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An analysis of ozone measurements at Cerro Tololo (30°S, 70°W, 2200 m.a.s.l.) in Chile

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ABSTRACT

Increases in tropospheric ozone (O_3) abundance are likely to take place in the near future in the populous and rapidly developing countries in the tropics and subtropics. An accurate evaluation of the future impact of increasing industrial activities in tropical and subtropical areas requires knowledge of the background levels of ozone. New ozone monitoring stations have been installed at several sites by the World Meteorological Organization (WMO) since the mid-90s. We analyze ozone data collected during two years since April 1996 at Cerro Tololo (30°S, 70°W, 2200 m.a.s.l.) some 50 km east from the city of La Serena. In this paper, we describe some of the atmospheric chemistry and meteorology that characterizes the Tololo site. The data show a seasonal variation with maximum mixing ratios in late winter and spring and minimum mixing ratios in late summer and early fall. These variations are most likely associated with the large-scale subsidence of the Hadley circulation and the location of the subtropical jet stream (STJ). Also, there is a diurnal variation that is probably partly associated with a mountain wind flow which is strongest in late spring and summer months. No significant mixing with marine boundary layer air perturbed by anthropogenic activities is apparent from the data. We find the Cerro Tololo site to be generally representative for background conditions of free-tropospheric air in the subtropics of the Southern Hemisphere. This work is done within the framework of a larger effort recently started by several Chilean institutions in cooperation with research centers abroad.

1. Introduction

Changes in ozone abundance are likely to take place in the near future in populous and rapidly developing countries in the tropics and subtropics (Crutzen, 1995). Knowledge about the unperturbed atmosphere is required to assess the magnitude of the impact of anthropogenic activities on the atmospheric composition in these areas. Within this framework, efforts have been made to install ozone-monitoring stations at several background sites in the tropics and subtropics. One of these recently installed stations is the one located at Cerro Tololo (30°S, 70°W, 2200 m.a.s.l.) near the city of La Serena in Chile (Fig. 1). This station is part of the Global Atmospheric Watch (GAW) program developed by the World Meteorological Organization (WMO). The station is run by the Chilean Meteorological Service in cooperation with the Interamerican Southern Astronomical Observatory.

This paper addresses the analysis of ozone data collected during two years at Cerro Tololo

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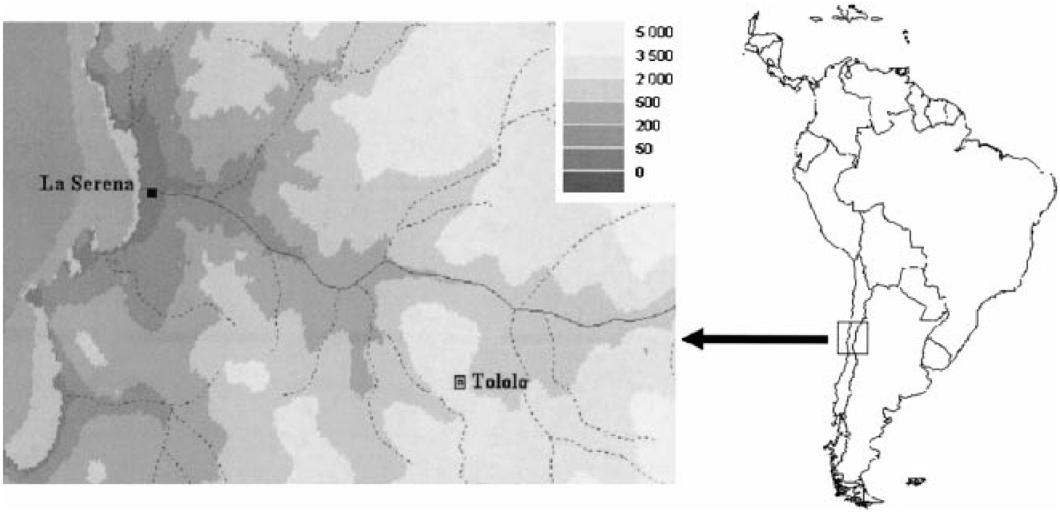

Fig. 1. Location of the observation site at Cerro Tololo (30°S, 70°W, 2200 m.a.s.l.). The map to the left shows altitude contours in meters for the intervals: <0, 0–50, 50–200, 200–500, 500–2000, 2000–3500, 3500–5000 and and above 5000 m.

beginning in April 1996. Our purpose is an initial characterization of the observation site with respect to meteorology and atmospheric chemistry aspects. Also, within the framework of a larger effort started by several Chilean institutions in cooperation with research centers abroad, our aim is to improve Chilean capabilities in atmospheric chemistry and meteorology. We envisage the Cerro Tololo site to become a fully equipped station, which will be helpful for a better understanding of global and regional climate change issues. This program is known under the Quechua Indian word "QHAWAYRA" which means "Air survey".

In the following sections, we describe the ozone data collected at Cerro Tololo and the weather patterns of the area (Section 2). Section 3 shows the analysis of data with respect to diurnal and seasonal variations. A few statements are made regarding interannual and day-to-day variations. Conclusions and some perspectives for future work are presented in Section 4.

2. Ozone measurements and weather patterns at Cerro Tololo

2.1. *Instrumentation and data*

The instrument used at the Cerro Tololo site is an Analyzer Sensor TECHO 49–003. The measurement principle is the absorption of ultraviolet (UV) radiation at 254 nm. This instrument is able to measure ozone-mixing ratios up to 500 ppbv, with 3% of accuracy. The data considered in the analysis are hourly averages of ozone, temperature and relative humidity for the period April 1996 through April 1998.

Synoptic scale analysis for individual, monthly and seasonal studies of the atmospheric circulation during the period under investigation, were carried out using the NCEP/NCAR reanalysis digital data (Kalnay et al., 1996).

2.2. *Meteorological description of the Cerro Tololo site*

The city of La Serena and the Cerro Tololo site are under the influence of the South Pacific high, which produces a permanent subsidence in the area interrupted occasionally by passing fronts or cut-off lows during winter. The thermal gradient between the equator and the pole is stronger in winter than in summer. This implies that the subsident branch of the Hadley circulation is more intense in wintertime than in summertime. Fig. 2 schematically depicts the main meteorological features that affect and modulate the climate along the West Side of the Southern South America.

A cold oceanic current system (Humboldt

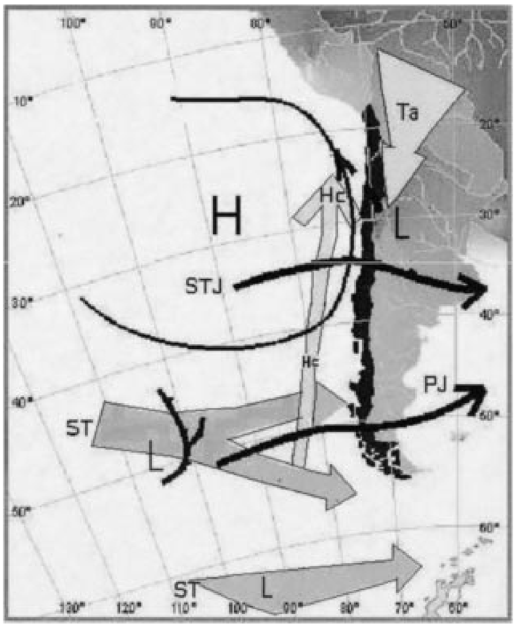

Fig. 2. Main atmospheric features that affect and define the climate of Chile. H: southern high-pressure center. L: low-pressure system associated with fronts. FT: main trajectories of frontal systems. Ta: Tropical air, convective tropical activity during the summer. Hc: Oceanic Humbolt current.

Current) that moves equatorwards along and offshore of the Chilean coast (Fig. 2), contributes to the strength of a low-level subsidence inversion between approximately 28°S and 36°S. According to the average wind profile at the Quintero radiosonde station (31°55′S, 71°31′W), southwesterly winds of about 3 ms^{-1} affect the region from 900 to 1500 m.a.s.l. during spring, summer and autumn. Northwesterly winds of about 5 ms^{-1} prevail during the winter season due to more frequent storm systems that cross the central-northern part of Chile. Above 1500 m.a.s.l., northwesterly winds of about 4–6 ms^{-1} prevail year round (Gilford et al., 1992). A monthly behavior of the wind can also be derived from the NCEP/NCAR reanalysis data. For this, a grid point concurrent with the Cerro Tololo site was selected (30°S, 70°W). At 925 hPa level, the reanalysis data show that southwesterly winds of about 2 ms^{-1} prevail from November to March, while northwesterly winds of about 3–4 ms^{-1} prevail from April to October. The same behavior is resolved at 850 hPa. From 700 hPa and above, northwesterly winds prevail year round and the wind speeds increase during the winter months.

Persistent stratus clouds off the northern Chilean coast, which are formed within the marine boundary layer, affect La Serena and surrounding coastal region. The vertical development of these low clouds is capped by the subsidence at about 1500 m.a.s.l.. An examination of radiosondes data from Quintero revealed that the subsidence inversion most of the time is below 2000 m. In addition, the diurnal variation of the air temperature obtained from the NCEP/NCAR reanalysis data suggests that, on average, the boundary layer is constrained below 850 hPa. This indicates that the atmospheric coastal conditions described before do not reach the Cerro Tololo site because its location (2200 m.a.s.l.) should be above the inversion layer. Therefore, Cerro Tololo most of the time is immersed in the free atmosphere and affected by the subsidence regime of the South Pacific high that brings clear sky conditions most of the year.

The wind pattern above 700 hPa reveals that the area of Cerro Tololo is within the northern part of the westerly regime of the midlatitudes. During summer, the climatological maximum upper-level winds (subtropical jet stream, STJ) are located near 200 hPa and the jet crosses the Andes at about 37°S. A second branch with a more northerly direction flows across the northern Andes at about 23°S playing a role in the development of the Bolivian high. In winter, the STJ is, on average, located around 30°S. The polar jet stream (PJ) near 300-hPa is located around 50°S in summer and it moves to around 40°S in winter, however it occasionally reaches the La Serena/Cerro Tololo sector.

On top of the large-scale features, a mountain wind flow exists in the Cerro Tololo area, with up-slope winds during the daylight hours and down-slope winds at night. This radiation driven circulation is stronger in summer than in other seasons. Nevertheless, it is worth noting that recent observations in the Antofagasta region, some 600 km north of Tololo, show a stronger down-slope flow during winter (Rutllant, personal communication).

The Hadley and Walker circulation including the interannual variability associated with El Niño and La Niña events also play a role in the climatological characteristics of the Cerro Tololo site and surrounding areas (Rutllant and Fuenzalida, 1991). In fact, during the period described here, a strong El Niño event took place (Fawcett, 1998; Walland, 1998). It began around March 1997 and ended in March 1998. This event was preceded by a weak La Niña episode in 1996. These phases of the Southern Oscillation imply several changes in the circulation patterns in the area (Aceituno, 1988; Rutllant and Fuenzalida, 1991). During El Niño events, there is an anomalously weak and northward displacement of the South Pacific high, allowing atmospheric conditions for an enhanced frontal activity and rainfall in central Chile in winter. La Niña events show opposite conditions.

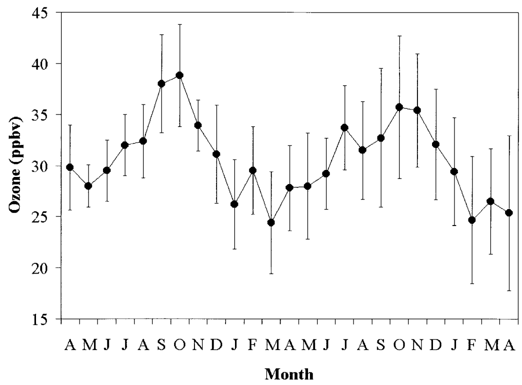

Fig. 3. Monthly averages of ozone volume mixing ratios at Tololo. The observation period is between April 1996 until April 1998. Error bars indicate one standard deviation of the mean.

3. Data analysis

3.1. Seasonal variation

Stratospheric air, rich in ozone, enters the troposphere through tropopause breaks that occur in connection with large-scale cyclogenesis, cut-off lows and quasi-adiabatic transport along isentropic surfaces (Holton et al., 1996). This air is brought down by large-scale subsidence in the subtropics. Therefore, it seems plausible to assume that the strength of the subtropical subsidence and especially the location of the STJ play a prominent role in the seasonal variability of ozone measured at Tololo.

The late winter and spring maximum in ozone observed at Cerro Tololo (Fig. 3) is apparently linked to downward transport of O_3-rich air from the upper troposphere and lower stratosphere. This transport is intensified during late winter and early spring when the southern branch of the Hadley circulation is strengthened. The result is that the Pacific high-pressure center takes a more southern position and the STJ crosses the Andes, on average, to the north of 30°S. Therefore, from early winter until spring the Cerro Tololo site is immersed in the large-scale subsidence of the Pacific high. The monthly average maximum wind speeds derived from the NCEP/NCAR reanalysis data for the period under consideration, revealed that during the two maximum peaks of ozone (cf. Fig. 3) the core of the STJ was around 25°S.

Model calculations made by Roelofs and Lelieveld (1997) suggest that there is a free tropospheric ozone maximum during spring in the southern hemisphere due to increased convective transport of ozone precursors probably associated with biomass burning. These calculations are consistent with tropospheric ozone column data derived from the total ozone mapping spectrometer (TOMS) and the stratospheric aerosol and gas experiment (SAGE) measurements that show maximum O_3 values during the austral spring, coincident with when biomass burning takes place in Africa and South America (Fishman et al., 1990).

At this stage, it is difficult to assess a contribution to ozone measured at Cerro Tololo from biomass burning sources in the tropics. However, the typical circulation patterns in the area make unlikely a major contribution of biomass burning sources in the tropics, at least in South America. If any, a contribution of tropical sources (see big arrow, Ta, in Fig. 2) is more likely in the austral summer when convective activity over the Andes takes place. On the other hand, as suggested by one of the reviewers, the effects of biomass burning may not be discernible as individual transport events but as a large-scale influence of biomass burning in the southern hemisphere's troposphere.

The lowest mixing ratios of ozone are observed in late summer when the STJ is bifurcated over the Pacific ocean giving rise to a branch that crosses the Andes far south of the Cerro Tololo area, typically at 37°S. This is consistent with the hypothesis that the main source of ozone arriving at Cerro Tololo is O_3-rich air that enters the troposphere in connection with the tilting of isentropic surfaces at the STJ. In such conditions, the air arriving at Cerro Tololo during summer should spend a longer time than in other seasons over the Pacific Ocean since its intrusion into the upper troposphere. During that time, the O_3-rich air should suffer dilution by mixing and depletion through photolysis and subsequent reaction of electronically excited oxygen atoms with water molecules (Crutzen, 1995 and references therein). Additionally, as discussed in Subsection 3.2, two other factors may explain the relatively low O_3 values observed in summer. One contributing factor is the entrainment of boundary layer air, poorer in ozone, into the Cerro Tololo area due to the enhanced radiation driven mountain-flow in daytime. In fact, the diurnal cycle of the westerly component of the surface wind derived from the reanalysis data shows a wind speed maximum around mid-afternoon and a minimum during night hours in summer. Moreover, a downward wind seems to develop between 0300 and 0900 UTC. In winter, a similar pattern appears, however no downward wind develops. Another factor, could be photochemical destruction in conditions of relatively low levels of reactive nitrogen oxides (NO_x) linked to transport of air masses which are poor in NO_x as they probably spend long periods over the ocean without sources of NO_x.

In summary, the late summer and fall minimum in ozone measured at Cerro Tololo seems to be mainly associated with the fact that the STJ is located far from the measuring site during this time of the year. Other reasons could be linked to the appearance of a mountain wind flow in summer, bringing in ozone-poor air from the lower atmospheric levels in which ozone was destroyed by photochemical processes.

It is worth noting that the seasonal behavior of ozone observed at other monitoring stations in the Southern Hemisphere, i.e., Samoa (14°S, 171°W, 82 m), Cape Point (34°S, 18°E, 75 m), Cape Grim (41°S, 145°E, 94 m), Syowa (69°S, 40°E, 21 m) and South Pole (90°S, 2835 m) (Oltsman and Levy, 1994), is different from that observed at Tololo. The former stations show maximum O_3 mixing ratios in winter months (JJA) whereas Tololo does in spring (SON). The summer minimum in O_3 mixing ratios is similar at all stations in the southern hemisphere. The different seasonality in ozone at Tololo as compared to other stations in the southern hemisphere may be partly due to differences in altitude of the stations. However, it cannot be ruled out that there are concurrent processes that may explain the observations at Tololo and elsewhere in the southern hemisphere.

It is interesting to note that a larger variance of the O_3-data was measured during 1997 (under El Niño conditions) than during 1996 (under La Niña conditions) (Figs. 3, 4). Also, the spring maximum in ozone is lower in 1997 than in 1996. These features are consistent with the circulation changes associated with positive and negative phases of the Southern Oscillation. In fact, the enhanced variance can be explained in terms of enhanced frontal activity in the area during the 1997 late winter and spring and the lower maximum in terms of a weaker Pacific high (Fawcett, 1998; Walland, 1998).

3.2. *Diurnal variation*

Fig. 4 shows the diurnal variation of the ozone measurements at Cerro Tololo for seven seasons from June 1996 to February 1998. Spring and, in particular, summer months show a clear diurnal variation, with maximum values at night and minimum values in early afternoon. Fall shows a less marked diurnal variation whereas winter months do not.

During spring and summer, the radiation driven circulation is stronger than in fall and winter. Therefore, it is likely that ozone values drop during the spring and summer days as up-slope winds transport O_3-poor air from lower atmospheric levels to the measuring site.

The mountain flow circulation indicated above appears not to be able to produce a significant entrainment of air from atmospheric layers below the inversion layer. If that was the case, we should expect to find an enhanced anticorrelation between O_3 and water vapor pressure measured at Cerro Tololo in summer months (DJF) as compared to winter months (JJA) due to a stronger radiation driven circulation in DJF than in JJA. We find more humid air in summer months but no significantly enhanced anticorrelation. This is illustrated in Fig. 5. Therefore, it appears that the Cerro Tololo site is generally representative of

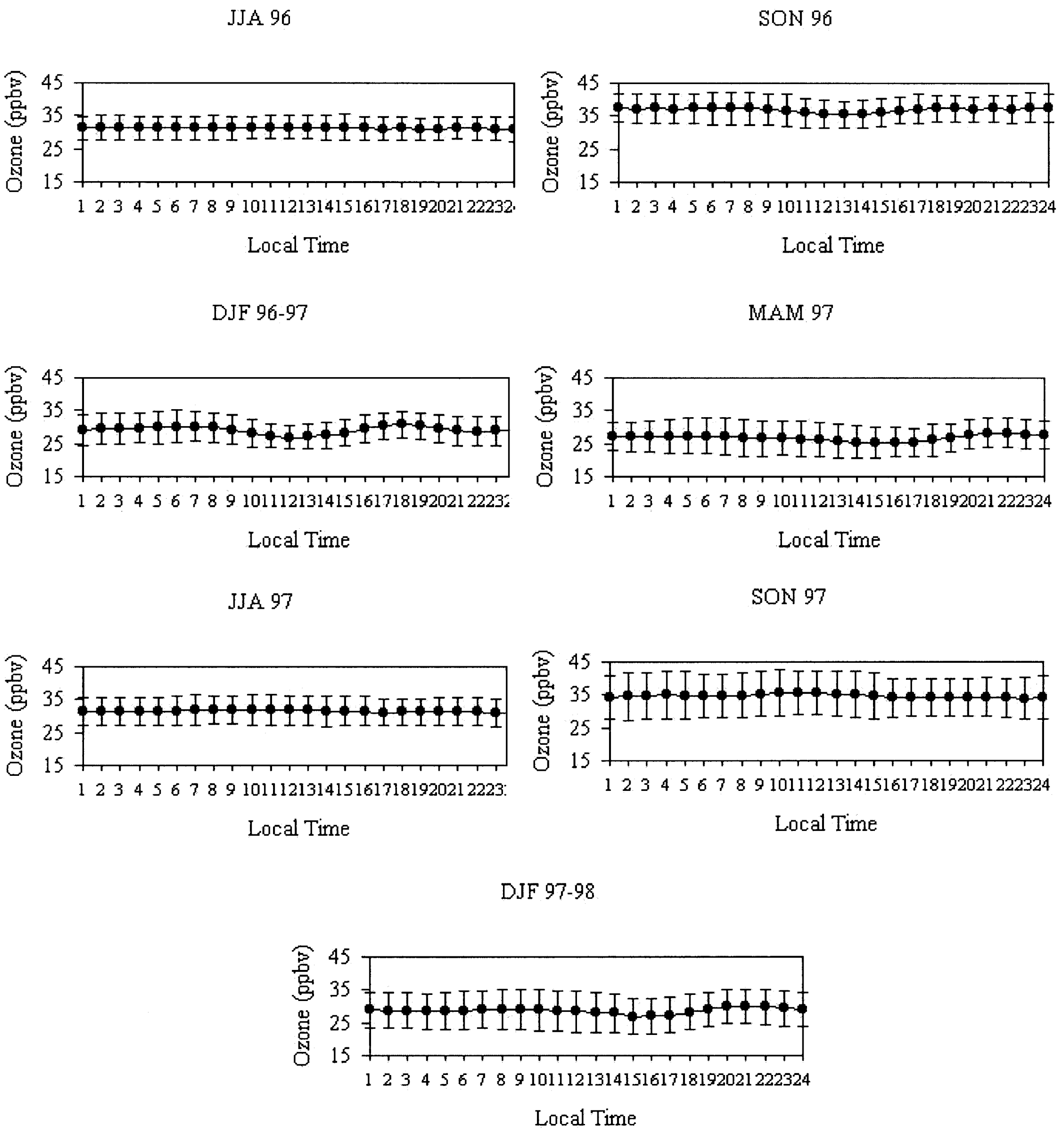

Fig. 4. Diurnal variation of ozone volume mixing ratios at Tololo for 7 seasons: June through August 1996 (JJA 96), September through November 1996 (SON 96), December 1996 through February 1997 (DJF 96–97), March through May 1997 (MAM 97), June through August 1997 (JJA 97), September through November 1997 and December 1997 through February 1998 (DJF 97–98). Error bars indicate one standard deviation of the mean.

the free tropospheric air above the subtropical inversion layer.

It is worth noting that in winter, in connection with occasional passage of frontal systems through the area, significant mixing of boundary layer air appears to occur. Ozone mixing ratios drop as relative and absolute humidity values go up. Such a situation from June 1997 is illustrated in Fig. 6. In this period, from 7 to 9 June, a coastal trough developed along the northern and central part of Chile, followed by a front that crossed the area on 12 June. In a situation like this, the coastal trough is restricted to the boundary layer. When its southern end is located to the south of Quintero

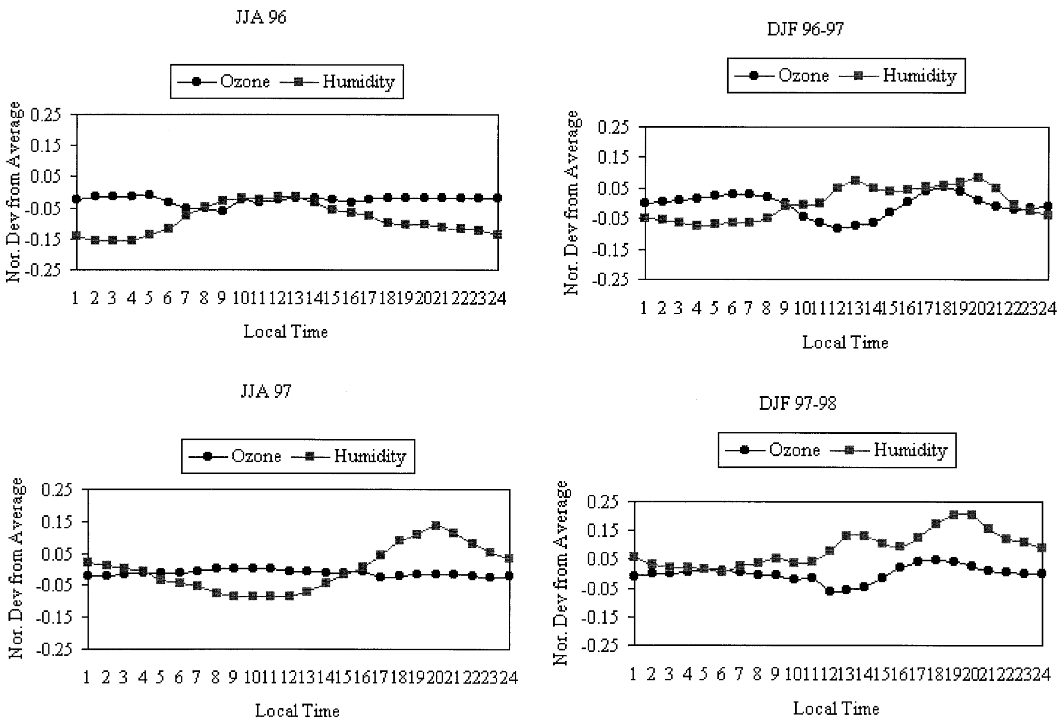

Fig. 5. Diurnal variation of the normalized deviation from average of ozone and water vapor pressure (humidity) at Tololo for four seasons: June through August 1996 (JJA 96), December 1996 through February 1997 (DJF 96–97), June through August 1997 (JJA 97) and December 1997 through February 1998 (DJF 97–98). The normalizing factor is the diurnal average for each variable.

station, the inversion layer goes up, allowing entrainment of maritime air inland (Rutllant and Garreaud, 1995). The weakening of the subsidence from 1200 UTC 8 June concurs with the water vapor pressure increase and the initial decrease of tropospheric ozone (Fig. 6). The relatively high level of water vapor and the increment of ozone coincided with the approach and arrival of the frontal system.

Another explanation for the low O_3-values and the afternoon O_3-minimum in summer at Cerro Tololo (cf. Figs. 3, 4) is a photochemical sink. In fact, it is well established that in absence of sufficient amounts of nitric oxide (NO) photochemical destruction of ozone takes place (Crutzen, 1995 and references therein). This could be the case at Tololo because the air arriving at this site is most likely poor in reactive nitrogen (NO_x) as it probably spends a long period over the ocean without sources of NO_x. No actual measurements of nitrogen oxides or other species are yet available at Cerro Tololo to test this hypothesis. However, a photochemical sink like this should not act on the local scale but on the synoptical scale as the time-scales for ozone production and destruction are of the order of a week or more. Hence, these photochemical processes should not act solely but in combination with the radiation driven circulation. In any case, a better understanding of these processes requires measurements of several chemical species at and upstream from Tololo.

Night-time values of ozone should be representative for free-tropospheric air whereas daytime values may, to some extent, be influenced by boundary layer conditions (i.e. below the subtropical inversion layer), particularly in summer. Also, in winter in connection with frontal passages similar conditions to those in summer may occur.

When mixing of boundary layer air into the Cerro Tololo site takes place, some impact of anthropogenic sources from La Serena and

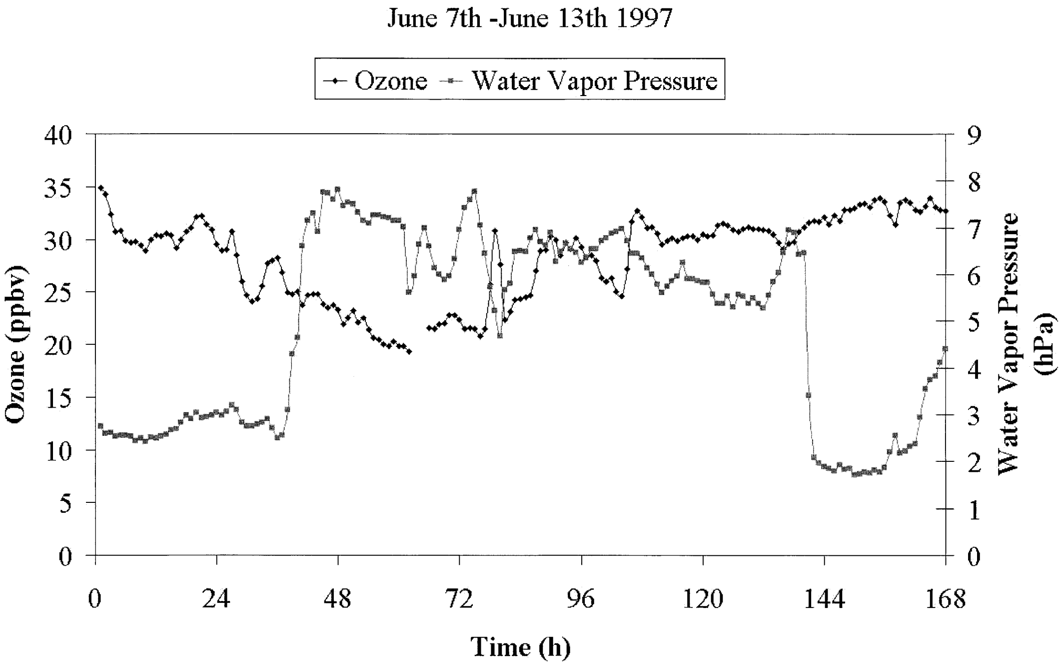

Fig. 6. Hourly averages of ozone (ppbv) and water vapor pressure (hPa) measured at Tololo during 7 June through 13 June 1997. The x-axis corresponds to hours since midnight between 6 June and 7 June. The water vapor pressure has been derived from temperature and relative humidity measurements at Tololo using a standard empirical formula.

surroundings may be detected at Cerro Tololo. However, occasions of such mixing from air below the subsidence inversion do not seem to be frequent. The data show no indication of in situ photochemically produced ozone, as there are no maximum mixing ratios at the hours when the sun is closest to the zenith and solar radiation is strongest. Again, measurements of relevant chemical species (carbon monoxide, nitrogen oxides, etc.) are required.

Oltmans and Levy (1992, 1994) have reported similar seasonal and diurnal variations of ozone as the ones measured at Cerro Tololo for a subtropical station in the Northern Hemisphere, namely at Bermuda (32°N, 64°W, 40 m.a.s.l). At both stations, there is a summer minimum and a spring maximum in ozone, however the amplitude of the seasonal and diurnal changes is stronger at Bermuda than at Cerro Tololo. The reasons for that are linked to differences in altitude and circulation patterns between the stations and also to the magnitude of the anthropogenic perturbation of the background composition of the atmosphere.

4. Conclusions and perspectives

A first analysis of the ozone data collected since April 1996 at the WMO station located at Cerro Tololo (30°S, 70°W, 2200 m.a.s.l.) has been presented. Meteorological features of the Tololo site have been described.

The Cerro Tololo area is mostly immersed in the free atmosphere and affected by the subsidence regime of the South Pacific high that brings clear sky condition for most of the year. Occasionally in wintertime, these conditions are interrupted by passing fronts or by cut-off lows. During winter and spring the STJ crosses the Andes around 30°S, right above Cerro Tololo and occasionally even the polar jet (PJ) extends northward to 30°S. In summer and fall the STJ shows a bifurcation over the Pacific Ocean with one branch north of Cerro Tololo around 23°S and another to the south around 37°S, when the PJ is located much further south of the Tololo site at around 50°S.

The ozone mixing ratios measured at Tololo show a distinct maximum during spring and a minimum during the late summer and fall (Fig. 3).

This feature appears to be associated with the strength of the subtropical subsidence and especially, the location of the STJ. Apparently, the main source of ozone arriving at Cerro Tololo is O_3-rich air that enters the troposphere along tilting isentropic surfaces near the STJ.

The time series of ozone data at Cerro Tololo is too short to allow establishing interannual variability patterns. Nevertheless, during the strong El Niño event of 1997 resulted in a significantly higher variance in the data and a lowering of the spring maximum.

Spring and particularly summer months show a clear diurnal variation pattern, with maximum values at night and minimum values in early afternoon. Fall months show a less marked diurnal variation whereas winter months do not show any. This diurnal variability is probably driven by the mountain wind flow that appears in late spring and summer.

Night-time values of ozone should be representative for free-tropospheric air, whereas daytime values may to some extent be influenced by boundary layer air transported up-slope to the East. This can occur particularly in summer in connection with a mountain flow or in winter in connection with frontal activity. However, this influence appears not to be significant.

Future work should consider trajectory analyses in order to establish the origin of the air arriving at Cerro Tololo and to quantify the transport of O_3-rich air from the upper troposphere to the measuring site. In addition, some meso-scale modeling work for the Cerro Tololo area is already ongoing (Fiebig, personal communication). Also, measurements of other species such as carbon monoxide (CO), reactive nitrogen (NO_x), etc., would be helpful in characterizing the air masses arriving at Tololo and in quantifying the impact of anthropogenic sources in area.

In summary, we find the chemical composition of the air arriving at the Cerro Tololo site to be generally representative of background conditions of free-tropospheric air in the subtropics of the southern hemisphere. Also, as the intrusion of O_3-rich air in connection with the tilting of isentropic surfaces near the SJT appears to be the main source of ozone measured at Cerro Tololo, the data collected at this site may provide an opportunity to better understand stratosphere-troposphere exchange processes in the subtropics. If the current QHAWAYRA program will prove to be successful, we expect the Cerro Tololo site to become a fully equipped station, which will significantly contribute to a better understanding of global and regional atmospheric chemistry and climate change studies.

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