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► **To cite this version:**

B. Sauvage, V. Thouret, Jean-Pierre Cammas, Jérôme Brioude, P. Nédélec, et al.. Meridional ozone gradients in the African upper troposphere.. Geophysical Research Letters, 2007, 34, (L03), pp.L03817. 10.1029/2006GL028542 . hal-00138042

**HAL Id: hal-00138042**

**<https://hal.science/hal-00138042>**

Submitted on 16 Jun 2022

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## Meridional ozone gradients in the African upper troposphere

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Received 31 October 2006; revised 29 November 2006; accepted 5 January 2007; published 13 February 2007.

[1] This study presents regular observations from the MOZAIC data base over Africa (1994–2004) to highlight the role of Hadley cells in tropical ozone production. We show the presence in the African upper troposphere (9–12 km) of a minimum of ozone and of a maximum of relative humidity that both follow the meridional migration of the Inter-Tropical Convergence Zone (ITCZ). We suggest that mechanisms contributing to meridional ozone gradients (0.3 to 1.5 ppbv deg<sup>-1</sup>) on both sides of the ozone minimum in the 20°S–20°N latitude band are (1) the vertical transport of relatively poor ozone air masses by tropical convection, and (2) the photochemical production of ozone in upper-level branches of the Hadley circulation. Depending on the season and on the meridional side of the ITCZ, ozone precursors injected in the ascending branch of the ITCZ include biomass burning, biogenic, lightning and anthropogenic emissions. **Citation:** Sauvage, B., V. Thouret, J.-P. Cammas, J. Brioude, P. Nédélec, and C. Mari (2007), Meridional ozone gradients in the African upper troposphere, *Geophys. Res. Lett.*, 34, L03817, doi:10.1029/2006GL028542.

### 1. Introduction

[2] The tropical troposphere has a major role in the photochemistry and energy budget of the atmosphere. Tropical regions present the combination of high chemical activity from biomass burning, biogenic and lightning emissions, and of dynamical processes allowing lifting and redistribution of the chemical species on a regional and global scale. Different analyses have investigated the role of previous sources on the ozone ( $O_3$ ) distribution especially over Africa. During TROPOZ campaign *Jonquière et al.* [1998] have shown the existence of enhanced  $O_3$  formation rates (15–35 ppbv d<sup>-1</sup>) over Northern Africa during the boreal winter due to biomass burning. This influence was confirmed by *Edwards et al.* [2003] with MOPITT carbon monoxide (CO) data. *Thompson et al.* [2000, 2003] provide meridional and zonal cross-sections showing an  $O_3$  maximum over the South Atlantic. As a complement, *Jenkins et al.* [2003] investigated the role of long-range transport and lightning in the build-up of this  $O_3$  maximum. Biomass burning and lightning seem to be the major contributors. Indeed, they both are important sources of  $NO_x$ . While biomass burning has strong influence on lower tropospheric ozone production, the  $LiNO_x$  ( $NO_x$  from lightning) ozone producing potential

by photochemistry is increased compared to other sources due to higher injection heights by convection and longer lifetimes in the upper troposphere. Lightning is the least known major source of  $NO_x$ . Estimates of the consequent  $O_3$  production is therefore uncertain.

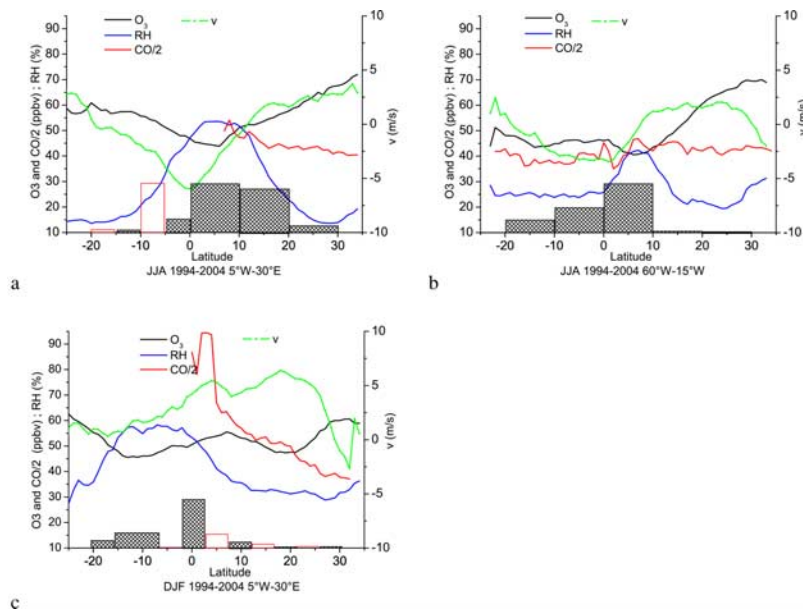
[3] The Hadley circulation drives vertical exchange via deep convection at the Inter-Tropical Convergence Zone (ITCZ). Each hemispheric Hadley cell is composed of a thermally driven rising branch of air in the equatorial band of low pressure. Divergence aloft is connected with horizontal poleward and sinking branches on the equatorward edge of the subtropical jets. Trade winds blow towards the ITCZ at the surface [*Piexoto and Oort*, 1992]. During the boreal summer, ascending branches are situated at their northernmost latitude (roughly 10°N over Africa) and migrate southward during the boreal winter. A rough zonal symmetry of Hadley cells is observed over Africa in boreal summer and over western Africa in boreal winter. *Chandra et al.* [2003] have attributed  $O_3$  maximum satellite data near 30°S and 30°N to downwelling associated with the Hadley circulation. However no study has so far investigated the Hadley cell's contribution to the redistribution of chemical species because (1) in-situ data were not previously available, and (2) there is uncertainty in the ability of a global chemistry transport model to accurately simulate convection, Hadley circulation strength and upper tropospheric ozone chemistry. This paper aims to investigate the role of Hadley cells in  $O_3$  redistribution, and to provide an accurate estimation of ozone gradients and production useful for model evaluation.

[4] During the boreal summer (monsoon season) over western tropical Africa, and during the boreal winter (dry season) over West and Central Africa, intense lightning activity [*Christian et al.*, 2003] and convection occur in the ITCZ. At the same time biomass burning occurs in the opposite hemisphere.  $O_3$  production downwind of  $LiNO_x$  and convected surface sources may then take place in the upper branches of the Hadley cells.

[5] Despite previous accomplishments, many critical gaps remain in our knowledge of the Hadley cell contributions to  $O_3$  production over the tropics. This study uses 10 years of regular in-situ measurements ( $O_3$ , relative humidity and CO) from the MOZAIC airborne program [*Marenco et al.*, 1998] to present a first comprehensive latitude cross-section of upper-tropospheric ozone over the tropics, with Africa as specific example, and to address the following questions: What could be the impact resulting from the combination of the Hadley cells and from the injection of  $O_3$  precursors (surface sources and  $LiNO_x$ ) on the tropical  $O_3$  distribution? Is there  $O_3$  formation in the upper-level branches of the Hadley cells? Can it be explained by  $LiNO_x$  injection in the ascending branches of the ITCZ? Do biomass burning emissions also participate in  $O_3$  formation in the tropical upper troposphere?

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**Figure 1.** Seasonal average of MOZAIC, LIS and ATSR data over meridional transects over (left) Africa and (right) the Atlantic during (a and b) the monsoon season (JJA), and (c) Africa during the dry season (DJF). Ozone (black, ppbv), carbon monoxide (red, ppbv divided by a factor of 2), relative humidity (blue, %), and meridional wind component  $v$  (green,  $\text{m}\cdot\text{s}^{-1}$ ). Bars represent the seasonal climatologies of LIS flash counts (light gray) and ATSR fires counts (red) averaged over  $10^\circ$  latitude bins. The time period and longitude bands used for averaging LIS and ATSR data are those used for MOZAIC data. LIS flash counts are averaged from  $0.5^\circ \times 0.5^\circ$  HRAC data. In order to match the visibility of the Y-axis on the left, a factor of  $5 \times 10^{-7}$  has been applied over the continent and of  $5 \times 10^{-6}$  over the ocean. For ATSR fire counts (red bars) a factor of  $5 \times 10^{-4}$  has been applied.

[6] Section 2 presents the first regional and seasonal analysis of  $O_3$  and water vapor meridional distribution from the 1994–2004 MOZAIC dataset, including recent CO data. This seasonal analysis is put into perspective with the meridional distribution of biomass fires and lightning flashes and a view of the zonally averaged meridional circulation over Africa. Finally, we discuss the fate of  $O_3$  precursors in the upper-level circulation over Africa.

## 2. Results of the Seasonal Analysis

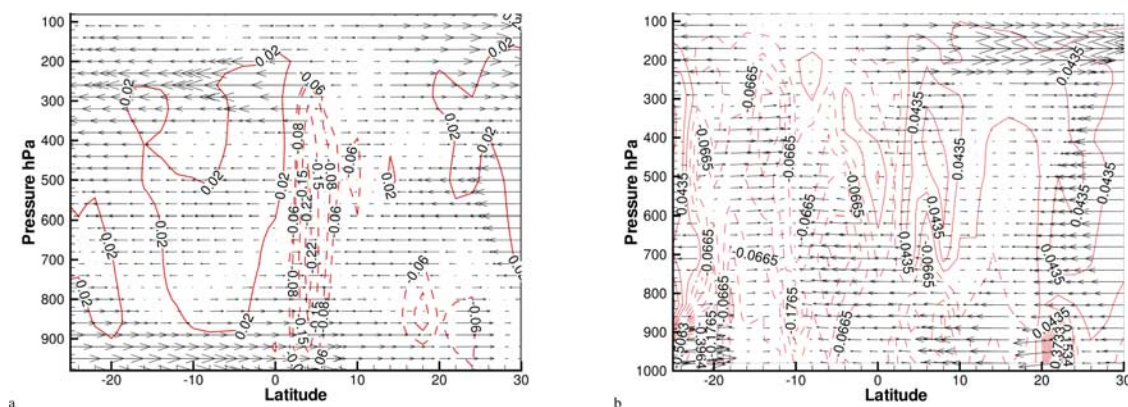
[7] MOZAIC measurements started in August 1994 on-board five longhaul aircraft for  $O_3$  [Thouret *et al.*, 1998] and water vapor [Helten *et al.*, 1998]. By the end of 2001 all aircraft were equipped with additional measurements of CO [Nédélec *et al.*, 2003]. Further details of the MOZAIC program are available at <http://mozaic.aero.obs-mip.fr/web/>. The present seasonal analysis includes about 6900 flights performed between Europe and Africa and about 3800 between Europe and South America during the 1994–2004 period. Hereafter, we will refer to the measurements for the regions of Africa and the Atlantic, respectively. In complement to aircraft data, we use the Along Track Scanning Radiometer (ATSR) space-based fire counts data <http://dup.esrin.esa.it/ionia/wfa/index.asp> and LIS (Lightning Imaging Sensor) lightning flash counts data <http://thunder.msfc.nasa.gov/data/>.

### 2.1. Perspectives From In-Situ Measurements

[8] Seasonal averages of MOZAIC, LIS and ATSR data along meridional transects over Africa and Atlantic are

shown on Figure 1. MOZAIC data collected from  $5^\circ\text{W}$  to  $30^\circ\text{E}$  (from  $15^\circ\text{W}$  to  $60^\circ\text{W}$ ) and averaged every one degree in latitude are used to build this seasonal mean over Africa (Atlantic). In order to avoid the contribution north of  $20^\circ\text{N}$  of stratospheric-origin air associated with subtropical tropopause folds [Cammis *et al.*, 1998], measurements with  $O_3$  in excess of 100 ppbv were discarded.

[9] For all regions and seasons, Figure 1 shows that the signature of the ITCZ aloft is composed of a minimum of  $O_3$  (40–45 ppbv) and of a maximum of relative humidity (RH) and lightning flashes. All features follow the ITCZ seasonal migration from  $5^\circ\text{N}$  during the monsoon season to  $10^\circ\text{S}$  during the dry season. Although the CO data set is not as complete as the others, CO over the continental ITCZ comes with relatively large volume mixing ratio (about 100 ppbv during the monsoon and 180 ppbv during the dry season) compared to the northern subtropics (80 ppbv). Anthropogenic and biomass burning emissions over Africa contribute to these relatively large CO values. The 180 ppbv CO peak just north of the equator over Africa during the dry season is located just south of the maximum of biomass burning fire counts (more than 30000 between  $0$ – $10^\circ\text{N}$ ). Uplifting of biomass burning products at the ITCZ near the coast of the Gulf of Guinea, previously advected in the lower tropospheric Harmattan flow probably explain those high CO values [Edwards *et al.*, 2003; Sauvage *et al.*, 2006]. More generally, the tropical mid to upper troposphere exhibits high CO concentrations during the local burning seasons [Jonquière *et al.*, 1998; McMillan *et al.*, 2005]. The high CO provides evidence of lower level influence and of possible associated emissions. Across the



**Figure 2.** Latitude-pressure cross-sections of meridional wind speeds (horizontal vectors in  $\text{m s}^{-1}$ ) superimposed with vertical velocities (contours lines in  $\text{Pa s}^{-1}$ , dashed for ascending motions) from ECMWF monthly analyses. (a) July 2003 in the  $5^{\circ}\text{W}$ – $30^{\circ}\text{E}$  longitude band, (b) January 2003 in the  $0$ – $15^{\circ}\text{E}$  longitude band.

Atlantic and during the monsoon season, low CO values are observed at MOZAIC cruise altitudes (80–90 ppbv). Note that we do not present Atlantic measurements during boreal winter because of a complex dynamic circulation associated with the South Atlantic Convergence Zone off Brazil.

[10] The outflow from deep tropical convection decreases  $O_3$  concentrations aloft [Folkins *et al.*, 2002]. The ozone minimum that follows the ITCZ in its migration would then be an indicator of the efficiency of the convective outflow to lower the upper tropospheric ozone background. This hypothesis is in agreement with the presence of meridional ozone gradients on both sides of the ITCZ in the meridional transport by Hadley cells. Over West Africa, emissions of ozone precursors (by either fires or lightning) and convection along the ITCZ are zonally distributed in a region of main easterly transport (mid-tropospheric African Easterly Jet and upper-tropospheric Tropical Easterly Jet). It allows the meridional 2-dimensional approach in which the role of Hadley cells is investigated. A similar process of injection of  $O_3$  precursors followed by zonal  $O_3$  buildup inside Walker cells has been addressed by Jacob *et al.* [1996], Thompson *et al.* [1996] and Moxim and Levy [2000]. In order to further establish the hypothesis that Hadley cells may be important in the regional budget of ozone in the tropical upper troposphere, we put the MOZAIC findings in a general circulation context with diagnoses built up from the ECMWF analyses and we evaluate the order of magnitude for an ozone photochemical production rate that would be associated with the observed meridional ozone gradient.

## 2.2. General Circulation Context

[11] Figure 2 represents wind projections along two latitude-pressure cross-sections over Africa. Winds are retrieved from monthly-mean ECMWF analyses for July 2003 (Figure 2a) and January 2003 (Figure 2b). The year 2003 is chosen as being the most representative of the MOZAIC dataset over Africa. In agreement with the hypothesis of a two-dimensional representation of the ITCZ, winds are averaged in longitude bands ( $5^{\circ}\text{W}$ – $30^{\circ}\text{E}$  for July and  $0^{\circ}$ – $15^{\circ}\text{E}$  for January).

[12] Figure 2a shows that during the monsoon season the  $O_3$  minimum and of maxima of RH and lightning flash counts (Figure 1) at about  $5^{\circ}\text{N}$ , are located in the upward branch of the meridional circulation. At 300–200 hPa, the

southward meridional component is the strongest, which could be related to a predominant winter cell [Tanaka *et al.*, 2004]. The vertical northward tilting of the divergence of the meridional component ( $6^{\circ}\text{N}$  at 300 hPa to  $12^{\circ}\text{N}$  at 200 hPa) agrees with MOZAIC observations, i.e. zero meridional wind observed at  $10^{\circ}\text{N}$  north of the  $O_3$  minimum.

[13] During the dry season (Figure 2b), patterns of the wind components are more complex. Over the ( $0$ – $15^{\circ}\text{E}$ ) longitude band (MOZAIC routes towards South Africa) used to average ECMWF data, the ITCZ bends southwards which interrupts its zonal symmetry. The latitude range of the upward branch of the meridional circulation is therefore extended southwards to  $10$ – $15^{\circ}\text{S}$  but nevertheless still corresponds with observations of minimum  $O_3$ , maximum relative humidity and lightning flash counts (Figure 1c). The agreement between ECMWF and MOZAIC upper-level meridional components is good in the northern part of the cross section, less good on the southern side. One reason is the few observations by MOZAIC near  $25^{\circ}\text{S}$ . The other reason is that the South Atlantic Anticyclone brings south easterly flow along MOZAIC flight routes.

## 2.3. Ozone Production Rates

[14] The hypothesis of  $O_3$  precursors injection ( $\text{LiNO}_x$  and/or biomass burning emissions) in the ITCZ followed by  $O_3$  formation in meridional branches of the upper-level circulation is pursued by the assessment of the  $O_3$  production rate that would be associated with the observed meridional  $O_3$  gradient (Table 1). Assuming steady and two-dimensional climatological mean meridional circulations, the local rate of change of ozone is zero, and the meridional advective derivative of ozone and the ozone production rate balance each other in the upper level branch of the meridional circulation. Further consideration of more realistic flow would require a 3-dimensional global chemical and transport model beyond the scope of this study.

[15] Table 1 summarizes MOZAIC upper-tropospheric seasonal means of the meridional  $O_3$  gradients and of the meridional wind component on both sides of the ITCZ, according to regions and seasons presented in Figure 1. Maximum local sources of  $O_3$  precursors are also indicated. Although lightning frequency is less over oceans than over continents [Christian *et al.*, 2003], it is intense enough over coastal African Atlantic (MOZAIC aircraft between Europe

**Table 1.** Upper Tropospheric Seasonal Means for the Meridional  $O_3$  Gradient, the Meridional Wind Component, and the Inferred  $O_3$  Production Rate Depending on Seasons and Regions

	Africa		Coastal Atlantic (Africa/Brazil)
	Boreal Dry Season DJF	Boreal Monsoon Season JJA	Boreal Monsoon Season JJA
Northern latitude band	15°S–10°N	12°N–30°N	8°N–24°N
Maximum local sources	biomass fires	lightning	lightning
Meridional $O_3$ gradient, ppbv deg <sup>-1</sup>	0.34 ± 0.07	0.94 ± 0.18	1.48 ± 0.29
Meridional wind component, m s <sup>-1</sup>	3.06 ± 5.59	2.16 ± 6.22	1.49 ± 1.62
$O_3$ production rate, ppbv d <sup>-1</sup>	0.81 ± 0.29	1.58 ± 0.88	1.72 ± 0.35
Southern latitude band		19°S–5°S	18°S–7°N
Maximum local sources		biomass fires	lightning
Meridional $O_3$ gradient, ppbv deg <sup>-1</sup>		0.62 ± 0.09	0.30 ± 0.06
Meridional wind component, m s <sup>-1</sup>		-2.95 ± 6.61	-2.22 ± 7.53
$O_3$ production rate, ppbv d <sup>-1</sup>		1.42 ± 0.51	0.52 ± 0.34

and Brazil) to be mentioned. We calculate the meridional gradients, mean wind speed and the resulting production rate between the position of the minimum of  $O_3$  (and of maximum RH) and a northern or southern limit. This limit is the latitude where the humidity is starting to increase again while the meridional wind speed component is still positive (negative) for the northern (southern) limit. If the wind speed becomes negative before a noticeable increase of RH, the limit is defined as the last latitude with positive (negative) meridional wind component.

[16] Extrema of the meridional  $O_3$  gradient are observed along flight routes off Africa and Brazil (Coastal Atlantic, right column) during the boreal monsoon season, with a maximum (minimum) of 1.48 ppbv deg<sup>-1</sup> (0.30 ppbv deg<sup>-1</sup>) in the northern (southern) cell. Table 1 generally confirms the role of the upper tropospheric meridional wind component in the poleward transport of the main precursors of  $O_3$ , either  $LiNO_x$  or biomass burning products. As explained above the seasonal-mean meridional wind component deduced from MOZAIC observations for the dry season on the southern side of the ITCZ is subject to caution due to the representativeness of data and the complex circulation in this region. A notable exception is the direction of the meridional component with respect to Hadley cells for the dry season on the southern side of the ITCZ over Africa where the seasonal mean meridional wind component deduced from MOZAIC observations is found to be equatorward instead of poleward. Only 200 flights are available over the Africa during that season.

[17] The highest production rates (1.58 and 1.72 ppbv d<sup>-1</sup>) are associated with summer cells (primarily fed by  $LiNO_x$  injection, Figure 1) in comparison to winter cells. The possible addition over Africa of the Hadley cell and of the western northward branch of the upper level high associated with the Asian monsoon [Hoskins and Rodwell, 1995] may explain the larger meridional component compared to the one over coastal Atlantic. A consequence could be the weaker meridional  $O_3$  gradient over Africa (1.58 ppbv d<sup>-1</sup>) compared to over coastal Atlantic (1.72 ppbv d<sup>-1</sup>).

[18] Winter cells that would mostly be associated with injection of  $O_3$  precursors by biomass fires get intermediate production rates (0.81 and 1.42 ppbv d<sup>-1</sup>). These rates are comparable to previous estimates obtained during the TRACE-A campaign downwind of biomass burning regions [Jacob et al., 1996; Pickering et al., 1996; Thompson et al.,

1996]. However our rates are estimated along meridional transport, in contrast with zonal transport for previous studies. The lower production rate found for the northern winter cell (0.81 ppbv d<sup>-1</sup>) compared to the southern winter cell (1.42 ppbv d<sup>-1</sup>) is consistent with the confinement of biomass burning emissions observed in the lower troposphere of West Equatorial Africa compared to Central-South Africa [Sauvage et al., 2005]. Finally, in relation to the minimum of local sources, the lowest production rate (0.52 ppbv d<sup>-1</sup>) is found over the coastal Atlantic in winter cells.

[19] The order of magnitude for  $O_3$  production, 0.5–1.7 ppbv d<sup>-1</sup>, is comparable with results of  $O_3$  production by  $LiNO_x$  of DeCaria et al. [2005] when averaged for their entire model domain [see DeCaria et al., 2005, Figure 12b]. Relative variations of this order of magnitude seem to highlight the importance of each source (lightning versus biomass burning) and of the strength of the meridional transport in the construction of the meridional  $O_3$  gradient.

### 3. Conclusions and Perspectives

[20] This study demonstrates the persistent role of the Hadley cell meridional transport in the tropical  $O_3$  distribution. It highlights the observation in the upper troposphere over equatorial Africa and Atlantic of a minimum of ozone ( $O_3$ ) and of a maximum of RH that follow the meridional migration of the ITCZ. In-situ measurements of  $O_3$  and water vapour come from the MOZAIC programme since 1994. Meridional gradients of  $O_3$  are observed on both sides of the ITCZ.

[21] To test the hypothesis of  $O_3$  formation in meridional branches of the upper level circulation, MOZAIC measurements for  $O_3$ , water vapour and the meridional wind component have been analysed at the seasonal time scale for the monsoon and dry boreal seasons. Extrema of the meridional  $O_3$  gradient are observed along flight routes off Africa and Brazil (Coastal Atlantic) during the boreal monsoon season, with a maximum (minimum) of 1.48 ppbv deg<sup>-1</sup> (0.30 ppbv deg<sup>-1</sup>) in the northern (southern) cell. The order of magnitude for the  $O_3$  production rate, calculated as the meridional component times the meridional  $O_3$  gradient, is 0.5–1.7 ppbv d<sup>-1</sup>. Highest production rates are shown to be associated with summer cells likely fed with  $LiNO_x$  injections and associated with relatively weak meridional components. Winter cells associated with injection of  $O_3$  precursors by surface sources have a relatively stronger

meridional component and hence intermediate production rates. These results are consistent with the dynamics of Hadley cells – winter cell dominating the summer cell in intensity [Tanaka et al., 2004] – and with chemistry model results for the order of magnitude for the  $O_3$  production rate in the environment of thunderstorms [DeCaria et al., 2005].

[22] Finally, this study emphasizes the role of the Hadley cells in the meridional  $O_3$  formation over the tropics. The quantitative assessment of this estimation of the  $O_3$  rate needs to be further investigated with modeling and the input of new data sets, like that from the recent AMMA experiment [Mari and Prospero, 2005], the MOZAIC data collected by Air Namibia in 2006, and space-based data from sensors having observational capacities in the tropical upper troposphere. It would also help in further investigate the chemistry-climate coupling by evaluating global modeling capability in the reproduction of Hadley cell circulation and  $O_3$  photochemistry along.

[23] **Acknowledgments.** We acknowledge MOZAIC funding agencies, the European Commission, INSU-CNRS (France), Forschungszentrum Jülich (Germany), and Météo-France. We are indebted to participating airlines (Lufthansa, Air France, Austrian Airlines, and former Sabena) for carrying free of charge the MOZAIC equipment and performing the maintenance. We also acknowledge the European Agency, ESA/ERIN. Randall V. Martin (Dalhousie University) is acknowledged for proofreading the final version of the paper.

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