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# A case study of extreme tropospheric ozone contamination in the tropics using in-situ, satellite and meteorological data

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**Abstract.** Since 1992, more than 150 radiosoundings have been performed from Reunion Island in the Indian Ocean in Southern tropics. On August 25, 1995, tropospheric ozone amounts of over 200 ppbv through the entire troposphere were encountered. Typical values during that period do not usually reach more than 80 ppbv. In order to explain these disturbances, we performed an analysis using satellite and ECMWF data. Results indicate that several mechanisms are working together, including photochemical effect of biomass burning associated with convection, and stratosphere-troposphere exchange near the subtropical jet stream. Based on results shown in this paper, it is difficult to estimate the relative contribution of biomass burning and stratospheric intrusions. Quantitative estimates should stem from other chemical component measurements and from a more detailed dynamical analysis.

## 1. Introduction

Since *Crutzen and co-workers'* studies [1979], biomass burning is well known to be responsible for strong enhancements of tropospheric ozone in the tropics. Contamination in low altitudes was observed over the Atlantic ocean near the south-west African coast [*Fishman et al.*, 1991]. Due to the Hadley and Walker cells, easterlies are dominant in low altitudes (trade winds), and westerlies are dominant above the wind inversion level [*Preston-Whyte and Tyson*, 1988]. That involves differences in the ozone contamination mechanisms between the western (Atlantic Ocean) and the eastern (Indian Ocean) coast of Africa. Concerning the Indian Ocean, the convection mechanisms near source regions play an important role in the ozone contamination, which occurs in the free troposphere [*Baldy et al.*, 1996].

The objective of this paper is to further scrutinize the influence of mechanisms (dynamical and chemical) responsible for tropospheric ozone contamination by analyzing a case study of extreme contamination of the free troposphere over Reunion Island (21.1°S, 55.3°E).

## 2. Radiosounding of August 25, 1995

Semi-monthly PTUO<sub>3</sub> radiosoundings (Pressure, Temperature, Humidity and Ozone) are performed at Reunion

since 1992. ECC (Electro-Chemical Cell) sondes are used to measure ozone [*Baldy et al.*, 1996]. Many radiosoundings launched in the year 1995 show strong enhancement of tropospheric ozone concentration, in particular the one launched on August 25, 1995 (Fig. 1). This radiosounding indicates tropospheric ozone amounts of over 200 ppbv above the trade wind inversion level. The extreme contamination occurs within the entire troposphere as shown by more than 150 ppbv up to the tropopause. Mean values observed during August do not exceed 80 ppbv (Fig. 1). The humidity profile, as observed typically in winter, is relatively low in the high troposphere. The value of the airflow rate of the ozone sonde sensor (202 ml.mn<sup>-1</sup>), the temperatures of ECC sonde ( $T_{ECC} < 33^{\circ}\text{C}$ ), PTU sonde ( $T_{PTU} < 27^{\circ}\text{C}$ ) and stratospheric ozone partial pressure (maximum of 160 nbar near 23 km) do not show any anomalies.

## 3. Analysis of anthropogenic source

### 3.1. Back-trajectories

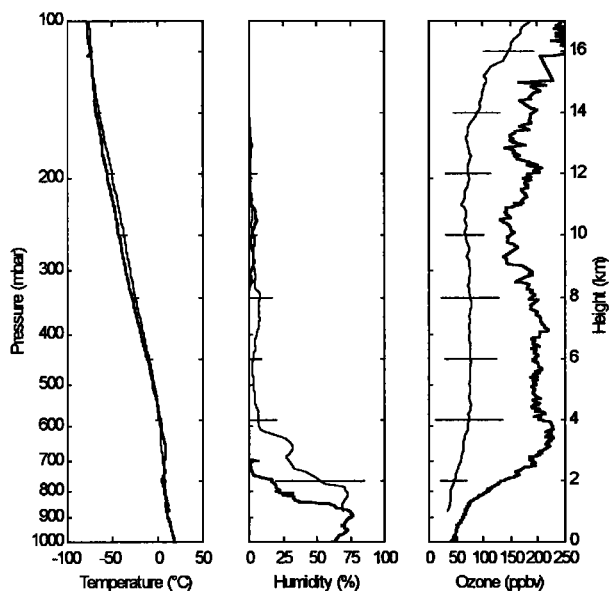
To determine the origin of the air masses on August 25, 1995, four day back-trajectories ending over Reunion were calculated with 0.75° grid ECMWF data (Fig. 2). At high altitudes (200 to 400 mb), the air masses are in recirculation above the Indian Ocean, after crossing over the northern part of Madagascar. From 450 to 500 mb, the air masses are coming from South Africa and reach the Indian Ocean after the passage to the south of Madagascar. From 600 to 750 mb, the air masses come from the south-eastern part of Africa and pass above Madagascar before reaching Reunion. Under 825 mb, the air masses have a source located to the east of Reunion. For this case study, the back-trajectories indicate that the sources of strong ozone enhancement are located to the west of Reunion.

### 3.2. Satellite

NOAA data were collected by a specific antenna of ORSTOM center on Reunion. For several days before the studied radiosounding, satellite data show a significant number of fires, in the south-eastern Africa and in Madagascar (Fig. 3a). The fires observed in the north of Mozambique are associated with strong development of smoke plumes and are propagated toward the south-west [*Randriambelo et al.*, 1998]. Ozone and its precursors emitted by biomass burning can reach the Indian Ocean, where they are injected by convective perturbations from low altitude towards the free troposphere [*Thompson et al.*, 1996]. In our case study, deep

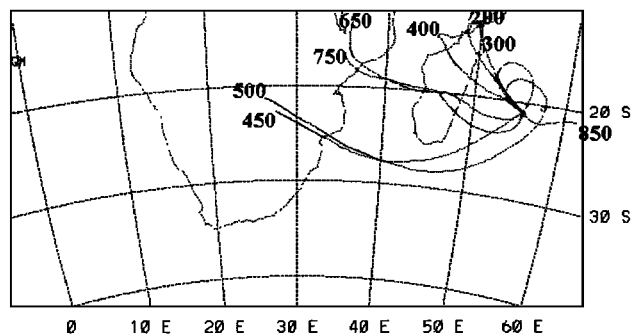
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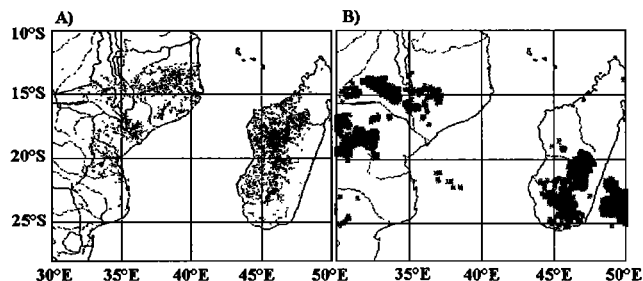


**Figure 1.** PTUO<sub>3</sub> radiosounding performed at Reunion Island on August 25, 1995 (thick line), compared with 7 averaged profiles obtained during August from 1993 to 1998. Error bars represent  $\pm 1 \sigma$ .

convective cloud events have been detected near the fire area (Fig. 3b). Moreover the rather slow horizontal transport (1500–2000 km in 4 days), can contribute to the ozone formation by “mix-then-cook” photochemistry scheme [Chatfield and Delany, 1990]. As such, the juxtaposition of fires and convection suggests a contamination of the ozone profile of August, 25, 1995 over Reunion by African and Madagascar savanna and forest fires. However such a contamination (200 ppbv) is exceptional and seems not to be explainable entirely by the vegetation fire photochemistry, whose maximum values around 100 ppbv have been observed in tropics [Thompson et al., 1996]. A soft vertical ozone gradient is to be noted just under the tropopause, between the levels 150 and 100 mb and low humidity is observed in the entire free troposphere (Fig. 1). This suggests the influence of other mechanisms. Consequently, we need to consider another ozone source. In addition to the influence of fires, did the stratosphere play a role?



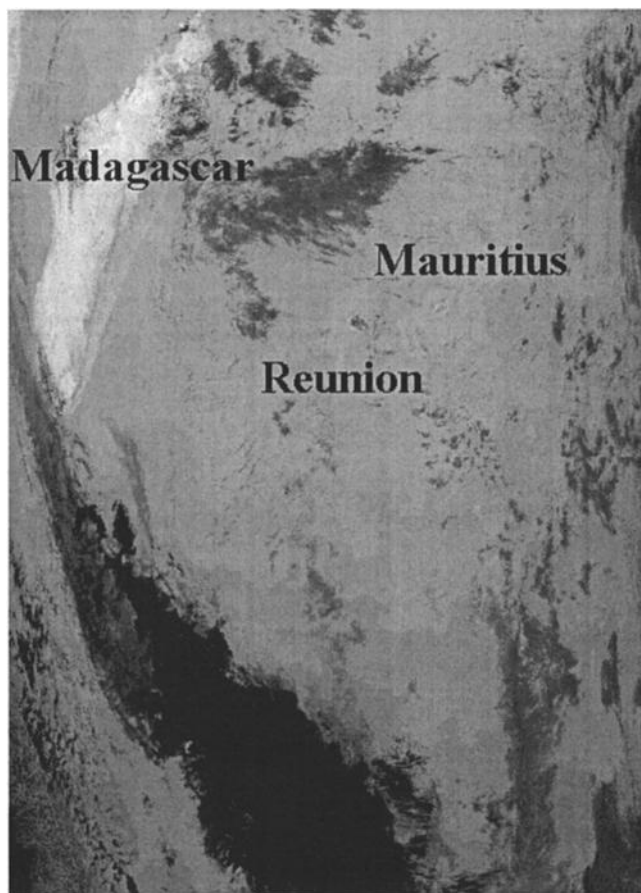
**Figure 2.** Three dimensional back-trajectories (August 21 to 25) ending over Reunion from 200 mbar to 850 mbar.



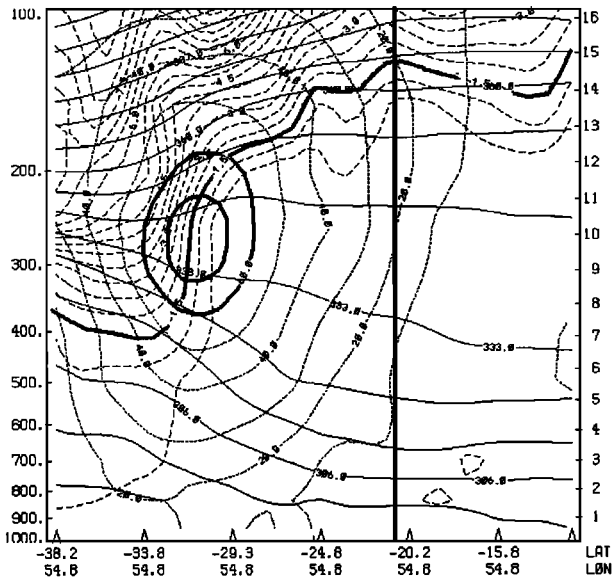
**Figure 3.** Maps showing location of (a) fires during August 20-24, and (b) deep convective cloud during August 19-20, 1995, detected from NOAA satellite

#### 4. Analysis of stratospheric source

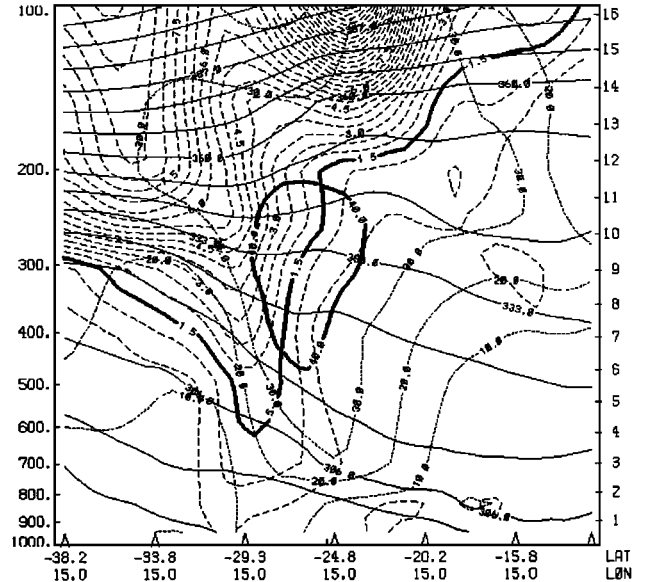
During this period (August 21-24), NOAA images (Fig. 4) show an intense cloud activity at mid-latitude, in the south-eastern part of Reunion. The frontal zone moves easterly towards Reunion. On August 21, this front is located on the southern edge of South Africa. On August 23, it is located in the Mozambique channel and reaches the south of Madagascar on August 24 (Fig. 4). It passes above Reunion on August 25, then it is in the east on August 26. The active part of the front is associated with an intense convective zone composed of strong cumulonimbus (brightness temperature



**Figure 4.** Infrared imagery (NOAA satellite) showing frontal cloud zone on August 24, 1995.



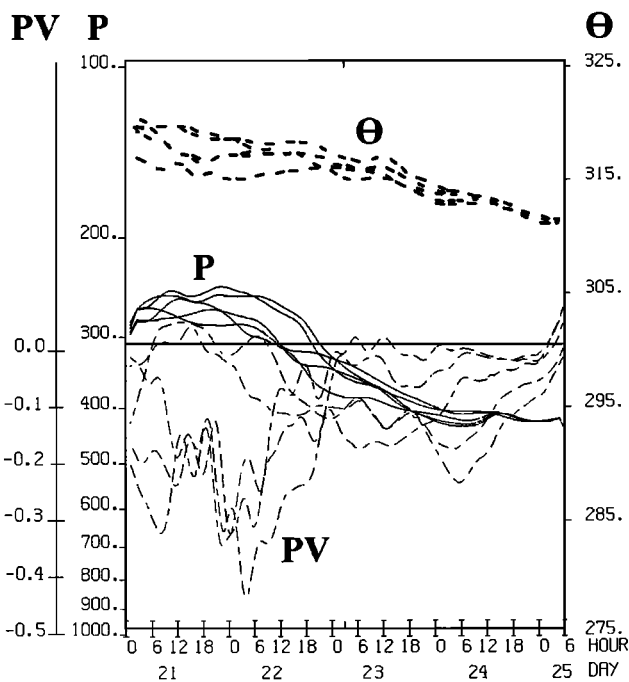
**Figure 5.** Vertical cross section (55°E) of PV (dashed curve),  $\theta$  (solid curve) and horizontal winds (dotted curve) on August 25, 1995. The PV tropopause level, jet streak and Reunion latitude have been highlighted.



**Figure 7.** Same as Fig. 5, on August 21 at 15°E.

Tb < 210°K). This frontal evolution, concordant with the back-trajectories (Fig. 2), leads us to study the possibility of a jet-streak influence and of possible stratospheric inputs. This front coincides with an intense jet streak to the south of Reunion (winds > 60 m.s<sup>-1</sup>, Fig. 5). However, on August 25, Reunion is on the anticyclonic side, in the entrance zone

of the jet streak (Fig. 5). In the following days, the jet-front system loses its strength and moves easterly. The trajectories followed by the air masses which arrived at Reunion at the 500 and 450 mb pressure levels (Fig. 2) pass near the assumed position of the subtropical barrier at this period of the year. In fact, these trajectories show the strongest subsidence. Indeed, the history of the air masses has been calculated from a back-trajectory cluster ending over Reunion ( $\pm 0.5^\circ$ ) at the 450 mbar pressure level. Figure 6 indicates that particles are characterized by strong subsidence (from 250 mb on August 21, 18h to 450 mb on August 25, 6h) associated with fluctuations of potential vorticity (August, 21 to 22) by quasi-isentropic mechanisms (potential temperature ( $\theta = 315^\circ\text{K}$ )). These characteristics suggest stratospheric intrusions during transport. Accordingly, the vertical longitudinal cross section at 15°E longitude on August 21, 1995, shows that a deep tropopause fold between 25°S and 30°S has occurred above South Africa (Fig. 7). The 1.5 PV units surface, often taken as dynamical definition of the tropopause, has descended during this episode down to 4 km (600 mb). This indicates that strong quantities of stratospheric air masses have been injected into the troposphere. These strong disturbances are in the vicinity of the origin of the back-trajectories (Fig. 2). These analyses thus showed that the tropopause fold, which occurred over South Africa, has contributed to the strong ozone concentrations of August 25, 1995, below 400 mb. On August 21, large quantities of stratospheric ozone would have been injected into the troposphere and would have been both dispersed and advected towards the Indian Ocean by the upper level westerlies. Subsidence are consistent with the near-zero humidity values observed by radiosounding. This type of exchange between stratosphere and troposphere seems close to the subtropical tropopause fold analyzed by *Baray et al.* [1998]. That event had occurred on August 11, 1993 between Madagascar and Reunion, and had been the cause of an increase in total ozone content of about twenty DU.



**Figure 6.** Temporal evolution of PV,  $\theta$  and pressure level calculated on back-trajectory cluster ending over Reunion island (450 mbar,  $\pm 0.5^\circ$ ).

## 5. Discussion

Ozone peaks were already observed in tropical zones at low altitude where photochemical ozone formation rate is very high [Thompson *et al.*, 1996]. The origin, only ascribed to biomass burning and convection, had led to enrichments generally in quite thin and localized layers. Convective zones associated with biomass burning induce vertical mixing and offer an explanation ozone concentration up to 150 ppbv. Another study has shown strong enhancements of ozone (> 200 ppbv) in high altitudes, attributed more particularly to stratospheric intrusions [Suhre *et al.*, 1997]. In our case study, the conjunction of dynamical and chemical mechanisms appears to have led to an extremely significant contamination of all the free troposphere. The very low relative humidity values associated to strong subsidence and PV anomalies suggest stratospheric influence below 450 mb. On the other hand, most of backtrajectories pass through biomass burning, as well as convection, that mix ozone precursors well up into the troposphere. Since significant convection has been observed, lightning, in addition to fires, can be a potential contributor to the ozone production, by generating NO<sub>x</sub> in the middle and upper troposphere [Thompson *et al.*, 1996]. As reported by Krishnamurti *et al.* [1993], tropospheric ozone behavior over South Atlantic Ocean is strongly influenced by a complex set of horizontal and vertical advections, transport from regions of biomass burning, and stratospheric influence. The accurate quantification of the relative contributions is very complex, because there could be an interconnection between chemical and dynamical mechanisms. For instance, the frontal systems, associated with upper level jet-streaks and westerly waves, can be at the origin of stratosphere-troposphere exchanges. Hence, they could play a role in the pollutant redistribution by ageostrophic circulations related to the jet-front system. Progress in the understanding of the complex ozone behavior in the tropics and quantitative estimates of the relative contributions should stem from the use of contour advection surgery and from mesoscale 3-D chemical and dynamical models.

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