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Tropospheric ozone climatology at two southern subtropical sites, (Reunion Island and Irene, South Africa) from ozone sondes, LIDAR, aircraft and in situ measurements

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Abstract

This paper presents a climatology and trends of tropospheric ozone in the southwestern part of Indian Ocean (Reunion Island) and South Africa (Irene and Johannesburg). This study is based on a multi-instrumental dataset: PTU-O_3 radiosoundings, DIAL LIDAR, MOZAIC airborne instrumentation and Dasibi UV ground based measurements.

The seasonal profiles of tropospheric ozone at Reunion Island have been calculated from two different data sets: radiosondes and LIDAR. The two climatological profiles are similar, except in austral summer when smaller values for the LIDAR profiles in the free troposphere, and in the upper troposphere for all seasons occur. These results show that the LIDAR profiles are at times not representative of the true ozone climatological value as measurements can be taken only under clear sky conditions, and the upper limit reached depends on the signal.

In the lower troposphere, climatological ozone values from radiosondes have been compared to a one year campaign of ground based measurements from a Dasibi instrument located at high altitude site (2150 m) at Reunion Island. The seasonal cycle is comparable for the two datasets, with Dasibi UV values displaying slightly higher values. This suggests that if local dynamical and possibly physico-chemical effects may influence the ozone level, the seasonal cycle can be followed with ground level measurements. Average ground level concentrations measured on the summits of the island seem to be representative of the lower free troposphere ozone concentration at the same altitude (∼2000 m) whereas night time data would be representative of tropospheric concentration at a higher altitude (∼3000 m) due to the subsidence effect.

Finally, linear trends have been calculated from radiosondes data at Reunion and Irene. Considering the whole tropospheric column, the trend is slightly positive for Reunion, and more clearly positive for Irene. Trend calculations have also been made separating the troposphere into three layers, and separating the dataset into seasons. Results shows that the positive trend for Irene is governed by the lower layer most probably by industrial pollution and biomass burning. On the contrary, for Reunion

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Island, the strongest trends are observed in the upper troposphere, and in winter when stratospheric-tropospheric exchange is more frequently expected.

1 Introduction

Tropospheric ozone has a significant climatic impact especially in the middle and upper troposphere, where ozone acts as a greenhouse gas. In the lower troposphere, ozone is known to be a major oxidant and is involved in other oxidant production, such as OH. Model calculations show that ozone formation can be enhanced with deep convective transport of boundary layer precursors (Thompson et al., 1997). Knowledge of the climatological characteristics of tropospheric ozone production and fluxes in subtropical regions would allow a better understanding of its direct effect on climate, and its role in atmospheric photochemistry. Subtropical regions, although poorly studied, experience at different times of the year significant stratospheric-tropospheric exchanges (STE), and also intense photochemical ozone production due to biomass burning, especially in our region of interest which covers the southern parts of Africa and the Indian Ocean.

Data used in this paper come from long term series of radiosonde data from Reunion Island and Irene in South Africa, both of which belong to the SHADOZ network (Thompson et al, 2003), and allow a comparative study of the ozone sources climatology in the subtropics. Reunion island (20.8° S; 55.5° E) is located near the southern limit of the tropics, in the Indian Ocean, 1000 km east of Madagascar. During winter, the island is affected by the proximity of the subtropical jet stream which may have a role in stratospheric air masses intrusions via tropopause breaks (Baray et al., 1998) and Rossby wave breaking (Postel et al., 1999). Results describing stratospheric intrusion associated with a tropical cut off low in South Africa have been previously published (Baray et al., 2003). A study of the effect of tropical convection on tropospheric ozone has also been reported (Leclair de Bellevue et al., 2006) and showed some differences between the two sites, Reunion and Irene. Biomass burning activity in southern

Africa and Madagascar during austral spring[1] associated with long range transport of air masses are also a cause of the tropospheric ozone increase over Reunion Island (Taupin et al., 2002).

Johannesburg (26.1° S; 28.0° E) and Irene (25.5° S; 28.1° E) are located further south and about 3000 km west of Reunion. These sites are located closer to photochemical sources, but from a dynamical perspective, the site of Irene is less affected by tropical convection but more affected by the subtropical jet stream activity than Reunion.

In this region, the ozone sources have been identified as both photochemical (Baldy et al., 1996) and stratospheric (Baray et al., 2003). Thompson et al. (2003) derived a Tropical Ozone climatology from the SHADOZ network data only and Diab et al. (2004) focused their climatology study on Irene and Johannesburg sites. A climatology of stratospheric intrusions has been conducted over the Pacific and Atlantic Oceans (Waugh, 2000). However, the ozone climatology and the influence of photochemical and stratospheric sources have not been analyzed using the same climatological approach at two distant subtropical sites ; and as such this is the objective of the present study.

A wide range of ozone data is available. In addition to ozone sonde profiles, ground based data from a UV absorption analyzer, in-situ measurements from commercial aircraft (MOZAIC data) and vertical ozone profiles from LIDAR will be used. In this paper the geophysical context for each site and the regional sources of ozone precursors will be presented together with a brief description of the database features. The first step is to examine and compare all databases in order to ensure of the coherence of the various data sets. The influence of data sampling and bias on ozone values between LIDAR and radiosonde data in Reunion is discussed. All data sets will be used to derive climatological properties of ozone in the subtropics. A comparative study between ground based ozone measurements at an elevated site (2100 m) on Reunion Island and low altitude radiosonde data focus on the seasonal cycle of ozone in the lower

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[1] September, October, November.

free troposphere. Finally, the last part of this paper deals with the tropospheric ozone trends over the past decade at Reunion and Irene.

2 Geophysical context and regional sources of ozone precursors

2.1 Reunion Island

2.1.1 Dynamical context

Reunion is located in the subsidence area of the Southern Hemisphere Hadley cell. The island meteorology is subject to subtropical, tropical and temperate influences on the general circulation, namely the south Indian anticyclone, the subtropical jet stream, and perturbations carried in the westerlies (Randriambelo et al., 2003). Preston-Whyte and Tyson (1988) have documented the circulation patterns in South Africa and in the adjacent Atlantic and Indian Oceans.

2.1.2 Regional sources

Of the two types of ozone sources already described, the photochemical source of ozone influencing the ozone profile over Reunion is mainly due to the long range transport of pollutants and ozone precursors from biomass burning activity in southern Africa and Madagascar (Fig. 1). Southern Africa is one of the main biomass burning regions in the Southern Hemisphere. The burning season starts in July and ends in October (Marenco et al., 1990). Measurement campaigns such as TROPOZ (TROPospheric Ozone) (Marenco et al., 1990), SAFARI (Southern African Fire-Atmosphere Research Initiative), TRACE-A (Transport and Atmospheric Chemistry near the Equator-Atlantic) (Andreae et al., 1996), SAFARI 2000 (Swap et al., 2003) have been undertaken in order to study the atmospheric photochemistry and the circulation patterns leading to the redistribution of southern African emissions and resulting in an ozone increase over southern Africa and the Atlantic and Indian Oceans (Randriambelo et al., 1999).

Aghedo et al. (2007) recently determined that 70% of the tropospheric ozone produced by African emissions is exported outside the continent. Meteorological analysis based on trajectory statistics showed that a large part of the African emissions might be advected to the Indian Ocean (Garstang et al., 1996; Piketh et al., 2002). Mechanisms responsible for regional contamination by biomass burning by-product have been documented (Baldy et al., 1996; Taupin et al., 1999, 2002). A climatology based on radiosonde and satellite data focuses on the relative importance of the different mechanisms (Randriambelo et al., 1999).

Ozone increase due to stratospheric intrusions have been detected in South Africa and over the Indian Ocean. These intrusions take place in association with meteorological disturbances such as tropopause breaks induced by the subtropical jet stream, cut off lows, tropical cyclones (Baray et al., 1998, 1999, 2003), westerly waves and frontal zones.

2.2 Irene

2.2.1 Dynamical context

The atmospheric circulation at Irene (25.9° S, 28.22° E) is dominated by the subtropical anticyclone within which subsidence and recirculations cause the accumulation of pollutants on large temporal and spatial scales (Garstang et al., 1996; Tyson et al., 1996; Piketh et al., 2002). As a consequence a 5km deep haze layer is formed over the southern African subcontinent. This layer is capped by a stable and persistent layer due to subsidence resulting from circulations inside the subtropical anticyclone (Cosin and Tyson, 1996). The frequency of this anticyclonic circulation is higher during winter (79% in June, July) than during summer (11% in December) (Tyson and Preston-Whyte, 2000). The system is disrupted when a mid-latitude westerly wave crosses the south of the African subcontinent. The 5 km stable polluted layer is then spread out. Trace gases including ozone precursors and aerosols circulate inside the stable layer until they are finally released eastward as a giant plume centred at 31° S along the east coast (Tyson et al., 1996).

2.2.2 Regional sources

In addition to stratospheric sources, regional sources of ozone in Irene include: biomass burning, biogenic, lightning and anthropogenic emissions (Aghedo et al., 2007).

Irene is located southwards of the main biomass burning region in Africa (Fig. 1). The biomass burning area extends between 10° N and 20° S and eastward (~25° E) during the dry season from June to October. Although the most important ozone increase is expected north of Irene, the impact of biomass burning can be detected at the station as a result of long range transport in the anticyclonic gyre. Results of the SAFARI 2000 campaign emphasize the importance of biomass burning in the tropospheric ozone budget of the southern African subcontinent (Swap et al., 2003). Volatile Organic Compounds (VOCs) are naturally emitted by vegetation (Kesselmeier and Staudt, 1999). The emission of VOCs significantly influence atmospheric chemistry because of their high reactivity and result in the formation (or destruction) of tropospheric ozone in high (or low) NO_x conditions (Aghedo et al., 2007).

During summer, the tropical easterlies from the Indian Ocean cause moist air to be advected over Irene. Strong convective activity is a daily phenomenon and can promote a rapid vertical uplift of surface pollutants, which leads to an ozone enhancement in the mid- to upper troposphere (Pickering et al., 1990, 1996). Lightening, which often accompanies these convective storms, can also be responsible for an increase in tropospheric ozone load.

Anthropogenic sources are an important factor in the ozone budget. Irene is located close to two urban-industrial areas: Pretoria and Johannesburg. The station is also 100 km from the main power generating area of South Africa, with 11 large coal generating power plants (Diab et al., 2004). Domestic use of biofuels also contributes to ozone precursors. Emissions due to power plants and domestic fuels are stronger during winter because of heating requirements. An emission increase during summer can also be expected because of air conditioning.

3 Instrumentation and database features

3.1 Radiosondes

A radiosonde program started in 1992 at Reunion and the database represents 15 years of usable data. At Irene, radiosondes were regularly launched between July 1990 and October 1993 (Diab et al., 2004). Since the two sites have been involved in the SHADOZ program in 1998, radiosondes are launched regularly on a weekly basis.

The SHADOZ program aims to provide a consistent database for tropospheric ozone in the Southern Hemisphere. The program originally intended to supplement the sparse amount of tropospheric ozone data in the Southern Hemisphere compared to the abundant data in the Northern Hemisphere (Thompson et al., 2003). The SHADOZ network involves 15 stations, distributed so as to have a good zonal coverage of the Southern Hemisphere.

Irene and Reunion stations use ECC (Electro Chemical Cells) sondes for ozone and Vaisala RS80 sondes for temperature, pressure and humidity. The KI Solution for cathode is 1% buffered at Irene and 0.5% buffered at Reunion. Profiles give data from ground level (24 m at Reunion, 1524 m at Irene) up to burst altitude, which is located between 30 km and 35 km in most cases. Ozone content precision for ECC sondes is estimated at 5% for the stratosphere and 10% for the troposphere (Thompson et al., 2003). Since 2007, horizontal wind measurements have been made at Reunion by GPS, simultaneously with ozone and PTU measurements.

3.2 LIDAR

Two LIDAR (Light Detection And Ranging) instruments are operating at Reunion. One of them, devoted to the measurement of a tropospheric ozone profiles, has been operating since 1998. The approach is to process the differential absorption of two UV wavelengths (289 nm and 316 nm) generated by a high pressure deuterium cell. LIDAR profiles are taken several times a week with a 150 m vertical resolution between 3 km and the tropopause height of 17 km in the tropics. The limits of the vertical domain for each profile are closely related to the atmospheric conditions. Details on the DIAL ozone LIDAR at Reunion are given in Baray et al. (1999) and on all the instrumentation of OPAR (Observatoire de Physique de l'Atmosphère de la Reunion) in Baray et al. (2006).

3.3 MOZAIC

The MOZAIC program (Measurement of Ozone and Water Vapour by Airbus In-Service Aircraft) was launched in 1994 by European scientists and Airbus in order to collect ozone water vapour, carbon monoxide and nitrogen oxides data to validate global chemistry transport models (Marenco et al., 1998). The precision of the ozone measurements is ±2 ppbv±2% (Thouret et al., 2006). The program uses equipment installed aboard long haul Airbus A340 aircraft flying from Europe. The MOZAIC data used in this paper corresponds to 577 flights from and to Johannesburg between July 1995 and January 2003. The seasonal distribution is given in Fig. 2.

3.4 Temporal coverage

Vertical ozone profile data is available since the early 1990s at Reunion and Irene. Both sites joined the SHADOZ network in 1998. In this article, the climatological study (Figs. 3, 4) focuses on the 8 common years of data (1998–2006, Table 1) for the two sites. Hence the ozone climatology benefits from a greater regularity in the radiosoundings time distribution regarding a considerable number of profiles (Table 1). The trend section uses the whole dataset for the two sites in order to view the widest time range.

At Reunion, profiles are collected regularly each week and the frequency of soundings is less affected by weather considerations than by possible technical issues. For each site, the total number of profiles over the 8-year period ranges between 20 and 25 profiles per month. Differences between January to July and August to December in numbers of radiosonde profiles for Reunion (Fig. 2) can be explained by measurement campaigns that have been conducted for satellite validation purposes or to describe particular atmospheric events in July 1998 and 1999.

MOZAIC and LIDAR databases show a great number of profiles, respectively 577 and 265 profiles respectively (Fig. 2, Table 1) but the profiles are sporadically distributed in time compared to the SHADOZ data. In addition to their climatological value, MOZAIC and LIDAR data are interesting to describe singular atmospheric events. Since the vertical domain of these data is variable, more work is necessary to define precisely the most convenient vertical domain for a climatology. MOZAIC data were recently used to derive an ozone climatology in the upper troposphere (9–12 km) (Thouret et al., 2006). In the next section, seasonal profiles enable a comparison between LIDAR data and ozone probe data. The climatological significance of LIDAR data will be discussed.

4 Data analysis

4.1 Comparison of ozone DIAL and sonde climatological profiles at Reunion

LIDAR seasonal profiles show lower ozone content than radiosounding profiles above 11 km for spring and winter, above 13 km for autumn and summer. This difference which appears on climatology is not visible on simultaneous measurements (Baray et al., 1999). Because of the absorption of the LIDAR beam by ozone molecules, when the LIDAR signal is weakened in the upper part of the profiles, the vertical limits of LIDAR ozone profiles are often set below an altitude where high ozone content is found, then the number of profiles used to build the climatological profiles lower in the upper part of the climatological profiles, and profiles with high ozone content in the upper troposphere are small in number in the LIDAR database. Contrary to the LIDAR technique, the measurement of ozone by radiosonde is not affected by the amount of ozone. This gives rise to some differences in the upper tropospheric climatology. The upper tropospheric DIAL ozone climatology is not representative of the real ozone climatology, despite individual DIAL profiles which covers the whole troposphere are available.

In summer, seasonal profiles are concordant in the lower troposphere below 5 km. Above 13 km, the trend is similar to the other seasons i.e. the LIDAR profile depicts smaller values than the sounding profile. Between 5 km and 13 km, the LIDAR profile shows higher values than the sounding profile. The maximum difference between the two profiles in this altitude range is 12.83 ppbv at 9.5 km. This difference is comparable to the difference between the two ozone climatological profile obtained in distinguishing the radiosonde database in presence of tropical convection and not (Fig. 4 of Leclair de Bellevue et al., 2006).

While radiosonde measurements can be launched in any weather conditions, LIDAR profiles are constrained to meteorological conditions: LIDAR measurements can be made only during nights with clear sky conditions. Then, summer corresponding to the rain season at Reunion and taken into account the complex role of convection on ozone, (uplift of ozone poor air masses into the troposphere in convective systems giving low values of ozone in the upper troposphere, and stratospheric-tropospheric exchange in the periphery of convective systems giving high values of ozone in the middle troposphere (Leclair de Bellevue et al., 2006), this gives some differences between ozone DIAL and radiosonde climatologies during summer.

4.2 Monthly mean distribution

The whole database has been processed so as to obtain the mean monthly evolution of ozone content in the troposphere. Figure 4 depicts the mean annual cycle in ozone content in ppbv (0–130 ppbv) between 3 and 16 km. Panels (a) to (d) depict LIDAR data, SHADOZ data from Reunion, MOZAIC data from Johannesburg, and SHADOZ data from Irene respectively. The lower altitude limit of 3 km is imposed by the LIDAR data; and 16 km is below the tropopause height (around 17 km in the tropics). The maximum value of the ozone mixing ratio is set 130 ppbv in order to keep variations of the SHADOZ data perceptible. This scale allows variations in the ozone content in the mid and lower troposphere to be distinguished.

4.2.1 Observations and analysis

Although the database includes many LIDAR and MOZAIC profiles (Table 1), Fig. 4b and d derived from SHADOZ data have a smoother appearance most likely due to the greater regularity of radiosonde launches. Each data type show a springtime maximum linked to biomass burning activities, that occurs between September and November according to the site. The greatest values are found in the SHADOZ data (Panels b, d), and Irene data shows values greater than 90 ppbv above 10 km altitude. MOZAIC and LIDAR data also exhibit a springtime peak but the values are seldom above 80 ppbv in the lower and mid-troposphere. The LIDAR ozone content at Reunion (Panel a) shows greater values in January than the content derived from SHADOZ data. Specifically LIDAR data display ozone values varying between 80 and 90 ppbv between 9 and 12 km. For the same altitude, the SHADOZ ozone content has values between 50 and 70 ppbv. This result is consistent with summer seasonal profiles shown in Sect. 3.

Upper tropospheric ozone content at Irene is greater than at Reunion. During winter, the ozonopause is lower at Irene. Values greater than 100 ppbv are found above an altitude of 13 km whereas at Reunion, such values are found only above 14 km. Irene is located at a more southerly latitude than Reunion, and consequently experiences a greater influence of the subtropical jet stream during winter than Reunion. Thus a strong influence of stratospheric intrusions during winter in South Africa can be assumed. Moreover, Irene is under the influence of the anticyclonic subtropical gyre which is responsible for large scale subsidence and recirculations of air masses in the lower troposphere above the southern African subcontinent. This regime is capped by a stable layer (Diab et al., 2004) which inhibits vertical mixing of air masses. Tyson and Preston-Whyte (2000) have shown that this system is highly prevalent during winter.

During summer, values above 90 ppbv are found at Irene at 10 km altitude and above.

At Reunion, such values are not recorded below an altitude of 11 km. Irene is located nearer to the photochemical sources than Reunion. Moreover, Pickering et al. (1990, 1996) show that during summer the southern African subcontinent is under the influence of the easterlies, which gives rise to convective activity and allows advection and mixing of tropospheric air masses. The quick redistribution of surface pollutants may be responsible for a mid- and upper tropospheric ozone enhancement. Considering the South African data for January, an ozone enhancement appears in the mid-troposphere where values exceed 60 ppbv. That increase, evident on both the MOZAIC and the SHADOZ plots, might be due to anthropogenic pollution. According to Diab (2004) Irene is located in an active urban industrial region. Domestic emissions during summer coupled with convection might explain this phenomenon.

5 Ground based measurements and comparison with the radiosounding climatology at Reunion

One year of ground-based ozone measurements have been performed at Piton Textor (2150 m ASL, southeast of Reunion) from October 1998 to October 1999. The instrument was a commercial photometer Dasibi 1008 RS allowing continuous measurements with a precision of about 5%. Monthly means of ground based ozone have been calculated, separating night-time (from 10 p.m. to 7 a.m.) and day-time (8 a.m. to 8 p.m.) measurements. These measurements are compared in Figure 5 with average ozone concentrations measured at an altitude of 2100 m by radiosoundings launched at Roland Garros airport (north of Reunion) in the morning. Radiosonde monthly data averaged over the period 1992–2006 are quite comparable to the values measured at ground level by the Dasibi instrument. Radiosonde data for the period 1998–1999 are somewhat lower. It is noted, however, that during this period only 40 balloons launches were performed, therefore monthly means are based on only 3 to 4 days of data. We also observe that ozone values recorded during the night by continuous ground level measurements are slightly greater than those recorded during the day by about 5 ppb. When the radiosondes reach 2000 m altitude they are rather far from the influence of the mountain boundary layer and the data corresponds to the free troposphere ozone concentration at 2100 m altitude. In contrast, ground measurements at the same altitude are strongly influenced by vertical motions and exchanges between the lower layers and the free troposphere, these exchanges being forced by orography. Figure 6 represents average vertical ozone profiles for the period 1998 to 2006 between sea level and 5000 m altitude. All seasonal profiles exhibit a positive gradient of 3 to 7 ppb/km, the largest gradient being observed during austral spring which corresponds to the fire season in southern Africa and Madagascar. Air motions along the mountain slopes are generated by a combination of land/sea breeze and slope breeze due to the differential heating of mountain slopes. This effect can be more or less pronounced depending on the force of the synoptic wind (i.e., the south-east trade wind). During the daytime, sea breezes and upslope breezes may combine to bring ozone-poor air from the boundary layer to the top of the mountain. Turbulent motions then mix this air with ozone rich air from the free troposphere resulting in concentrations similar to the free tropospheric concentration at the same altitude. During the night, land breezes combine with downslope breezes generating a subsiding motion as observed for instance on the slopes of Mauna Loa volcano (Garett, 1980). This leads to a reverse diurnal cycle of ozone, with maximum values recorded during the night, which has been observed at various mountain sites in continental regions (Zaveri et al, 1995). This original ozone cycle with ozone nighttime recovery has already been observed at Reunion, even in the marine boundary layer and was attributed to dynamical effects (Bremaud et al., 1998a). In another paper, Bremaud and Taupin (1998b) also discussed the influence of heterogeneous chemistry within orographic clouds in the marine boundary layer to explain the daytime ozone depletion. However, this is probably a second order effect compared to the vertical mixing between layers presenting important vertical ozone gradients

A seasonal cycle comparable to those observed in the free troposphere is observed with a maximum during Winter-Spring and a minimum in summer-Autumn. This seasonal cycle is observed in both Dasibi and radiosounding data at 2 km altitude, with Dasibi values slightly above radiosounding data. Average ground level concentrations measured on the summits of the island seem to be representative of the lower free tropospheric ozone concentration at the same altitude (~2000 m) whereas nighttime data would be representative of tropospheric concentration at a higher altitude (~3000 m) due to the subsidence effect. Ground level measurements at an altitude station at Reunion Island therefore allow to document seasonal variations of regional free tropospheric ozone concentrations. To better understand local dynamics and its influence on trace gas concentrations, we are currently starting a high resolution modelling program with a meso-scale non-hydrostatic model in order to better understand ground level measurements which will be performed at the future observatory built on the top of the Maïdo Peak in the north-west of the island at 2200 m altitude. This observatory, expected for 2010, will be devoted to long term atmospheric composition measurements in the framework of monitoring networks such as GAW (Global Atmospheric Watch) and NDACC (Network for Atmospheric Composition Changes).

6 Long-term Tropospheric ozone behaviour at Reunion Island and Irene based on linear trend calculations

The ozone databases of Reunion Island and Irene cover the periods 1992–2006 and 1990–2006 respectively (time series of 15 and 17 years). In this section, weexamine the linear trend of tropospheric ozone over these periods, and we compare the different tropospheric layers of the both measurement sites.

6.1 Methodology

To examine the linear trend, the following equation is used:

$$\text{Ozone} = \tau t + \beta \tag{1}$$

τ is the linear trend calculated by linear regression using the least squares method, t is the time and β is a residual term. The error is calculated using the student's t-test with 99% confidence interval. This error gives a statistical estimation of the influence of the variability of ozone data on the estimation of the linear trend (Montgomery et al., 2006). Radiosonde data can be given in concentration (molec/cm^3), partial pressure (nbar) or volume mixing ratio (ppbv). Ozone trends have been calculated in atmospheric layers in ppbv by decade and in DU by decade (1 DU=2.69×10^{16} molec/cm^2), with the two calculations giving similar results. We present only the results in DU/decade in the following section.

6.2 Results

Ozone trends calculated at Reunion and Irene are presented in Fig. 7. The lower limit of the tropospheric column is 1 km at Reunion, and 2 km at Irene, because of the different altitudes of the two sites. The upper limit has been fixed at 16 km, below the tropopause height at the two sites (Sivakumar et al., 2006). Considering the whole column, the tropospheric trend is slightly positive (0.5 DU/decade) for Reunion, and more clearly positive for Irene (1.97 DU/decade). These two values are superior to the statistical error calculated for the two sites (0.33 DU/decade and 0.08 DU/decade for Reunion and Irene respectively). All the trends calculated in DU by decade and the corresponding percentages by decade are presented in the Table 2.

In order to examine the potential influence of the two main sources of ozone in the region, *viz.* anthropogenic and biomass burning pollution of the lower layers and stratospheric–tropospheric exchange, the troposphere has been separated into three layers: one below 4 km, the altitude where the trade wind inversion occurs (Taupin et al., 1999), and the free troposphere which is separated into two equal parts: the lower free troposphere between 4 and 10 km and the upper free troposphere between 10 and 16 km. At Reunion, below 4 km, because of the easterly trade wind regime, ozone is not expected to be directly influenced by biomass burning and stratospheric-tropospheric exchange, but only by local sources and exchanges within the free troposphere. At

Irene, local sources of pollutions and biomass burning can both occur. For the two sites, the two layers 4–10 km and 10–16 km can be directly influenced by biomass burning and stratospheric-tropospheric exchange. In regard to these considerations, some differences between the two sites appear (Fig. 8):

- The trend for the lower layer at Irene is clearly positive (4.2 DU/decade), while at Reunion the trend is slightly negative (–0.3 DU/decade). Under the hypothesis of an increase of biomass burning and pollution in South Africa, a positive trend of ozone over South Africa is consistent because of a persistent synoptic recirculation which occurs over South Africa (Preston-Whyte and Tyson, 1988). On the contrary, lower layers over Reunion are less directly influenced by biomass burning because of the easterly trade wind regime (Baldy et al., 1996).
- The trend for the lower free troposphere at Irene is positive but less than the layer below. Almost no trend is observed for the upper free troposphere. This suggest that the tropospheric trend at Irene is mainly governed by the lower layers of the troposphere, and specifically by the biomass burning and pollution influences. For Reunion, the trend behaviour is opposite: almost no trend for the lower free troposphere and a slightly positive trend for the upper free troposphere.

The reason for this positive trend for the upper layers is an interesting issue and we draw two hypotheses: first, the influence of the increase of biomass burning and pollution. The less important values than Irene could be due to the complex dynamical mechanisms which are necessary for Reunion Island tropospheric ozone to be influenced by biomass burning: injection of pollutants in the free troposphere by convection, mix-then-cook scheme and ozone production during the westward transport (Chatfield and Delany, 1990, Baldy et al., 1996). The second hypothesis is an increase in stratospheric-tropospheric exchange which could be induced by the climate warming.

In order to estimate the influence of the two mechanisms, seasonal trends are presented in Figure 9 for Reunion and Fig. 10 for Irene. A climatological study based on the Lagrangian determination of the origin of ozone is necessary to quantify with precision the respective origin of each mechanism. However, for Reunion Island, it has been established that:

- The influence of biomass burning is at a maximum in September-October-November season (Baldy et al., 1996, Randriambelo et al., 2000).
- The influence of stratospheric-tropospheric exchange induced by the subtropical jet stream is at a maximum in June-July-August, because of the location of the jet stream close to the latitude of Reunion (Baray et al., 1998; Randriambelo et al., 2000).
- The influence of stratospheric-tropospheric exchange associated with tropical convection is at a maximum in December-January-February (Leclair de Bellevue et al., 2006).
- The March-April-May season could be representative of background ozone levels, since the influences of stratospheric-tropospheric exchange and biomass burning are weak.

Taking account of these considerations, we observe effectively no-trend, or a weak trend, for the two sites during the March-April-May season: –0.52±0.58 DU/decade for Reunion and 0.12±0.11 DU/decade for Irene. For Reunion, the most positive trends have been observed in December-January-February (1.87±1.12 DU/decade) and in June-July-August (2.88±1.42 DU/decade). These values suggest that stratospheric-tropospheric exchange is the most important influence as it is the most active ozone source during these seasons. In contrast, for Irene, the trend is strong in September-October-November (2.68±0.23 DU/decade), and also in December-January-February (3.32±0.38 DU/decade) and in June-July-August (3.09±0.25 DU/decade). For Irene, the two mechanisms can play a role in the annual trend observed. An overview of long-term changes in tropospheric ozone has recently been published (Oltmans et al., 2006). This study was based on a global network of observations, with some stations located in the tropical and subtropical regions of the Southern Hemisphere: American Samoa Island, New Zealand, Australia and South Africa. For Cape Grimm (Australia) and Lauder (New Zealand), Oltmans et al. (2006) observe no change in summer, but a significant increase during the late winter and early spring. For Lauder (New Zealand), they show an interesting vertical structure of the ozone trends, positive below 500 hPa, and slightly negative above the 500 hPa level (but not significant), very similar to the vertical structure that we have observed for the ozone trends over Irene. For Cape Point (South Africa) some increase has been observed throughout the year, but the largest increase is again during the late winter and early spring, similar to the trends observed over Irene in our study. For all the sites of the subtropical Southern Hemisphere, Oltmans et al. (2006) point out that the seasonal increase occurs during a time of the year when biomass burning in the Southern Hemisphere is very active. However, trends in this period on a decadal scale have not been reported, nor is there a significant increase in CO (Langerfelds et al., 2003), hence it is not possible to conclude with certainty which are the main factors influencing the ozone trends for these sites.

In summary, this section was a first attempt to determine the long-term evolution of ozone based on 15 and 17 years databases respectively at Reunion Island in the Southern Indian Ocean and at Irene in South Africa. The two sites were compared and the influence of possible mechanisms playing a role on the long-term ozone evolution discussed. Significant differences in the ozone trends have been observed at Reunion and Irene. The increase in tropospheric ozone observed over Irene occurs mainly in the lower layers, similar to observations of Oltmans et al. (2006) over other sites of the Southern Hemisphere. It could be associated with the increase in pollution and biomass burning. Our study outlines some differences between Irene and Reunion Island, which has larger positive trends in the upper troposphere than in the lower troposphere. Some additional studies are necessary to demonstrate the origin of the positive trends in the upper troposphere over Reunion Furthermore, this work has highlighted the need to determine the trend with more precision. In order to do that, more complex methods and trend models must be applied, which could be, for example, based on a multiple regression neuronal network (Lu and Chang, 2005).

7 Conclusions

In this study we have presented a climatology of tropospheric ozone in the south-western part of the Indian Ocean (Reunion Island) and South Africa (Irene, Johannesburg). The main conclusions are the following:

- The comparison of the seasonal profiles of tropospheric ozone of Reunion Island obtained from radiosondes and LIDAR showed that the two climatological profiles are generally in good agreement, except in austral summer with smaller values for the LIDAR profiles in the free troposphere, and in the upper troposphere for all the seasons. Because of the nature of LIDAR data (measurements performed only under clear sky conditions, and an upper limit depending of the signal) the LIDAR profile is not representative of the true ozone climatology in the whole troposphere.
- Climatological ozone values from radiosonde for the lower troposphere have been compared to a one year campaign of ground based measurements from a Dasibi instrument at a high altitude site (2100 m) on Reunion Island. The seasonal cycle is comparable for the two sets of data, with Dasibi values slightly exceeding radiosounding data at the equivalent altitude. This suggests that if local dynamical and possibly physico-chemical effects may influence the ozone level, the seasonal cycle can be followed with ground level measurement at a high altitude site. Average ground level concentrations measured on the summits of the island seem to be representative of the lower free troposphere ozone concentration at the same altitude (∼2000 m) whereas night-time data would be representative of tropospheric concentration at a higher altitude (∼3000 m) due to the subsidence effect.

- Finally, linear trends have been calculated from radiosondes data at Reunion and Irene. Considering the whole tropospheric column, the trend is slightly positive for Reunion, and more clearly positive for Irene. The trend calculations have been repeated, stratifying the troposphere into three columns, and separating the dataset into seasons. Results shows that the positive trend for Irene seems to be governed by the lower layer and consequently by industrial pollution and biomass burning. On the contrary, at Reunion, the strongest trends are observed in the upper troposphere, and in winter when stratospheric-tropospheric exchange peaks.

Our short term intention is to study the temporal and vertical distribution of anomalies in the tropospheric ozone column over Irene and Reunion. In order to discuss the influence of different sources, climatology and trends of tropospheric ozone, as well as CO measurements and the convective transport index from the MOPITT satellite (Deeter et al., 2003) will be analysed. The long term perspective is to provide a complete quantification of all the sources influencing the tropospheric ozone budget in the region using a Lagrangian approach and to compare the results with those obtained from a global chemistry transport model.

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Table 1. Features of the database: period of measurements, total number of ozone profile and altitude range.

	Number of years of data	Total number of profiles	vertical range of data
SHADOZ RUN	8	269	sea level – 30 km
1992–2006 RUN radiosoundings	15	358	sea level – 30 km
SHADOZ IRN	8	208	1524 m – 30 km
1990–2006 IRN radiosoundings	17	363	1524 m – 30 km
LIDAR RUN	8	265	3 km – 16 km
MOZAIC JOB	9	577	1 km – 12 km

Table 2. Trend values calculated at Reunion and Irene, in DU by decade and the correspondence in percentage per decade. The lower limit A is 1 km for Reunion Island and 2 km for Irene.

	Reunion		Irene	
	DU/decade	%/decade	DU/decade	%/decade
A-16 km	0.50±0.33	1.5±1.0	1.97±0.08	5.6±0.3
A-4 km	–0.22±0.02	3.6±0.3	0.87±0.01	13.8±0.1
4–10 km	–0.54±0.09	0.3±0.5	0.67±0.03	4.3±0.2
10–16 km	0.94±0.04	9.0±0.4	0.39±0.02	3.2±0.2

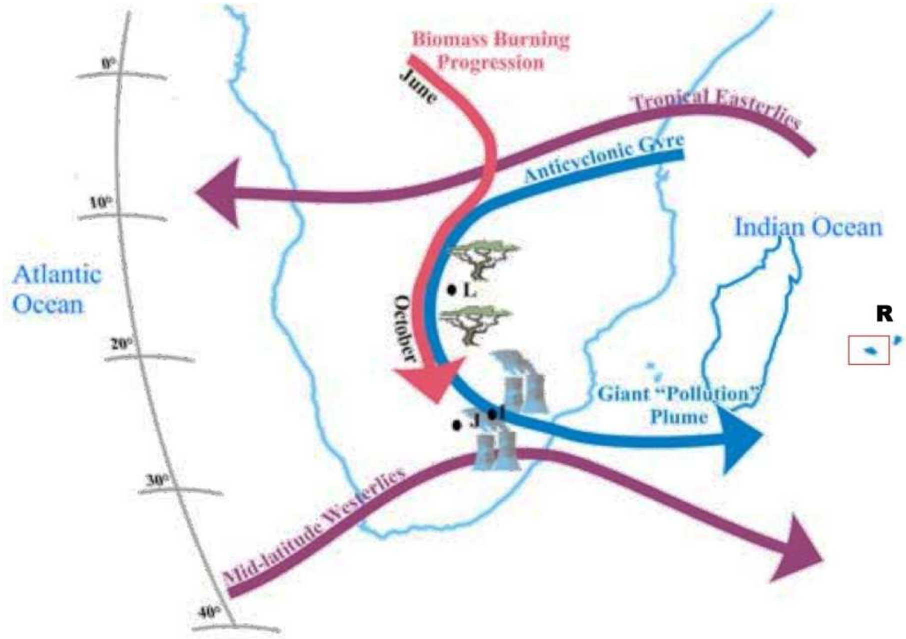

Fig. 1. Schematic representation of meteorological processes affecting tropospheric ozone over subequatorial Africa. Industrial, biomass burning, and biogenic sources of ozone precursors are also shown. The locations of Johannesburg, Irene, Lusaka, and Reunion are represented by "J,","I,", "L" and "R" respectively. Adapted from Fig. 1, Diab et al., 2004.

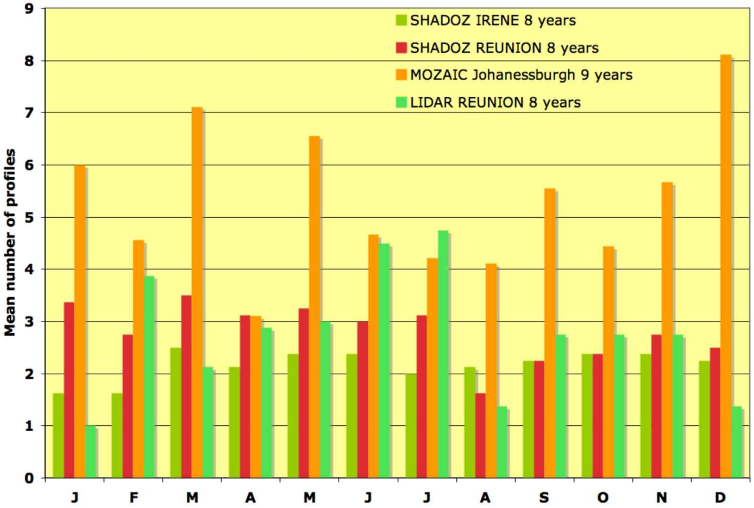

Fig. 2. Monthly distribution of the profiles used for each type of data.

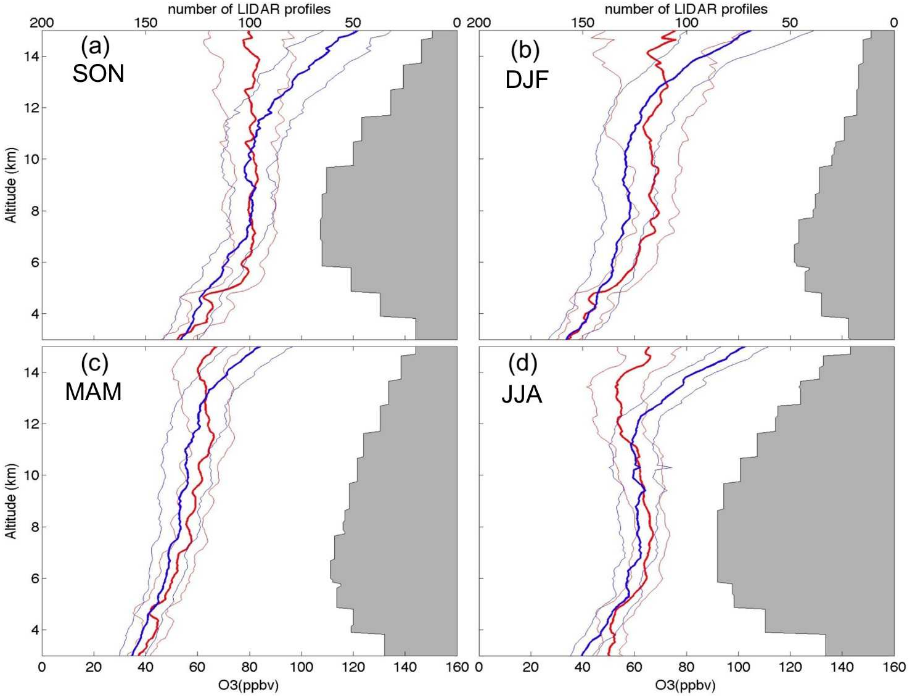

Fig. 3. Seasonal ozone profiles and standard deviation (ppbv) between 3 and 15 km derived from radiosonde data (blue lines) and from LIDAR data (red lines) at Reunion during **(a)** spring (SON) **(b)** summer (DJF) **(c)** autumn (MAM) and **(d)** winter (JJA). The number of profiles used for each DIAL LIDAR climatological profile is given on the right in grey.

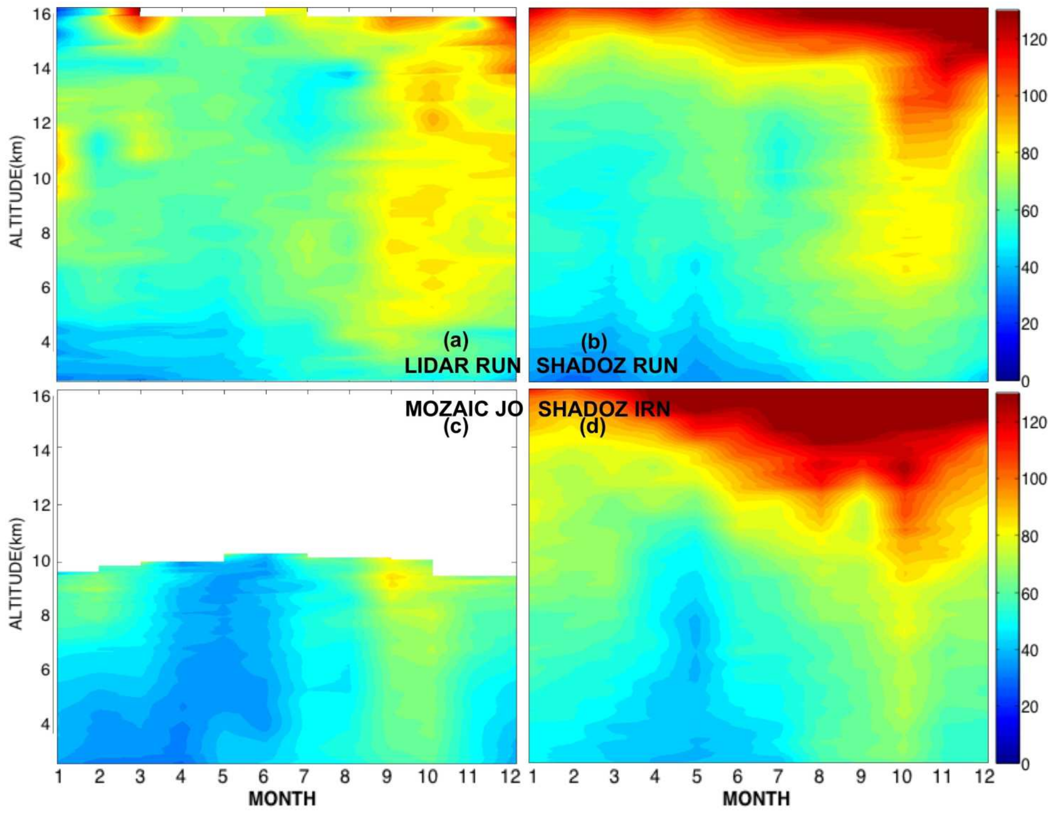

Fig. 4. Monthly distribution of the mean tropospheric ozone content (0–130 ppbv) between 3 and 16 km altitude for different sites and different types of data. **(a)** LIDAR Reunion, **(b)** SHADOZ Reunion, **(c)** MOZAIC Johannesburg, **(d)** SHADOZ Irene.

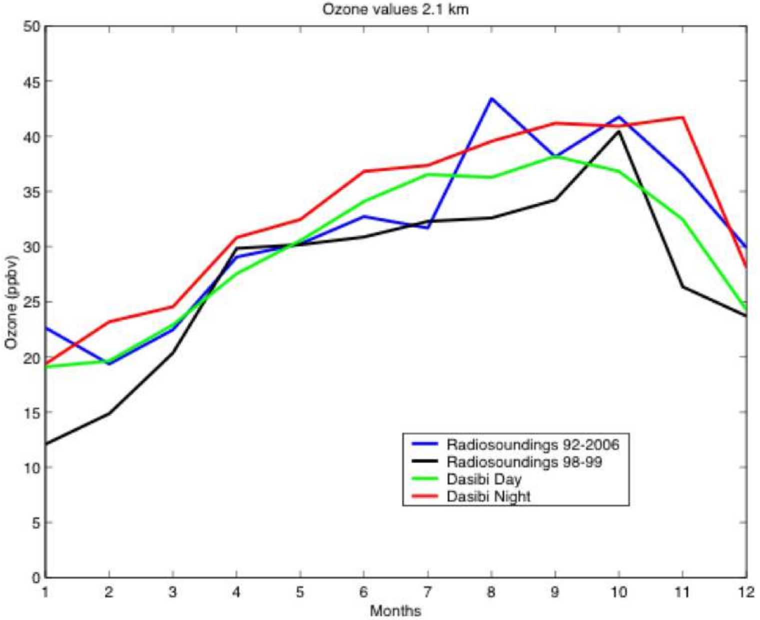

Fig. 5. Seasonal variations of ozone concentration at Piton Textor at 2100 m altitude in the South East of Reunion Island during daytime (green curve) and nigh-time (red curve), from October 1998 to October 1999, and average value of ozone concentration at the same altitude from radiosoundings (average monthly data from 1992 to 2006 (blue curve), and monthly data from October 1998 to October 1999, black curve).

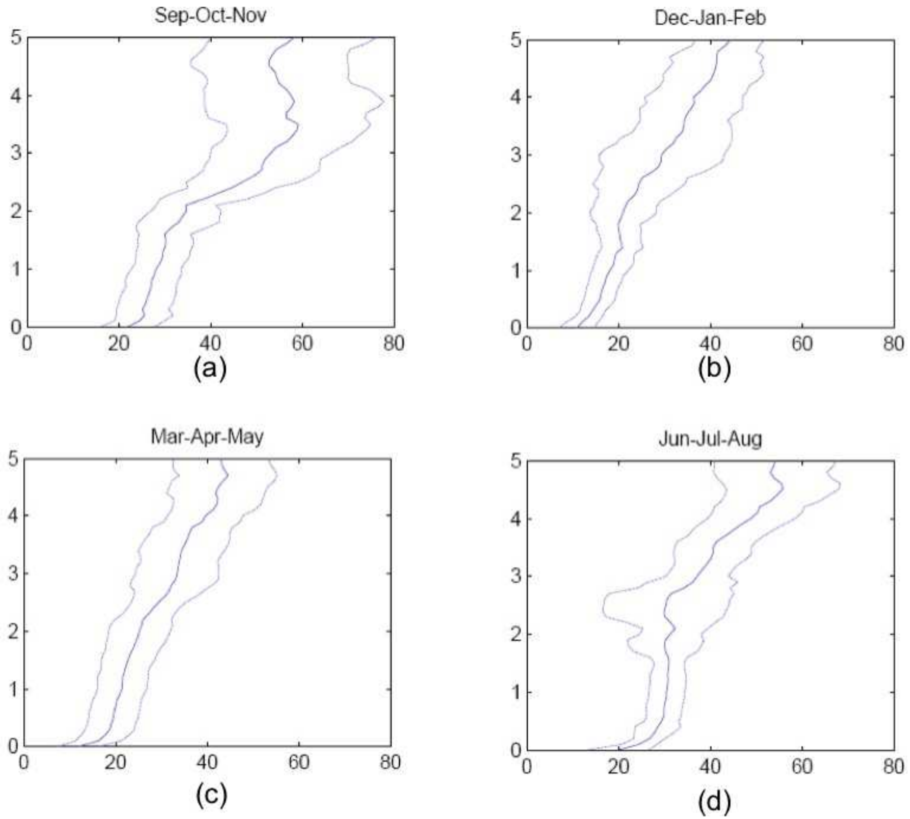

Fig. 6. Seasonal ozone profiles and standard deviation (ppbv) between 0 and 5 km derived from radiosondes at Runion **(a)** spring (SON) **(b)** summer (DJF) **(c)** autumn (MAM) **(d)** winter (JJA).

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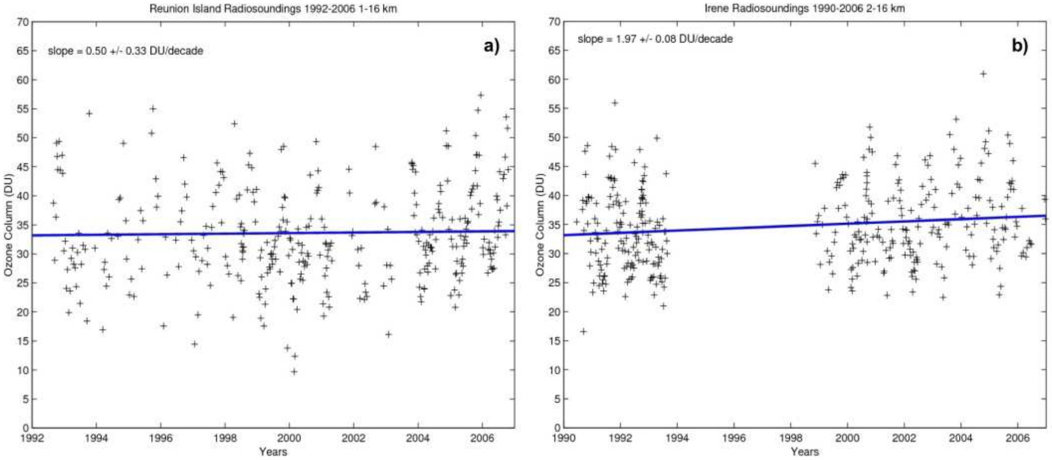

Fig. 7. Ozone trends for the tropospheric layer between 1 and 16 km at Reunion **(a)** and between 2 and 16 km at Irene **(b)**.

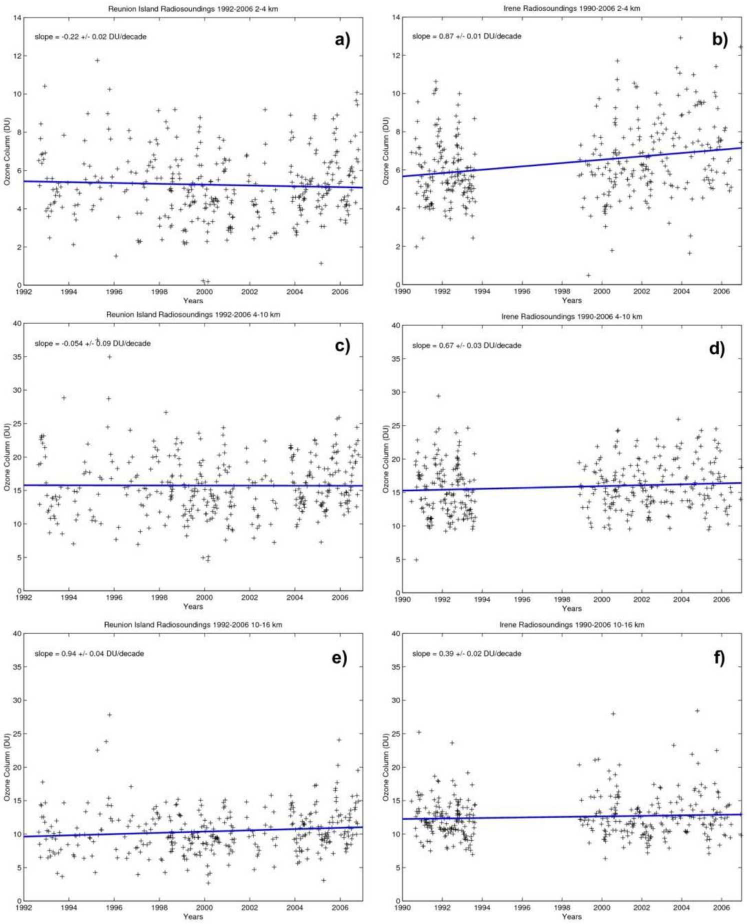

Fig. 8. Ozone trends between 2 and 4 km at Reunion **(a)** and Irene **(b)**, between 4 and 10 km at Reunion **(c)** and Irene **(d)**, and between 10 and 16 km at Reunion **(e)** and Irene **(f)**.

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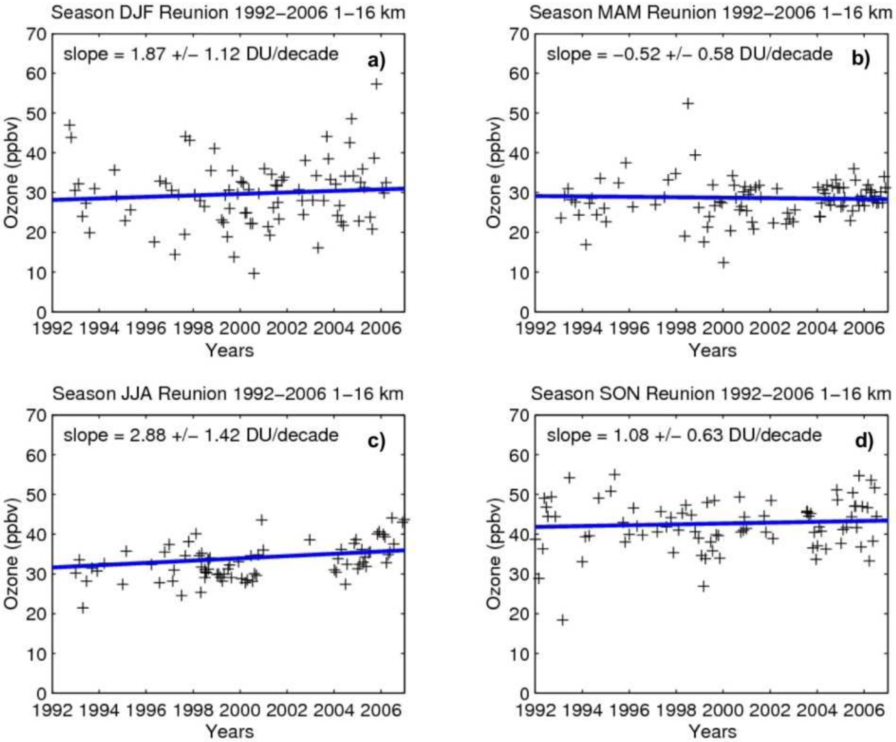

Season DJF Reunion 1992–2006 1–16 km

Season MAM Reunion 1992–2006 1–16 km

Season JJA Reunion 1992–2006 1–16 km

Season SON Reunion 1992–2006 1–16 km

Fig. 9. Ozone trends for the tropospheric layer between 1 and 16 km at Reunion in December-January-February **(a)**, March-April-May **(b)**, June-July-August **(c)**, et September-October-November **(d)**.

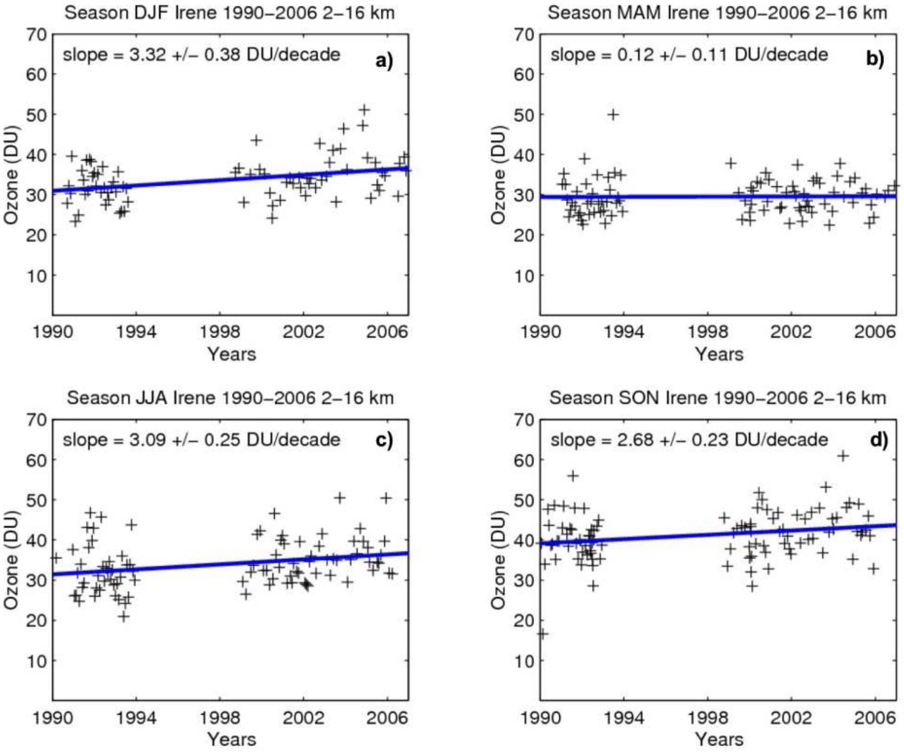

Fig. 10. Same as Fig. 9 but between 2 and 16 km at Irene.