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Climatology of tropospheric ozone and water vapour over Chennai: a study based on MOZAIC measurements over India

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ABSTRACT: Climatology of tropospheric ozone (O₃) and water vapour (H₂O) over a metro city of Chennai in southern India has been presented based on the Measurements of OZ one and water vapour by Airbus I n-service AirCraft (MOZAIC) data during 1996–2001. The seasonal trends in mixing ratio of O₃ are discussed in view of annual changes in the transport pattern, rainfall, and its relationship with ambient H₂O. The vertical distributions of both O₃ and H₂O show large seasonal variations. The observed high mixing ratios of O₃ in winter and spring months can be attributed to long-range transport of pollutants by the northwesterly winds associated with winter monsoon circulation. The lowest mixing ratio of 15 ppbv of O₃ from July to September coincides with the summer monsoon circulation over Indian subcontinent. The surface level mixing ratio of O₃ was observed to be below 30 ppbv throughout the year. On the other hand, monthly rainfall and vertical distributions of H₂O show just opposite seasonal variations to that of O₃ mixing ratio. Chennai is a coastal city; therefore, the seasonal change in surface wind flow causes large variability in H₂O concentrations of 6–30 g/kg in the lower troposphere. More or less similar annual patterns of mixing ratios of O₃ and H₂O are repeated during different years of 1996–1998, except for some small-scale differences. High level of O₃ mixing ratio, particularly during the early winter months of year 1997, seems unusual as we do not see such trends for other year of measurements over Chennai. The case studies of profiles of O₃, relative humidity (RH), and temperature in view of long-range transport over Chennai during the different seasons have been also discussed. Significant enhancements in tropospheric O₃ were observed over Chennai due to transport of air from forest fire regions of Indonesia during October 1997.

KEY WORDS rain; Chennai; transport; marine; troposphere; tropical; ozone; fire

1. Introduction

Tropospheric ozone (O₃) plays a key role in the chemistry of the atmosphere, with effects on both the environment and Earth's climate. In the troposphere, photolysis of O₃ initiates the oxidation process through the formation of hydroxyl radicals (OH). Ozone, because of its strong absorption band centred at 9.6 μm, also plays an important role in global warming, particularly in the upper troposphere (Forster and Shine, 1997; Gauss *et al.*, 2003). The photochemical reactions are dominated by the cycles of O₃ and water vapour (H₂O) in the tropical region, which receives intense solar radiation. H₂O is also a greenhouse gas (Ramanathan *et al.*, 1985; Wigley, 1987).

Tropospheric O₃ levels have increased since pre-industrial times (1860), particularly in the Northern

Hemisphere (NH) (Oltmans *et al.*, 1998). It is estimated that O₃ has contributed +0.35 W/m² to the radiative forcing since 1860, compared with +1.66 W/m² from the CO₂ (Forster *et al.*, 2007). Tropical region is a large contributor to the global O₃ budget. Moreover, it plays a key role in controlling the chemistry and climate of the tropical troposphere. Human activities increase O₃ concentration in the troposphere through emissions from fossil fuel combustion and biomass burning (Crutzen *et al.*, 1979; Logan *et al.*, 1981; Crutzen and Andreae, 1985).

The tropical continental boundary layer (CBL) is an important source region for photochemical production of O₃. In tropics, biomass burning and fossil fuel combustion generate large amount of precursors such as CO, methane (CH₄), and non-methane hydrocarbons (NMHCs) (Houghton *et al.*, 1996; Kirchhoff, 1996). Therefore, vertical variations of O₃ reflect the heterogeneity of this region, arising from O₃ precursor sources (biomass burning), sinks (photolysis, photochemistry),

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and convective processes (Chatfield and Crutzen, 1984; Liu *et al.*, 1987; Davies *et al.*, 1998). Lateral transport from a source in the upper troposphere or perhaps the lower stratosphere, or O₃ and its precursors from a photochemically very active source region have also been observed to play important role in the distributions. An important factor in transport studies is the lifetime of O₃. Ozone is photodissociated by short-wave solar radiation (<340 nm) into electronically excited O(¹D) atoms and with H₂O to form OH radicals. Thus, the lifetime of O₃ is basically determined by the amount of H₂O and solar radiation. The lifetime of O₃ increases from 2 to 5 days in the moist tropical marine boundary layer (MBL) to approximately 90 days in the free troposphere (Fishman *et al.*, 1991). Thus, once O₃ is lifted from the boundary layer it can be transported far away from its source regions.

Typical vertical distributions of O₃ in the tropics exhibit a minimum at the surface, a maximum at ~6.5 km, another minimum at 11.2 km, and a subsequent increase towards the tropopause (Folkins *et al.*, 2002). The major factors that determine the vertical variation of O₃ are *in situ* production/destruction, deep convective outflow, vertical advection, shallow convection, eddy exchange with the lowermost stratosphere, convective downdrafts, and vertical mixing near the tropical tropopause associated with convective overshooting (Sherwood and Dessler, 2001) and Kelvin waves (Fujiwara *et al.*, 1998).

The detailed characteristics of O₃ in the tropical troposphere remain unresolved because of a lack of comprehensive observations. The number of ozonesonde stations in the northern tropics is limited (Logan, 1999), although Hilo, Kuala Lumpur, Surinam, Benin (Thouret *et al.*, 2009), and Costa Rica have all participated in SHADOZ (Thompson *et al.*, 2003; <http://croc.gsfc.nasa.gov/shadoz>). The main objective of this work is to study the tropospheric O₃ profile in view of annual changes in the transport pattern and also its relationship with H₂O based on the Measurements of O₃ and H₂O by Airbus In-service Aircraft (MOZAIC) data over Chennai, India. We have also presented the case studies to investigate the impact of long-range transport in the vertical distributions of O₃ and H₂O over Chennai.

2. Overview of MOZAIC program and measurements over India

The MOZAIC program was initiated in 1993 by European scientists, aircraft manufacturers, and airlines to collect experimental data. Ozone and H₂O sensors were installed on board five long-range commercial aircrafts (Airbus A340's) of four participating airlines. The MOZAIC project, in operation since August 1994, is a European Union (EU) supported project for the automatic measurements of O₃ and water vapour on board AIRBUS A340 aircraft on scheduled flights. MOZAIC is sponsored by

major European airlines: Air France (one aircraft), Austrian Airlines (one aircraft), Lufthansa (two aircrafts), and Sabena/Air France (one aircraft).

The ozone analyser is a dual-beam ultraviolet (UV) absorption instrument (Thermo-Electron Model 49–103), which has a detection limit of 2 ppbv. Uncertainties in the measurements are estimated to be $\pm(2 \text{ ppbv} + 2\%)$, although actual performance is better (Thouret *et al.*, 1998a, 1998b). The response time of the instrument is 4 s, which translates into a vertical resolution of 30 m. Full details of calibration procedures and mounting in the aircraft are also given by Thouret *et al.* (1998b). Relative humidity (RH) and temperature are measured with a capacitive sensor (Humicap-H) and a platinum resistance sensor (PT100), respectively. The Humicap-capacitor together with the PT100 resistor is mounted at the top of an axis-symmetric body, which is especially designed for installation in an appropriate housing.

MOZAIC data combined with meteorological data are being used to (1) establish a quasi-global climatology of the large-scale distributions of O₃ and H₂O in the troposphere; (2) establish the vertical and temporal distributions of O₃ and H₂O at various locations (airports) over the continents; (3) investigate the seasonal and geographical variations in relation to their natural and anthropogenic sources, stratosphere–troposphere exchange (STE), horizontal and vertical air circulation; and (4) to validate chemistry and transport models (Marenco *et al.*, 1998). Measurements taken during both take off and landing of the aircraft have been used as vertical profile data over the airports. To supplement some gaps in MOZAIC database during August or July months, we have used the Indian Metrological Department (IMD) data measured at Trivandrum (as this site also represents similar climatology as over Chennai). The MOZAIC vertical profiles of O₃, H₂O and meteorological parameters over the Indian subcontinent consist of measurements at three airports of metropolitan cities, namely Chennai (Madras) (12.99°N, 80.15°E), Delhi (28.56°N, 77.11°E), and Mumbai (Bombay) (19.09°N, 72.87°E). In this study, we have discussed the distributions of O₃ and H₂O over Chennai.

3. Seasonal transport pattern over Chennai

Chennai metropolitan area extends over 1180 sq km and has a population of more than 8.5 million. Rapid increase in urbanization with vehicle congestion has increased on the roads of Chennai. As a result of this, gaseous pollutants are continuously increasing in the ambient air of Chennai city (Senthilnathan, 2008). The daily rainfall data measured by the IMD for years 1996–2001 are plotted in Figure 1. Chennai is a coastal city and very often influenced by the transport of maritime air due to land see breeze circulation. The northeast (NE) monsoon period from October to December is the principal rainy season, whereas the southwest (SW) monsoon from June to September is the secondary rainy season for this place.

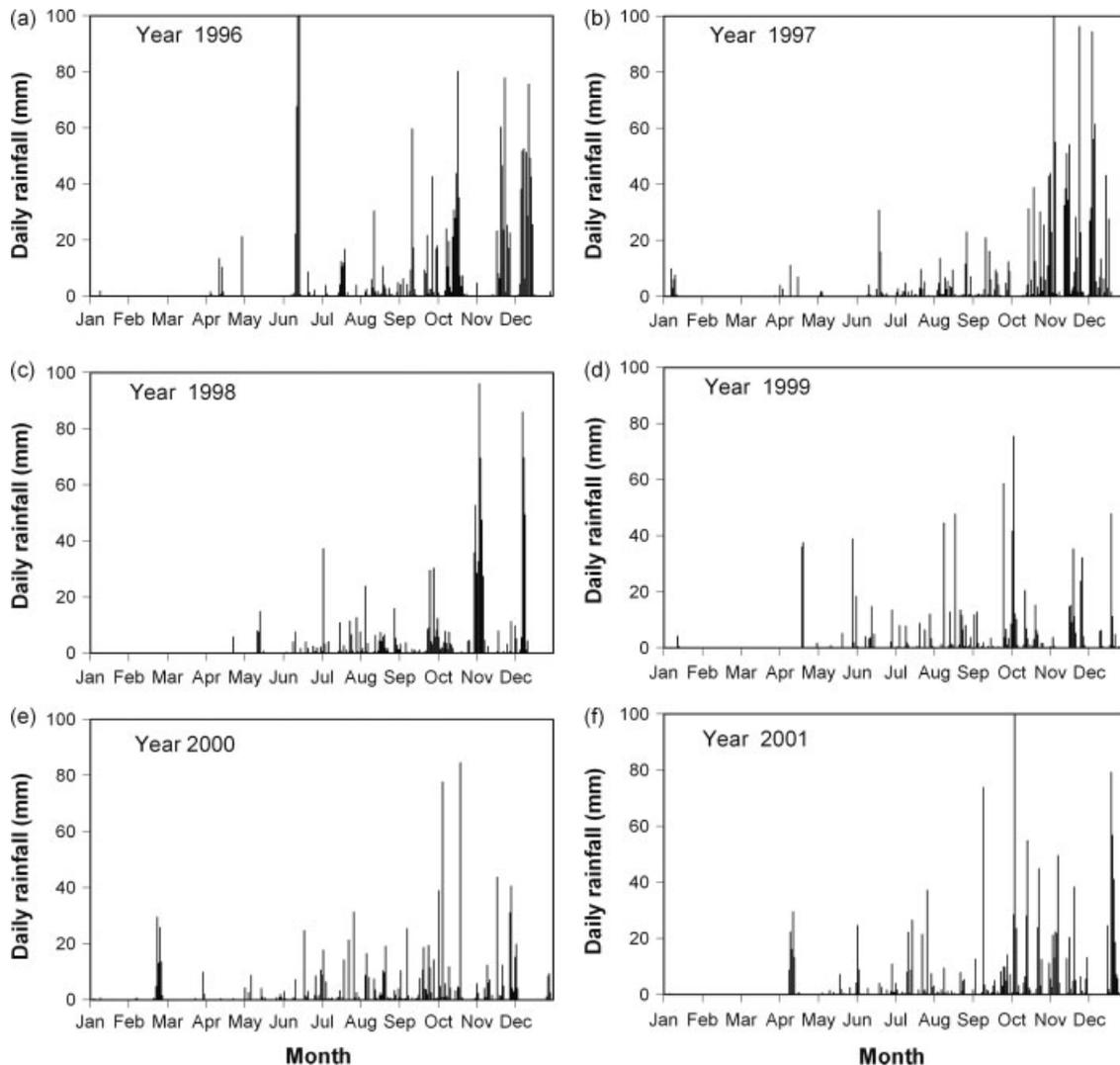


Figure 1. Daily rainfall variations measured at Chennai for different years during 1996–2001 period [data taken from Indian Meteorological Department (IMD)].

In the lower troposphere, the seasonal change in the wind flow pattern over Southeast Asia is mainly associated with the onset and withdrawal of monsoon circulations (Asnani, 1993). The summer monsoon circulation is generally visualized as large-scale convergence of mass and moisture over the Indian subcontinent in the lower levels, while a strong upper level divergence aloft. Surface heating during summer favours enhanced convection over land, which is fuelled by moist inflow at low levels. We have used the National Centre for Environmental Prediction (NCEP) data to derive the wind streamline flow over India and surrounding marine regions. Figure 2 shows the mean wind streamline plots during the winter months of January–March (JFM) at four different pressure levels of 925, 700, 400, and 250 mb during 1996–2001. In the winter season, wind flow at 700 mb is from NE, while in the upper troposphere (see plot at 250 mb) westerly wind flow prevails over Chennai. In the summer monsoon season from July to September (JAS) months, the wind streamlines at 925 mb show the SW flow, while in the upper troposphere the flow is easterly

over Chennai (Figure 3). In the middle troposphere, the wind patterns show large seasonal variations over Chennai.

4. Seasonal variations in O_3 over Chennai during 1996–1998

4.1. Monthly mean profiles of O_3 and H_2O during 1996

We have calculated the monthly mean of MOZAIC profile data to study the seasonal variations of O_3 (ppbv) and H_2O (g/kg) in the troposphere (0–8 km). Figure 4 shows the vertical distributions over Chennai for year 1996. It can be very clearly seen that profiles of both O_3 and H_2O show large seasonal variability. The mixing ratio of O_3 near surface is always below 30 ppbv throughout the year. In a good agreement to this result, the mean surface O_3 mixing ratio of ~10–25 ppbv was observed at an urban site of Chennai during year 2006 (Pulikesi *et al.*, 2006). During the winter months, mixing

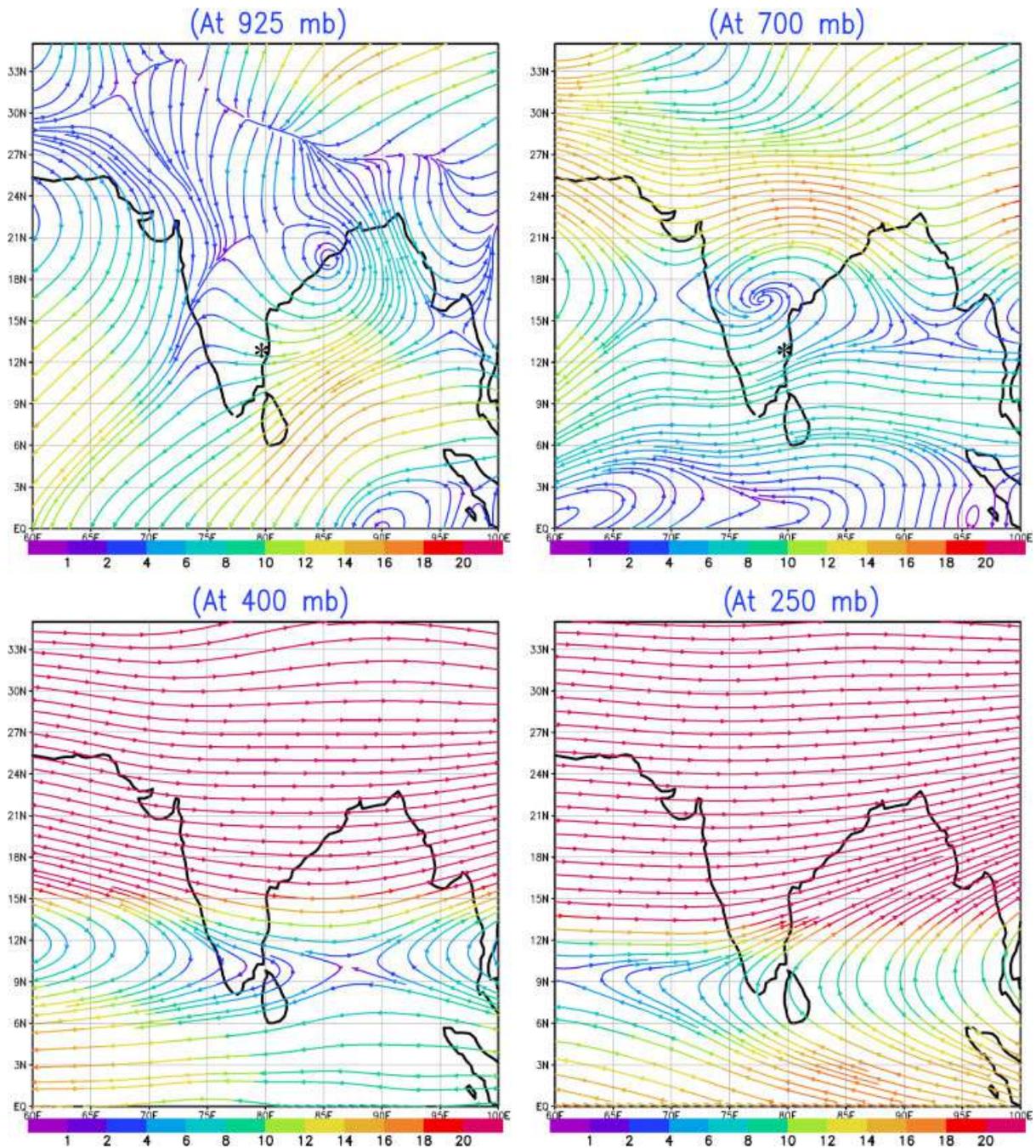


Figure 2. Mean wind fields (NCEP data) at four different pressure levels during winter (January, February, and March) of 1996–2001 period over continental and surrounding marine regions of India (* symbol shows the location of Chennai). Colour bar represents the magnitude of wind speed. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

ratio of O_3 varies from 40 to 50 ppbv without any significant vertical gradients in the free troposphere, between 1 and 8 km of altitudes. Measurements during April–June months exhibit elevated mixing ratio of 50–70 ppbv in the middle troposphere between 3 and 8 km of altitudes. While consistently low mixing ratios of tropospheric O_3 were observed from August to December months. In this period of summer monsoon circulation, vertical distributions of O_3 show steady increasing trend with height as the mixing ratios were 10–15, 10–20, 20–30 and 30–40 ppbv between the altitudes of 0–2, 2–4, 4–6 and 6–8 km, respectively. On the other hand, the

seasonal variability in H_2O concentration of 6–30 g/kg was observed in the lower troposphere over Chennai.

Chennai is a metro city of India where anthropogenic emissions of precursors of O_3 are continuous throughout the year. As expected for a typical urban site, the diurnal variations of ozone at Chennai exhibit elevated mixing ratio during daytime with low values at night (Pulikesi *et al.*, 2006). Therefore, O_3 mixing ratios near the surface were always low as the MOZAIC profile measurements were always taken during the night hours. In the spring months (April and May), observed high concentrations of O_3 in the lower and mid-troposphere could be due to

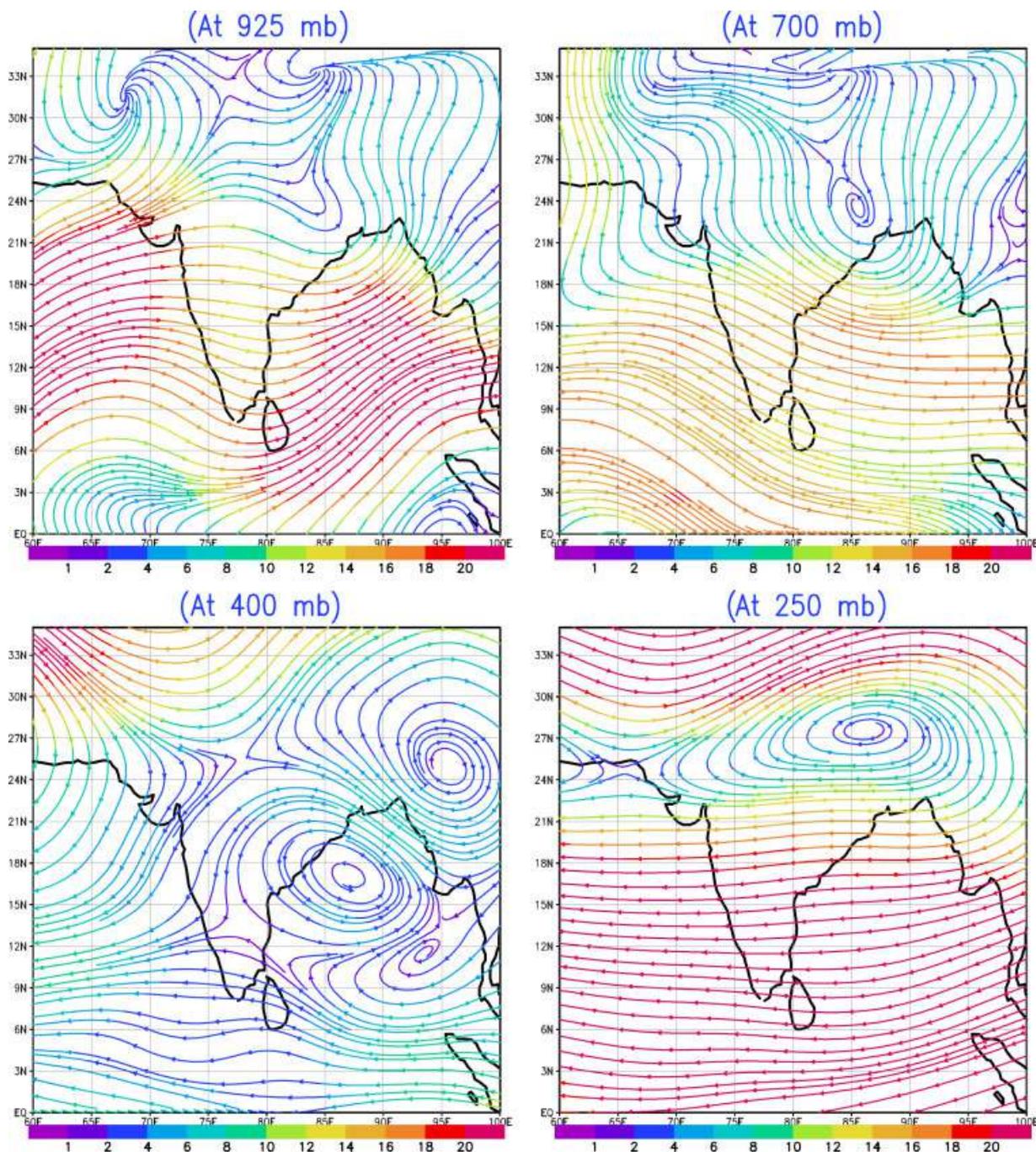


Figure 3. Mean wind fields (NCEP data) at four different pressure levels during summer (July, August, and September) of 1996–2001 period over continental and surrounding marine regions of India. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

the long-range transport of pollutants from the continental region of Asia (Hastenrath, 1985; Lal *et al.*, 2007). On the other hand, the observed low mean concentration of ~ 15 ppbv over Chennai is due to advection of cleaner air from the Indian Ocean during June to September. This is the period when frequency and amount of rainfall are highest at Chennai due to the influence of summer monsoon circulation. In a good agreement with this observation, the mixing ratios of O_3 were observed to be in the range of 10–20 ppbv over the southern region of Bay of Bengal during the summer monsoon campaign (Sahu *et al.*, 2006). To investigate how the rainfall

might have linked with the seasonality in O_3 distributions, the monthly mean tropospheric mixing ratio of O_3 and rainfall recorded per month over Chennai are plotted in Figure 5. Annual variations of O_3 and rainfall show just opposite trends for year 1996 (Figure 5(a)). The rainfall was lowest from January to June months (year 1996 was an exception), while mixing ratio of O_3 was elevated at 35–60 ppbv during this period of measurements over Chennai. The onset of summer monsoon circulation (cleaner marine air), usually associated with rainfall and convective clouds, coincides with the decline in mixing ratio of O_3 from ~ 60 to 25 ppbv in upper troposphere

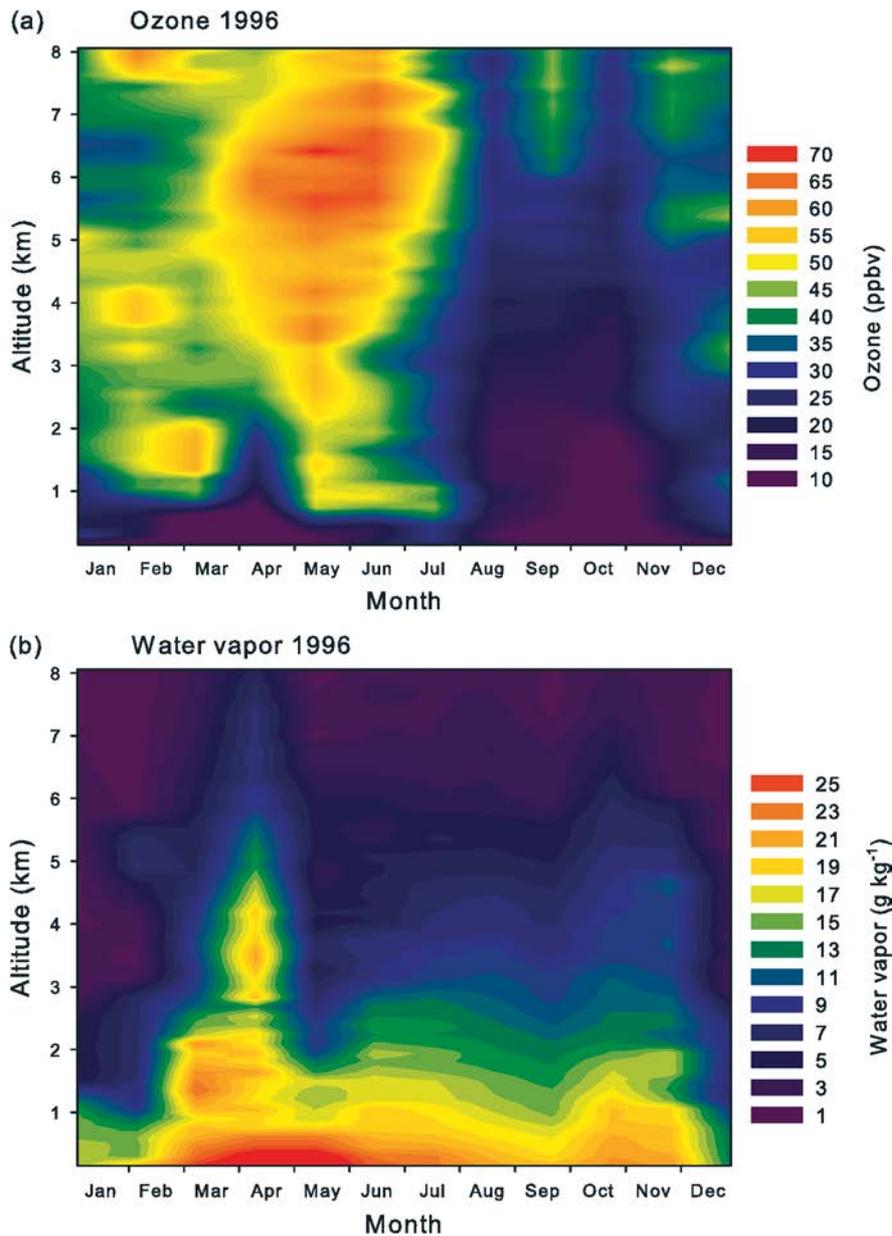


Figure 4. Monthly mean contour plots of (a) ozone and (b) water vapour representing the annual variations over Chennai during 1996. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

and ~ 40 to 15 ppbv in lower troposphere. Later, from October to December, the rainfall increases over Chennai. However, O_3 is almost constant at ~ 20 ppbv in the lower troposphere and ~ 40 ppbv in the upper troposphere partly due to the transport of polluted air from Indo-Gangetic plain (discussed in case studies). This insensitivity of O_3 mixing ratio to the increasing rainfall over Chennai could be partly due to the balance between the transport of pollutants and photochemical sink of O_3 . During the early winter monsoon circulation, the NE wind flow brings continental pollutants from Asia; however, concentrations of O_3 and other pollutants could be diluted due to intense rainfall and related chemical processes over Chennai. The importance of the processes leading to low mixing ratio of O_3 , whether simple dilution or presence of additional O_3 sink due to high H_2O ,

needs further investigation. The monthly means of tropospheric O_3 and H_2O over Chennai for measurements during 1996–1998 are shown in Table I.

Space-based observations from Measurements of Pollution in the Troposphere (MOPITT) (Kar *et al.*, 2004), the Microwave Limb Sounder (MLS) (Jiang *et al.*, 2007; Park *et al.*, 2007), the Atmospheric Composition Experiment (ACE) (Park *et al.*, 2008), and the Atmospheric Infrared Sounder (AIRS) (Randel and Park, 2006) also show how deep convection associated with the summer monsoon lofts boundary layer air with low O_3 and high H_2O from Asia into the upper troposphere (Gettelman *et al.*, 2004). This pattern occurs also over Nairobi, a continental site that is located in a meteorological regime more akin to the Indian Ocean than central or southern Africa (Chatfield *et al.*, 2004, 2007).

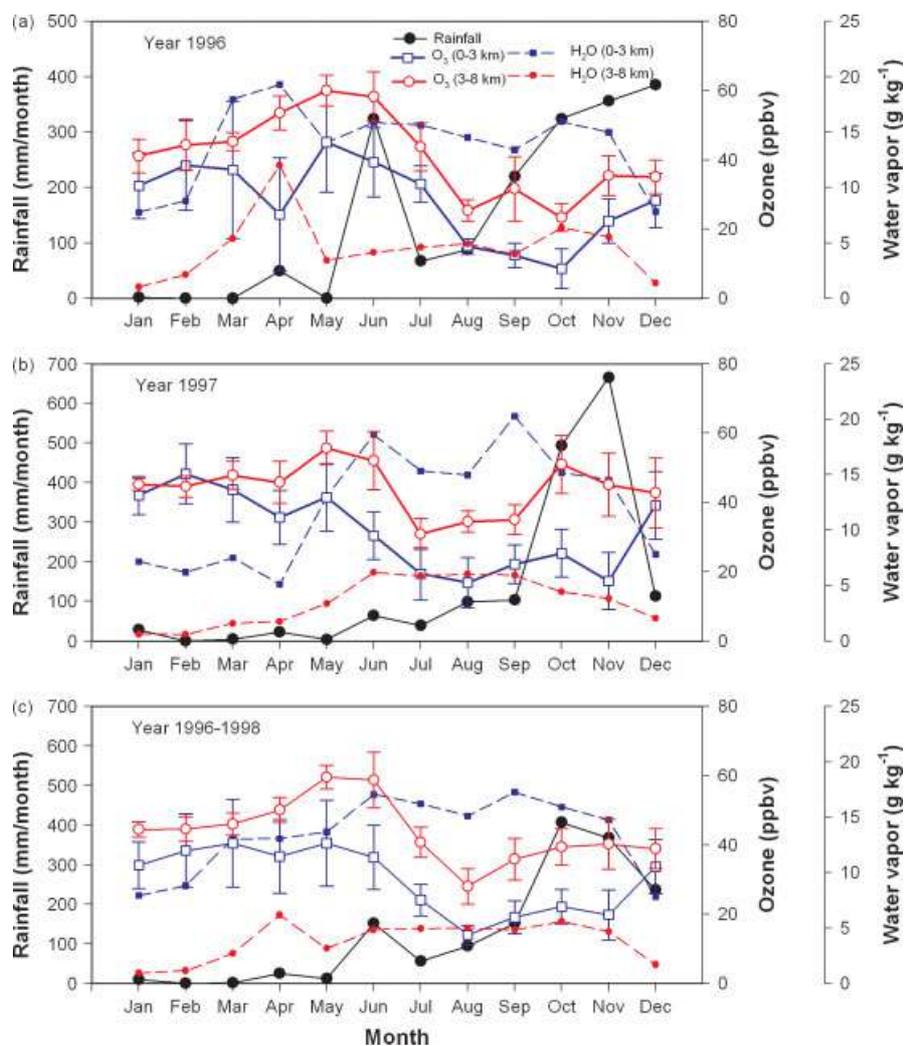


Figure 5. Monthly mean tropospheric (0–3 and 3–8 km of altitudes) ozone, water vapour, and rainfall per month over Chennai for 1996–1998 period (a–c). This figure is available in colour online at wileyonlinelibrary.com/journal/joc

Table I. Mean mixing ratios of O₃ and H₂O between 0 and 10 km of altitudes over Chennai during 1996–1998.

Month	Year 1996		Year 1997		Year 1996–1998	
	O ₃ (ppbv)	H ₂ O (g/kg)	O ₃ (ppbv)	H ₂ O (g/kg)	O ₃ (ppbv)	H ₂ O (g/kg)
January	32.8	3.5	45.7	3.0	39.3	4.4
February	41.8	4.5	45.6	2.6	41.5	5.0
March	41.8	9.8	47.0	3.7	43.2	7.9
April	43.5	14.2	41.7	2.9	43.4	9.6
May	50.6	7.1	51.4	6.7	50.0	8.4
June	48.4	8.3	46.4	10.4	47.6	10.9
July	37.3	8.5	28.4	9.1	32.4	10.6
August	19.7	8.2	30.0	9.0	21.0	10.1
September	23.3	7.2	32.3	10.9	27.5	11.0
October	18.3	9.6	45.8	8.1	30.8	10.8
November	25.2	8.7	38.8	7.5	30.0	9.7
December	29.2	3.7	42.1	4.0	36.3	4.8

4.2. Monthly mean profiles of O₃ and H₂O during 1997 and 1998

Figure 6 shows the seasonal variations of tropospheric O₃ and H₂O over Chennai for year 1997. Generally,

the seasonal pattern of mixing ratio of O₃ is similar to that observed during 1996. The major factors such as emissions of O₃ precursors from biomass burning sources and pattern of transport controlling the distribution of O₃ in the troposphere may change significantly from year

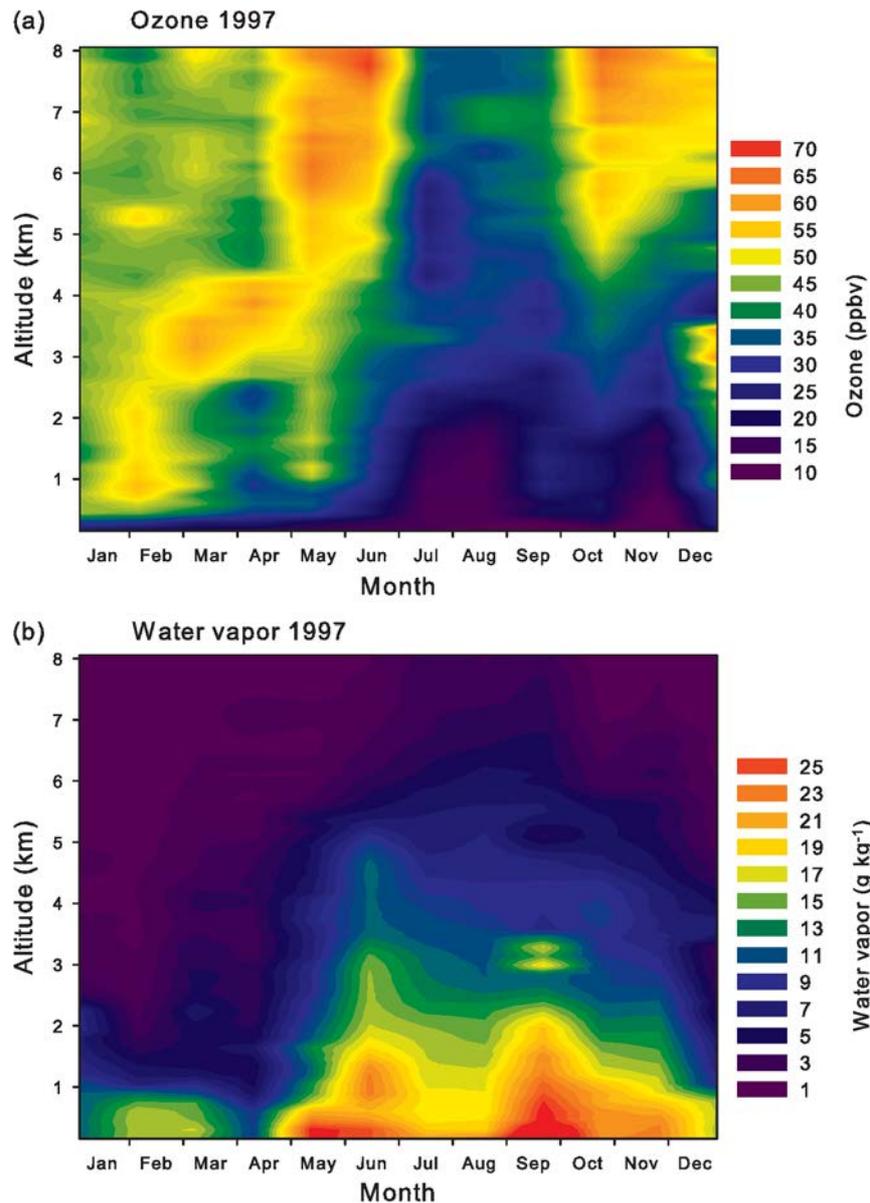


Figure 6. Monthly mean contour plots of (a) ozone and (b) water vapour representing the annual variations over Chennai during 1997. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

to year. The main features are same for the years 1996 and 1997; however, some small-scale differences can be noticed in the year-to-year distributions of O_3 over Chennai. The onset of episodes of low O_3 during the summer monsoon starts from June in 1997, while it was from July during 1996. In the upper troposphere (6–8 km altitudes), mixing ratio of O_3 varies between 30–40 and 50–60 ppbv during 1996 and 1997, respectively.

Similar to the observation for year 1996, the annual variations of O_3 and rainfall data also show opposite trends during 1997 (Figure 5(b)). The rainfall was lowest from January to June months, while mixing ratio of O_3 was elevated at 40–55 ppbv during this period of measurements. As has been explained the onset of summer monsoon brings cleaner marine air and rainfall, therefore mixing ratio of O_3 declines from ~ 45 to 35 ppbv during July–August period. Later, from August

to November, the rainfall data exhibit increasing trend, while mixing ratio of O_3 increases and it becomes ~ 45 ppbv in October month. The increasing trend in O_3 mixing ratio, particularly during the early winter months of year 1997, seems to be an unusual feature as we do not see such trends for both the years of 1996 and 1998 (Figure 5(a) and (c)). If we see the daily rainfall pattern during the late monsoon and early winter months, from August to mid-October, year 1997 seems to have influenced by spells of dry weather than those in other years of 1996–2001 (Figure 1). Interestingly, for the same period of year 1997 the mixing ratios of O_3 in middle and upper troposphere over Chennai were higher than those in 1996 and 1998. The year 1997 corresponds to El Niño events when convective activity becomes low in Indonesia, in this region, extensive forest fire activities were reported due to sparse precipitation (Fujiwara *et al.*,

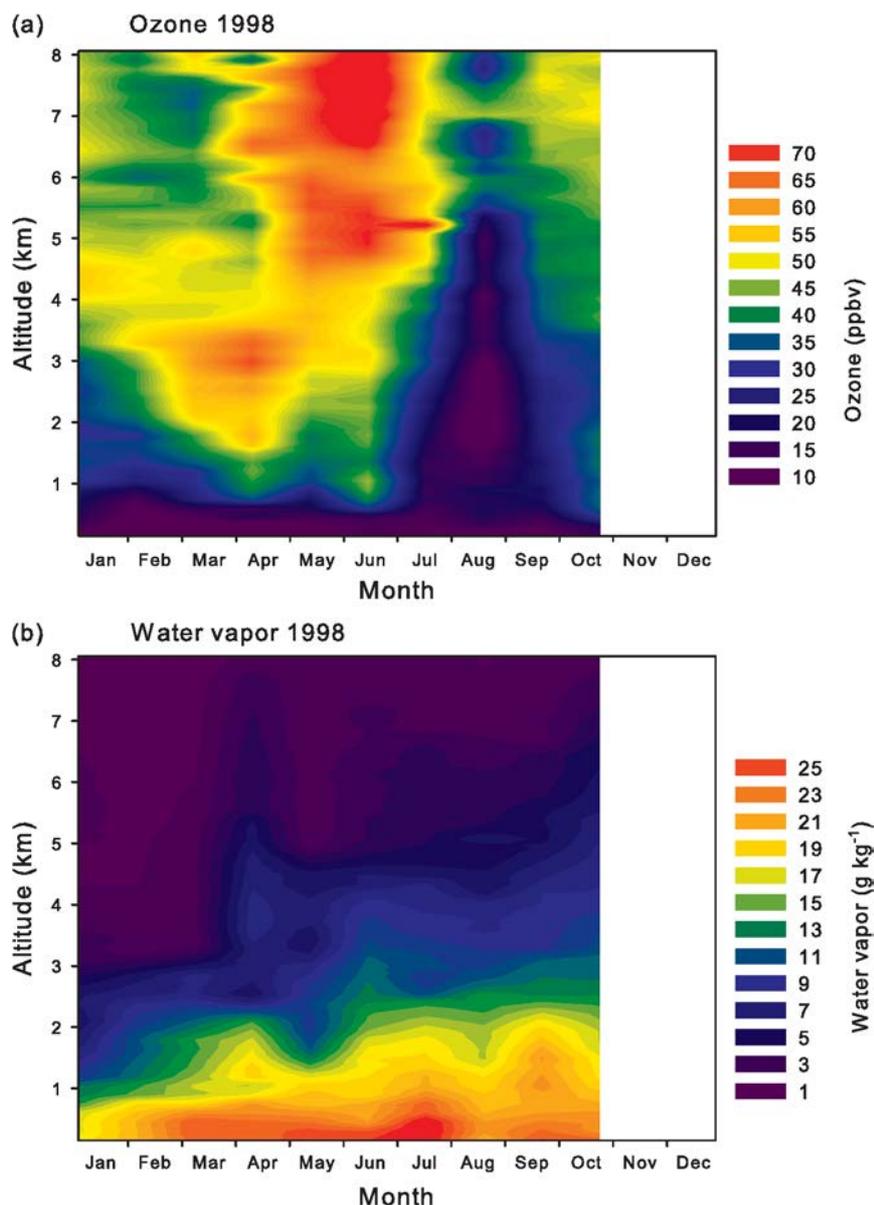


Figure 7. Monthly mean contour plots of (a) ozone and (b) water vapour representing the annual variations over Chennai during 1998. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

1999 and reference therein). Pronounced enhancements of total and tropospheric O₃ were also observed in the other parts of Southeast Asia when extensive forest fires were reported in Indonesia during 1997. The coincidences of the ozone enhancements with the forest fires suggest the photochemical production of tropospheric O₃ due to its precursors emitted from the fires (Fujiwara *et al.*, 1999). Case study discussing the impacts of Indonesian forest fires on the vertical distribution of O₃ over Chennai has been presented in Section 5.

Figure 7 shows the seasonal variations of O₃ and H₂O over Chennai for year 1998. There were no MOZAIC measurements during November and December months, otherwise the seasonal variability in O₃ during 1998 data is similar to that for year 1996. However, vertical extent of elevated O₃ (values up to 80 ppbv) during spring season of 1998 is higher compared to same period of

1996 and 1997. Seasonal variation in H₂O concentration during 1998 is also similar to 1996 over Chennai. For the period 1996–1998, the composite seasonal variations based on monthly mean vertical profiles of O₃ and H₂O are shown in Figure 8. The general features observed in the seasonal patterns of both O₃ and H₂O are similar to those discussed for individual years. The 1-sigma (standard deviation) variation of monthly O₃ mixing ratio is shown in Figure 9. The mixing ratio of O₃ shows some small-scale anomalies in the lower and middle troposphere. The 1-sigma values of ~20 ppbv between 0 and 1 km in the winter and spring months are due to exceptionally high values during year 1997. Another anomaly of 15–20 ppbv can be observed in the middle and upper troposphere (>6.0 km) during October. We have investigated all the individual profiles using back trajectories and fire count data to know the causes of this

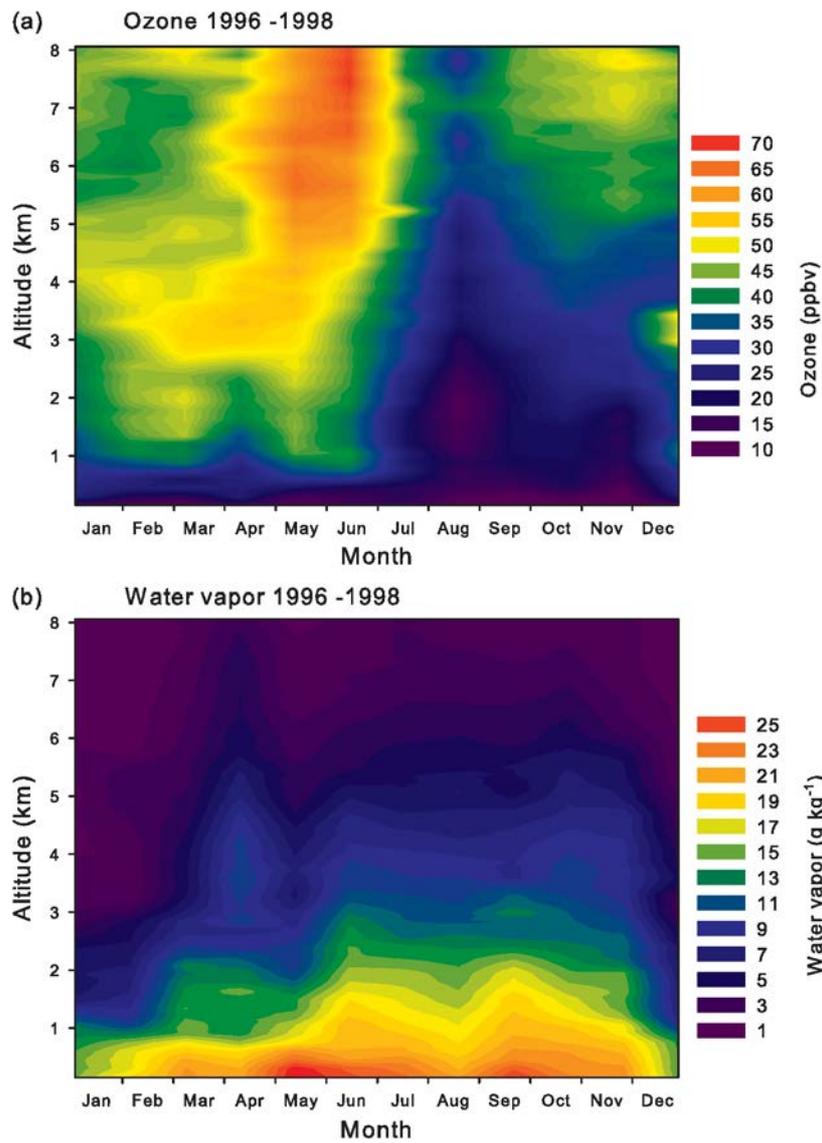


Figure 8. Monthly mean contour plots of (a) ozone and (b) water vapour representing the annual variations over Chennai during 1996–1998. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

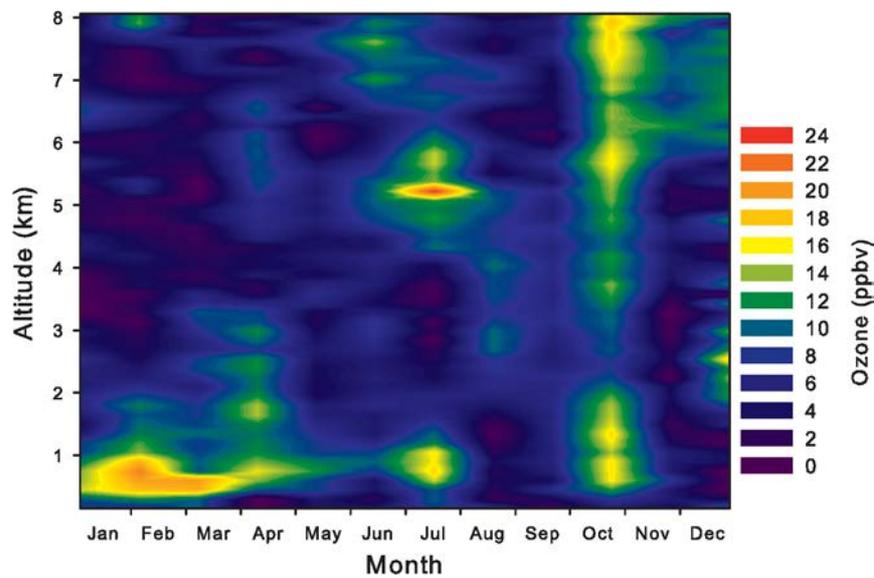


Figure 9. The 1-sigma (standard deviation) variations of monthly O₃ during 1996–1998 over Chennai. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

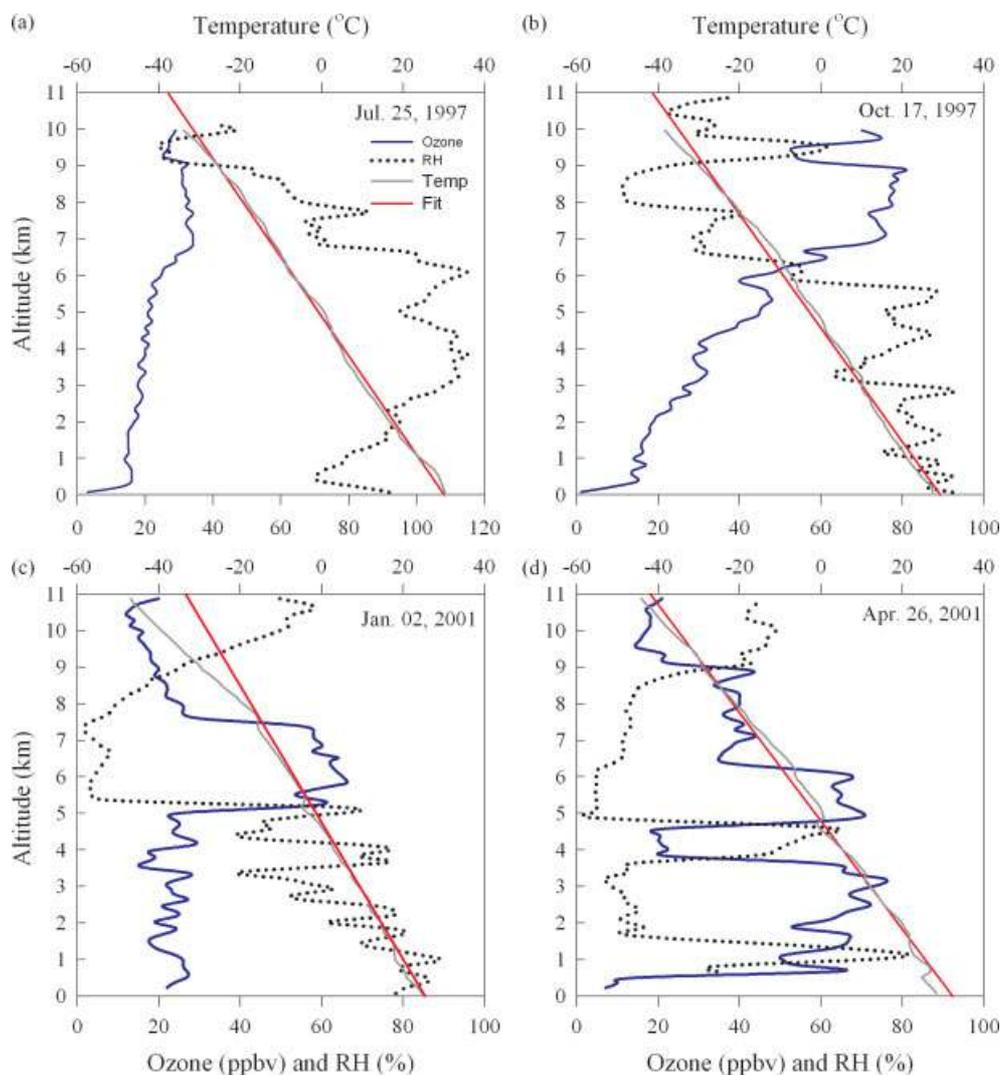


Figure 10. Vertical profiles of ozone, relative humidity, and temperature obtained from MOZAIC measurements over Chennai (a–d). This figure is available in colour online at wileyonlinelibrary.com/journal/joc

anomaly. We found that the long transport of O_3 -rich air from extensive biomass burning regions of Indonesia during year 1997 yielded this feature (case study is presented in Section 5.).

5. Case studies of O_3 profiles over Chennai

At any particular location, there can be large day-to-day variability in the vertical distributions of O_3 and meteorological parameters. The magnitude of variability can change significantly with altitude, sometimes exhibiting marked stratification, with a single or multiple peaks, and other times exhibiting a uniform profile and absence of structures.

Each O_3 profile is a unique response to photochemical and dynamic processes. Therefore, study of profiles in terms of its structure, particularly the magnitude and altitudes of maxima and minima, and vertical gradient should provide the insights about the underlying processes. Traditionally, mean profiles averaged by season or year together with some measure of variability about

these means have been analysed (Kirchhoff *et al.*, 1991; Diab *et al.*, 1996; Fortuin and Kelder, 1998; Thouret *et al.*, 1998b; Logan, 1999; Thompson *et al.*, 2003). On the other hand, case studies have been widely used to gain an understanding of the underlying atmospheric processes. Examples of studies focusing on dynamic processes, particularly STE, include Merrill *et al.* (1996), Cooper *et al.* (1998), Folkins *et al.* (1999), Ramdriambelo *et al.* (1999), Zachariasse *et al.* (2000), Scott and Cammas (2002), and Kim *et al.* (2002). Others in which strong photochemical forcing was targeted include Fujiwara *et al.* (1999) and Longo *et al.* (1999). The drawback of case study analysis is that, whilst an understanding of process is gained, there is little contribution to our knowledge of the frequency of events or their relative importance in the overall budget of O_3 .

In this study, we first describe the observed features in O_3 along with RH and temperature in the troposphere and then back trajectory analysis will be discussed to explain the features associated with the transport of different air masses. The isentropic 7-day back trajectories were

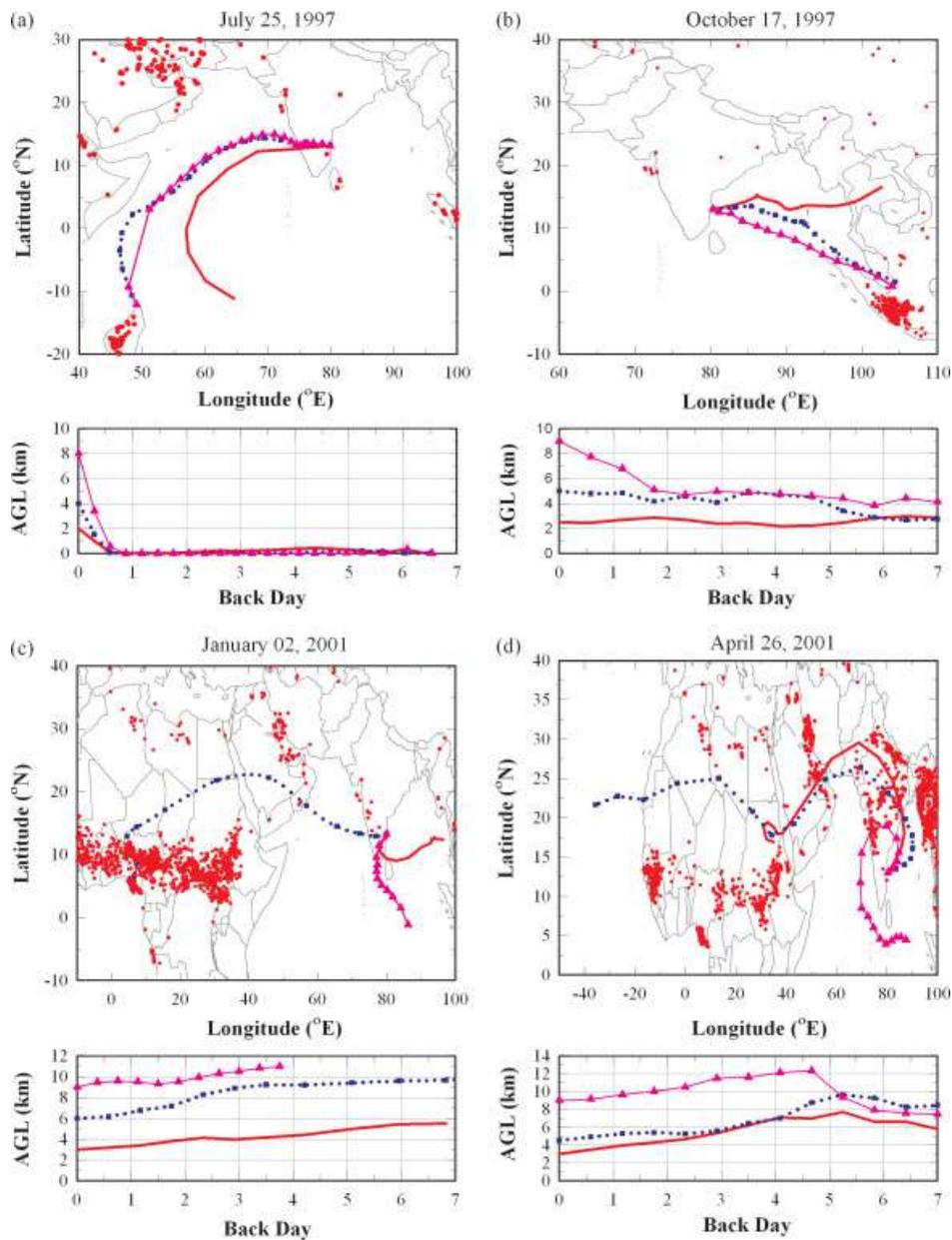


Figure 11. The 7-day back trajectory plots indicating the source and transport history of air masses at different altitudes for four selected dates of observation over Chennai (a–d). The monthly fire count data (filled circle in red) indicate the activities of biomass burning over the different continents. This figure is available in colour online at wileyonlinelibrary.com/journal/joc

calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPLOT) program (Draxler and Hess, 1998). Back trajectories represent the large-scale circulation and may be used to suggest potential source regions, although origin of a specific air parcel cannot be determined exactly. The monthly fire count data from Along Track Scanning Radiometer (ASTR) have also been shown to demonstrate the emissions activities. In the following subsection, we have chosen four different days of MOZAIC observations to represent the typical features in the different seasons over Chennai.

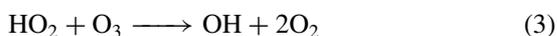
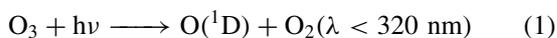
5.1. 25 July 1997

For 25 July 1997, the vertical profiles of O_3 , RH, and temperature over Chennai are shown in Figure 10(a).

This day of MOZAIC measurements have been chosen as a representative for the summer monsoon period. In the lower troposphere (0–5 km), vertical profile of O_3 shows constant but very low values of ~ 15 ppbv, while RH increased from 80 to 90% near surface to 110% in the free troposphere. The layer of very high RH in the middle troposphere indicates the presence of thick cloud due to deep convection of marine air over Chennai. In the upper troposphere, O_3 increases from 20 to 30 ppbv, while RH profile shows decreasing trend in the upper troposphere.

Such low levels of O_3 in the middle and upper troposphere are frequently observed in the tropical marine air (Fujiwara *et al.*, 2000). For this day of observation, the back trajectory plots calculated at three different altitudes

of 2, 4 and 8 km are shown in Figure 11(a). All these trajectories over Chennai were tracked to have originated in the MBL of southern Indian Ocean. Back day analysis of the trajectories suggests that the air masses were confined within the MBL and were convectively uplifted (up to 8 km) just hours before their measurements over Chennai [see lower panel of Figure 11(a)]. Owing to deep convection of marine air, the entire troposphere is well mixed resulting in very low mixing ratios of O₃ and very high RH values over Chennai for this day of observation. In the monsoon season, due to wet or rainy weather, RH is typically associated with low levels of O₃ provided by less intense photochemical production (Lelieveld and Crutzen, 1990). In addition, the chemical loss due to following set of reactions (Parrish *et al.*, 1986) could also play a major role.



Similarly, low values of O₃ mixing ratios over the Indian Ocean have been reported due to lifting of ozone-poor MBL air by convective cells of the Inter Tropical Convergence Zone (ITCZ) (De Laat *et al.*, 1999; Taupin *et al.*, 1999; Chatfield *et al.*, 2004). In support to the observations, the biomass burning activities over Indian subcontinent were also lowest during the July 1997.

5.2. 17 October 1997

For 17 October 1997, vertical profiles of O₃, RH, and temperature showing large variability over Chennai are plotted in Figure 10(b). This day of MOZAIC measurements have been chosen as a representative for the transition period from summer to winter monsoon circulations over the Indian subcontinent. The mixing ratio of O₃ is ~10 ppbv near the surface, which increases linearly with height in the lower troposphere (0–5 km), while profile of RH exhibits very high RH of ~90%. At higher altitudes, mixing ratio of O₃ increases sharply from ~45 ppbv at 5.5 km to ~82 ppbv at 7 km. On the other hand, RH shows just opposite trend as it decreases from 90% at 5.5 km to 30% at 7 km. The peak in O₃ mixing ratio of 82 ppbv coincides with a minimum in RH of 10% between 7 and 8.5 km of altitudes. However, this profile over Chennai appears to be unusual, as we have not noticed such enhancements in O₃ profiles during October and November months of other years.

Figure 11(b) shows back trajectory plots at altitudes 2.5, 5 and 9 km over Chennai for this day of measurements. The back trajectory arriving at 2.5 km of altitude originated over Thailand about 5 days before the observations. Subsequently, the air mass travelled through the MBL (which is a sink of O₃) of Bay of Bengal before reaching over Chennai. On the other hand, the sources of back trajectories calculated at 5 and 9 km of altitudes were located in the lower troposphere over Indonesia. It

has been well known that during the October–December months of year 1997, there were extensive forest fire activities over Indonesia as also shown in Figure 11(b). Therefore, transport of O₃ and its precursors from Indonesia could have impacted the measurements over Chennai. The year of 1997 corresponds to El Niño events when convective activity becomes low in Indonesia. In this region, it is likely that pronounced enhancements of tropospheric O₃ associated with extensive forest fires due to sparse precipitation may take place (Fujiwara *et al.*, 1999). Chan *et al.* (2003) have reported the substantial enhancements in O₃ mixing ratio over many parts of tropical Southeast Asia and the subtropical south China region including Hong Kong in mid-autumn and early winter (October–December) of 1997, when many forest fires occurred in the Indonesian region. Therefore, O₃ enhancements were caused by the large-scale outflows of O₃ and other pollutants from the fire active regions of Indonesia, which were related to the abnormal meteorological conditions associated with the strong El Niño phenomenon. The spread of O₃ plume, extended as far as India, was also detected by satellites observation of the Earth Probe-Total Ozone Mapping Spectrometer (EP/TOMS) during intense fire (August–November 1997) over Indonesia (Thompson *et al.*, 2001). The global importance of emissions from Indonesian peat fires is well established (Herman *et al.*, 1997; Thompson *et al.*, 2001; Cochrane, 2003)

5.3. 02 January 2001

For 02 January 2001 of measurements, the vertical profiles of O₃, RH, and temperature over Chennai are shown in Figure 10(c). This day of MOZAIC profiles have been chosen to represent the winter season measurements over Chennai. In the lower troposphere (0–5 km), mixing ratio of O₃ fluctuates between 20 and 30 ppbv, while corresponding RH profile shows decreasing trend with fluctuations between 40 and 90%. Interestingly, we can see several small-scale stratified layers in the profiles of both O₃ and RH, which are just opposite in phase. In the middle troposphere, a layer (5–7.5 km) of enhanced O₃ (~40 ppbv) was observed when mixing ratio increased from ~25 to 65 ppbv. On the other hand, RH profile exhibits just opposite variation and observed to be lowest of ~10% when O₃ mixing ratio was highest. In the upper troposphere, mixing ratios of O₃ decreases with height while RH increases as their values were ~20 ppbv and 60%, respectively at 10 km of altitude. Newell *et al.* (1999) found that approximately 20% of the troposphere is stratified and that layers are sometimes as thin as a few hundred metres (Thouret *et al.*, 2000).

To investigate the factors causing stratifications in both profiles of O₃ and RH, the back trajectories calculated at the three different altitudes of 3, 6 and 9 km over Chennai are plotted in Figure 11(c). Air parcels arriving at 3 and 9 km of altitudes were originated in the free troposphere over the oceanic regions of Indian Ocean and Bay of Bengal and do not show any influence of continental

air mass. In the tropics, maritime free tropospheric air mass is characterized by low O_3 and high RH due to lifting of ozone-poor MBL air by convective cells of the ITCZ (De Laat *et al.*, 1999; Taupin *et al.*, 1999). Further, additional depletion of O_3 in clouds might have also caused the observed low and high RH in their respective profiles (Kley *et al.*, 1996). On the other hand, trajectory arriving at 6 km of altitude shows continental influence as it originated over extensive biomass burning regions of west and central Africa (Ghana and Nigeria). The combination of high O_3 and low RH might be due to convectively lifted CBL air that has subsided over Chennai (Zachariasse *et al.*, 2000). January is the month characterized by the more intense fire activity in west central Africa, where emissions of pollutants including the precursors of O_3 are large (Eva and Lambin, 2000).

A field campaign named *EXPERiment for REgional Sources and Sinks of Oxidants* (EXPRESSO) was designed to investigate the O_3 formation in the tropical troposphere above central Africa and to consider the impact of biomass burning processes on the global atmosphere in the November month 1996 (Marion *et al.*, 2001). During this campaign period, which corresponds to an intense burning activity, the ozone production efficiency (OPE) ranging from 6.3 to 14.8 was estimated in the plumes. Satellite observations have shown high levels of tropospheric O_3 over large areas above the western African continent (Fishman *et al.*, 1990).

5.4. 26 April 2001

Vertical profiles of O_3 , RH, and temperature measured on 26 April 2001 have been shown in Figure 10(d). Typical for the distribution in spring season, profiles of both O_3 and RH show large variations resembling laminar structure over Chennai. The mixing ratio of O_3 is below 10 ppbv, while RH varies from 40 to 80% in the boundary layer. Above the boundary layer, the mixing ratio of O_3 increases rapidly to 50–75 ppbv and remain elevated up to 3.5 km of altitude, while reverse to O_3 profile RH value is down to $\sim 10\%$ in this layer. In the mid-troposphere, between 3.5 and 4.5 km of altitudes, O_3 decreases to 20 ppbv and RH increases to 70% simultaneously. Similar to layer observed just above the boundary layer, mixing ratio of O_3 shoots to the elevated values of 60–70 ppbv while RH becomes extremely low (below 5%) between 5 and 7 km of altitudes. A layer with O_3 of about 35 ppbv and RH of 15% was observed between 7 and 9 km of altitudes. In the upper troposphere, mixing ratio of O_3 is ~ 20 ppbv and RH is relatively high of 40% between 9 and 11 km of altitudes.

We have calculated the back trajectories at various altitudes for this day of measurements; however, trajectories only at 3.0, 4.5 and 8 km of altitudes are shown in Figure 11(d). In previous case study, we have explained that the enhancements in O_3 mixing ratio in the free troposphere over Chennai are mainly due to the long-range transport of fire plumes from Africa. However, in the present case study the observed low mixing ratio of O_3

and high RH near the surface is due to strong temperature inversion (Figure 11(d)), hence O_3 -rich free tropospheric air does not enter into the boundary layer. The trajectory at 3 km over Chennai, which corresponds to the elevated O_3 of ~ 80 ppbv, was originated over the central Africa and passed through Arabian Peninsula and northwest India. April is the end of fire season over central Africa (Eva and Lambin, 2000); however, apart from biomass burning other anthropogenic pollutants could be transported from continents of India and Arabian Peninsula. Diab *et al.* (2003) have also reported that the forest fire emissions over the central Africa are responsible for the low to mid-tropospheric enhancements in the spring season. Trajectory at 4.5 km originated in the upper troposphere over the Atlantic Ocean and passes through the marine region of Bay of Bengal, resulting in low O_3 and high RH over Chennai. This could have resulted in the dilution of a high O_3 layer as there are two layers with similar levels of O_3 and RH lying just below and above to this layer of lower O_3 mixing ratio. Trajectories arriving in the upper troposphere at 9 km and higher altitudes were originated over the southeast of Indian Ocean and could have transported the O_3 -poor and wet air masses over Chennai. While moderately low O_3 of 35 ppbv and high RH (40%) at 8 km was due to the transport of mixed air from Arabian Sea and continental region of central India. This transport could be due to the presence of easterly jet, which is characterized by O_3 -poor and wet layer at 9–14 km over the tropics (Fujiwara *et al.*, 2003).

6. Conclusions

Climatology of tropospheric ozone and water vapour over a metro city of Chennai in southern India has been presented based on the MOZAIC data during 1996–2001. We have also used the NCEP wind data and HYSPLIT back trajectory calculation to explain the transport and origin of various air masses. Chennai is a coastal city where NE monsoon period from October to December is the principal rainy season, whereas the SW monsoon from June to September is the secondary rainy season. The vertical distributions of both O_3 and H_2O show large seasonal variations for each year of measurements from 1996 to 1998. In the late winter and spring months, the observed high values of 60–80 ppbv of O_3 mixing ratio in the middle troposphere over Chennai were attributed to the long-range transport of pollutants from continents of Africa and Arabian Peninsula by the northwesterly winds. On the other hand, low O_3 mixing ratios of ~ 15 ppbv were observed during the JAS months, which coincide with the summer monsoon circulation over Indian subcontinent. On the other hand, monthly rainfall and vertical distributions of H_2O show just opposite seasonal variations to O_3 mixing ratio. The onset of summer monsoon circulation associated with rainfall and convective clouds coincides with the decline in mixing ratio of O_3 . While from August to December, the monthly rainfall data exhibit increasing trend, but O_3 was almost

constant, this insensitivity of O₃ mixing ratio was due to the balance between the transport of pollutants and photochemical sink of O₃. For year 1997, rather elevated but increasing trend in O₃ mixing during the early winter months seems to be an unusual feature as we do not see such trends for both the years of 1996 and 1998 of MOZAIC measurements over Chennai.

We have also selected some MOZAIC measurements for case studies of O₃, RH and temperature to investigate the impacts of long-range transport and special climatologically events. As a representative for the summer monsoon measurements, the profiles on 25 July 1997 exhibit low O₃ values of ~15 ppbv and high RH in the troposphere due to the deep convection of marine air over Chennai. The measurements on 17 October 1997 have been chosen as a representative for the transition period from summer to winter monsoon circulations over the Indian subcontinent. This day of O₃ profile exhibits strong enhancements from ~45 ppbv at 5.5 km to ~82 ppbv at 7 km over Chennai due to transport from extensive forest fire region of Indonesia during year 1997, which corresponds to El Niño events. This is for the first time we report the enhancements of O₃ over India caused by the extensive fires over Indonesia. On 02 January 2001, typical for winter monsoon, the vertical profiles of O₃ and RH show several small-scale stratified layers that are just opposite in phase. Typical for the distribution in spring season, profiles on 26 April 2001 show large variations of both O₃ and RH resembling laminar structure over Chennai.

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