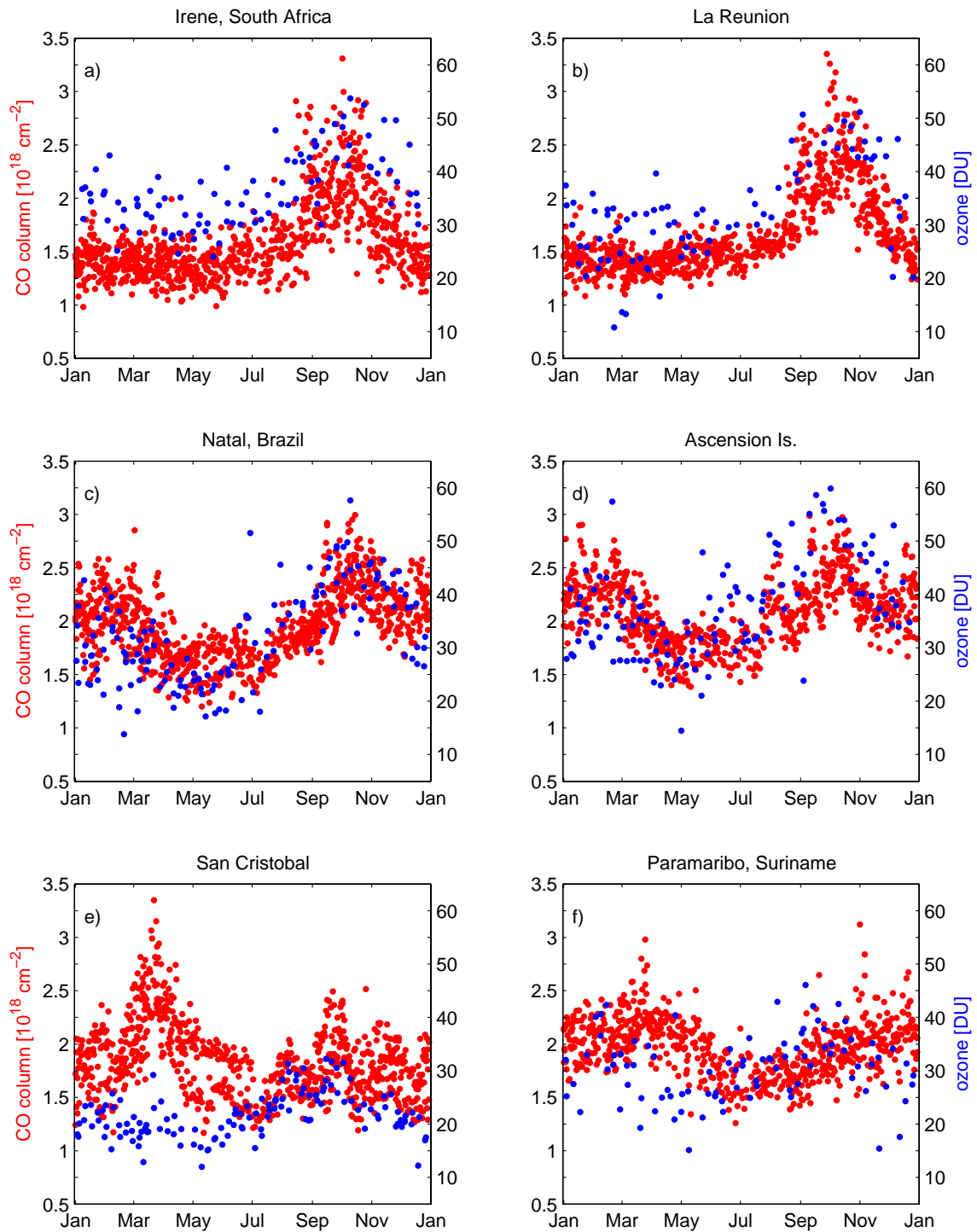


Figure 5:

