Seasonality of tropospheric ozone and water vapor over Delhi, India: a study based on MOZAIC measurement data

Lokesh K. Sahu • Shyam Lal • Valérie Thouret •
Herman G. Smit

Abstract Tropospheric distributions of ozone (O_3) and water vapor (H_2O) have been presented based on the Measurements of **O**Zone and water vapor by Airbus **I**n-Service Air**C**raft (MOZAIC) data over the metro and capital city of Delhi, India during 1996–2001. The vertical mixing ratios of both O_3 and H_2O show strong seasonal variations. The mixing ratios of O_3 were often below 40 ppbv near the surface and higher values were observed in the free troposphere during the seasons of winter and spring. In the free troposphere, the high mixing ratio of O_3 during the seasons of winter and spring are mainly due to the long-range transport of O_3 and its precursors associated with the westerly-northwesterly circulation. In the lower and middle troposphere, the low mixing ratios of ~ 20 – 30 ppbv observed during the months of July–September are mainly due to prevailing summer monsoon circulation over Indian subcontinent. The summer monsoon circulation, southwest (SW) wind flow, transports the O_3 -poor marine air from the Arabian Sea and Indian Ocean. The monthly averages of rainfall and mixing ratio of H_2O show opposite seasonal cycles to that of O_3 mixing ratio in the lower and middle troposphere. The change in the transport pattern also causes substantial seasonal variation in the mixing ratio of H_2O of 3–27 g/kg in the lower troposphere over Delhi. Except for some small-scale anomalies, the similar annual patterns in the mixing ratios of O_3 and H_2O are repeated during the different years of 1996–2001. The case studies based on the profiles of O_3 , relative

L. K. Sahu (✉)

Research Center for Advanced Science and Technology, University of Tokyo, 4-6-1 Komaba,
Meguro-ku, Tokyo 153-8904, Japan
e-mail: lokesh@atmos.rcast.u-tokyo.ac.jp

S. Lal

Physical Research Laboratory (PRL), Navrangpura, Ahmedabad 380 009, India

V. Thouret

Laboratoire d'Aérologie, Unité Mixte de Recherche 5560, Centre National de la Recherche
Scientifique/Université Paul Sabatier, Observatoire Midi-Pyrénées, Toulouse, France

H. G. Smit

Research Center Jülich, Institute for Chemistry of the Polluted Atmosphere (ICG-2), PO Box 1913,
52425 Jülich, Germany

humidity (RH) and temperature show distinct features of vertical distribution over Delhi. The impacts of long range transport of air mass from Africa, the Middle East, Indian Ocean and intrusions of stratospheric O₃ have also been demonstrated using the back trajectory model and remote sensing data for biomass burning and forest fire activities.

Keywords Rain · Delhi · 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. Radiatively, because of strong absorption band centered at 9.6 μm , tropospheric O₃ is a greenhouse gas contributing to the global warming (Forster and Shine 1997; Gauss et al. 2003). The photochemical reactions are dominated by the cycles of O₃ and water vapor (H₂O) in the tropical region, which receives intense solar radiation. Water vapor is also a greenhouse gas (Ramanathan et al. 1985; Wigley 1987). The photolytic dissociation of O₃ in the presence of H₂O provides the primary source of hydroxyl (OH) radical which initiates the oxidation of major trace gases like carbon dioxide (CO₂), carbon monoxide (CO) and hydrocarbons (Houghton et al. 1996).

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 0.2–0.6 Wm⁻² to the radiative forcing since 1860, compared to 1.5 Wm⁻² from the CO₂ (IPCC 1996). The tropical region is a large contributor to the global O₃ budget. Anthropogenic activities can enhance the O₃ concentration in the troposphere through emissions of various precursors from fossil fuel combustion and biomass burning (Crutzen et al. 1979, Crutzen and Andreae 1985; Logan et al. 1981; Kirchhoff 1996). Especially in the tropics, the growing economic activity is expected to have an enhanced impact on future chemistry and climate.

The tropical continental boundary layer (CBL) is an important region for photochemical production of O₃. The vertical variations of O₃ reflect the heterogeneity of this region, arising from precursor sources (biomass burning), sinks (photolysis, photochemistry), and convective processes. The combination of these factors, depending on their importance, ultimately results in distinct O₃ profiles (Chatfield and Crutzen 1984; Liu et al. 1987; Davies et al. 1998). 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 source regions.

Typically, 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 toward 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 and Costa Rica have all participated in SHADOZ (<http://croc.gsfc.nasa.gov/shadoz>). The main objective of this work is to study the tropospheric profiles of O₃ and H₂O in view of

seasonal changes in the transport pattern and rainfall using the Measurements of O₃ and water vapor by Airbus In-Service Aircraft (MOZAIC) data over Delhi (28.56°N, 77.11°E), India. This region is also strongly influenced by the location of the subtropical jet (STJ) and only in summer more frequently located equator-ward of the STJ. The case studies using the distinct profiles of O₃, RH and temperature are presented to investigate the impact of long-range transport of air masses from different continents and also the role stratospheric intrusion.

2 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 aircraft (Airbus A340's) of four participating airlines. The MOZAIC project, in operation since August 1994, is a EU (European Union) supported project for the automatic measurements of O₃ and H₂O on board AIRBUS A340 aircraft on scheduled flights. MOZAIC is sponsored by major European airlines: Air France (1 aircraft), Austrian Airlines (1 aircraft), Lufthansa (2 aircraft) and Sabena/Air France (1 aircraft).

The O₃ analyzer is a dual beam 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, b). The response time of the instrument is 4 s which translates in to a vertical resolution of 30 m. Full details of calibration procedures and mounting in the aircraft are given in Thouret et al. (1998b). Relative humidity and temperature are measured with a capacitive sensor (Humicap-H) and a platinum resistance sensor (PT100), respectively (Helten et al. 1998, 1999). 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. MOZAIC vertical profiles of O₃, H₂O and meteorological parameters over the Indian subcontinent were measured at three airports of metropolitan cities, namely Chennai (Madras) (12.99°N, 80.15°E), Delhi and Mumbai (Bombay) (19.09°N, 72.87°E). In this study we have discussed the distributions of O₃ and H₂O over Delhi. There were about 155 MOZAIC measurements during 1996–2001, the number of profiles for different months are shown in Fig. 1(a).

For the comparison, the profiles of tropospheric O₃ obtained from MOZAIC aircraft (August 15, 1996) and by the IMD balloon-sonde (August 14, 1996) measured within 24 h over Delhi are plotted in Fig. 1(b). Though, these profiles were made during two different dates, however, these are the closest data set which we managed to get for the inter-comparison. The IMD data, measured using electrochemical cell (ECC), overestimates the mixing ratio of O₃, however mostly in the middle and upper troposphere. On the other hand, the vertical trends show reasonable agreement between the IMD and MOZAIC measurements.

Fig. 1 **a** Number of MOZAIC profiles during different months of year 1996–2001 and **b** inter-comparison of MOZAIC (UV analyzer) and IMD (electro-chemical cell) ozone profiles measured over Delhi

Apparently, the small-scale differences could arise from the different vertical resolution, local variability in the mixing ratio of O_3 due to slightly different place and time over Delhi for MOZAIC and IMD measurements. A detailed inter-comparison study of the performances of different techniques of O_3 measurements are reported by Smit and Kley (1998).

3 Seasonal transport pattern over Delhi

Delhi is the capital city of India and home for over 10 million residents. The city has large number of industries and also surrounded by various industrial zones. The total number of vehicles has increased from about 3.03 million to 4.0 million during 1997–2004 (Ghude et al. 2008). Also referred to as National Capital Region (NCR) of Delhi, it incorporates satellite cities of Faridabad, Gurgaon, Ghaziabad and Noida. Gurgaon is the main city of the

NCR because it is the home to major companies and provides the best infrastructure. Gurgaon is located in the southeast of Delhi airport and the city's population is around 1,000,000 (as per the 2001 census). Among the major industries in Gurgaon are related to information technology (IT), auto manufacturing and pharmaceuticals. Faridabad is one of the main industrial cities of Haryana state and is bounded by Delhi on the north, Gurgaon on the west and the parts of Uttar Pradesh on the east and south.

The surface temperature varies from a maximum of 45°C in summer to a minimum of 1°C in winter. Except for the summer monsoon period, the dry climate persists during most of the year. The daily rainfall data measured by the Indian Meteorological Department (IMD) for different years of 1996–2001 are plotted in Fig. 2. The months of March to June are usually hot and dry, summer monsoon prevails during June–September and winter from November to February over Delhi.

In the lower and middle troposphere, the seasonal change in the pattern of wind flow 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 in the lower levels and strong upper level divergence aloft

Fig. 2 Annual patterns of daily rainfall for different years of 1996–2001 at Delhi (Data taken from Indian Meteorological Department (IMD))

over the Indian sub-continent. The strong surface heating during summer can enhance the convective activities over the land, which is also fueled by moist inflow at low levels. We have used the National Center for Environmental Prediction (NCEP) data to derive the seasonal wind streamlines at different pressure levels flow over India and surrounding regions. Figure 3 shows the mean wind streamlines at four different pressure levels of 925 mb, 700 mb, 400 mb, and 250 mb in the months of winter for 1996–2001. In the winter season, the wind flow at 700 mb is from the west-northwest directions over Delhi. The upper troposphere is influenced by the strong westerly winds during the months of JFM (see the plot for 250 mb level in Fig. 3). On the other hand, during the summer monsoon season (JAS), the mean wind streamlines at 925 mb level (see Fig. 4) shows SW flow. Similar to the flow during winter, the westerly wind

Fig. 3 Average wind fields at four different pressure levels during winter months (January, February and March) of 1996–2001 over continental and surrounding marine regions of India (* symbol shows the location of Delhi). Color bar represents the magnitude of wind speed

prevails in the upper troposphere (250 mb) during the summer season. In the middle and upper troposphere, the westerly wind prevails throughout the year over Delhi.

4 Seasonal distributions of O₃ and H₂O

4.1 Monthly variations of O₃, H₂O and wind flow

The monthly mean vertical profiles of O₃, H₂O and wind speed over Delhi for the year 2000 are shown in Fig. 5. We have selected year 2000 as the MOZAIC measurements were made during all the months. For other years there were some gaps for a few months. The

Fig. 4 Average wind fields at four different pressure levels during summer monsoon (July, August and September) of 1996–2001 over continental and surrounding marine regions of India. Color bar represents the magnitude of wind speed

Fig. 5 Monthly mean contour plots representing the annual variations of ozone water vapor and wind speed over Delhi during year 2000

surface level mixing ratio of O_3 varies between 20–40 ppbv with a minimum during wet and a maximum value in the dry season. In the lower and mid-troposphere (2–5 km of altitudes), the mixing ratios of O_3 vary between 20–70 ppbv during the winter and spring. In the upper troposphere (6–10 km of altitudes), the mixing ratios of O_3 were observed to be about 50–120 ppbv during winter and spring.

On the other hand, the mixing ratios of O_3 were 20–40 ppbv in the lower troposphere and 45–60 ppbv in the upper troposphere during the summer season. Later, in October the mixing ratio of O_3 increases and were observed between 50–60 ppbv and 60–80 ppbv in the middle and upper troposphere, respectively.

The vertical distribution of H_2O also exhibits strong seasonal variation over Delhi, however, the cycle is opposite to that of O_3 mixing ratio (Fig. 5b). The surface level concentration of H_2O was ~6–9 g/kg during winter and spring. The higher concentrations of ~24–27 g/kg were observed in August. The decreasing trend begins toward the end of summer monsoon and lowest H_2O values of 3–6 g/kg were observed during the months of November and December. The free troposphere region was very dry as H_2O was often below 2 g/kg during winter and spring. Higher concentrations were observed in the lower and middle troposphere during the summer coinciding with the period of substantial rainfall at Delhi.

Delhi is a metro city of India where anthropogenic emissions are continuous throughout the year. The diurnal variation of surface level O_3 exhibits elevated mixing ratio of 40–65 ppbv during the daytime and lower values of 10–15 ppbv in the nighttime (Ghude et al. 2008 and references therein). The large seasonal variation (maximum in spring) observed in the mixing ratio of O_3 in the upper troposphere over subtropical regions could be mainly due to the STE (Thouret et al. 1998b; Gupta et al. 2007). The surface level mixing ratio of O_3 in the springtime over Asia is strongly influenced by the continental outflow of precursors and also by the downward transport of O_3 -rich air from the stratosphere (Hastenrath 1985; Carmichael et al. 1998; Lal et al. 2007). The annual variation of mean wind speed is shown in Fig. 5(c), and the pattern almost resembles the seasonal mixing ratio of O_3 for the year 2000. The middle to upper tropospheric region (5–10 km of altitudes) is influenced by strong winds ($>20 \text{ m s}^{-1}$) during the winter. In the summer season, this westerly flow is broken by the onset of summer monsoon circulation over Indian subcontinent. Generally, the occurrences of high mixing ratios of O_3 in the upper troposphere follow the propagation of dry air which indicates that O_3 is mainly of stratospheric origin rather than from photochemical production over Delhi. A recent study of vertical distributions over Kanpur in the north India highlights the role of downward transport of stratospheric O_3 associated with the STJ in the upper and middle troposphere (Gupta et al. 2007). In the summer season, the lower mixing ratios of O_3 observed over Delhi are mainly attributed to the flow of cleaner air due to prevailing summer monsoon circulation. The wind stream lines at 925 mb level show the influence of southwest flow carrying cleaner air from Arabian Sea and Indian Ocean. In the tropical region, convective processes over the continental region can uplift O_3 -poor air from the planetary boundary layer (PBL). In the summer season, these processes seem to contribute relatively high H_2O and low O_3 concentrations in the middle and upper troposphere over Delhi. The models and satellite observations also suggest similar distributions of O_3 over northern India (Kunhikrishnan et al. 2006; Worden et al. 2009).

Based on the MOZAIC observations for the period of 1996–2001, the composite seasonal variations using the monthly mean vertical profiles of O_3 and H_2O are shown in Fig. 6. The general features observed in the seasonal patterns of both O_3 and H_2O are similar to those discussed for the year 2000. The 1-sigma (standard deviation) variations of

Fig. 6 Monthly mean contour plots representing the composite annual variations of ozone and water vapor over Delhi during 1996–2001

the monthly O_3 and H_2O for 1996–2001 are shown in Fig. 7. The mixing ratio of O_3 shows some small scale anomalies in the free troposphere. For example, the 1-sigma values of ~ 30 ppbv between 8–9 km in the seasons of winter and spring are due to exceptionally high values of ~ 120 ppbv during the year 2000, while there are no such anomalies in the distribution of H_2O over Delhi. This indicates the large natural variability in the transport of O_3 -rich dry air from the stratosphere due to the STJ. Another anomaly of 20–30 ppbv can be noticed in the mid troposphere (5–7 km) during the months of May–July. We have investigated all the individual profiles using back trajectories and fire count data to know the causes of this anomaly. This feature can be attributed to the long-range transport of O_3 -rich air from biomass burning regions in Africa during the year 1999 (case study is presented in Section 5.0). The two major anomalies in H_2O distribution of up to 5 g/kg can be seen during the months of May–June and September–October, these were mainly associated with the onset and withdrawal of summer monsoon circulation, respectively. Similar anomalies can also be deduced from the daily rainfall data (see Fig. 2).

Fig. 7 The 1-sigma (standard deviation) variations of monthly O₃ and H₂O during 1996–2001 over Delhi

4.2 Relationship between O₃ and rainfall

To investigate how the seasonality in the mixing ratio of O₃ is related with rainfall and concentration of H₂O, the tropospheric means (between 0–5 and 5–10 km of altitudes) over Delhi for the period of 1996–2001 are plotted in Fig. 8. The annual variation of O₃ show just opposite cycles to rainfall and H₂O concentration in both the regions of lower and upper troposphere. However, the seasonality of both O₃ and H₂O are particularly stronger in the lower troposphere. The mixing ratios of O₃ were elevated in both the regions during winter and spring. The onset of summer monsoon circulation (marine air), usually associated with heavy rainfall and convective clouds, marks the decline in the mixing ratio of O₃. In the lower troposphere, the mixing ratio of O₃ was lowest of ~25 ppbv in August which coincides with the highest rainfall and H₂O concentration (17 g/kg) over Delhi. Later, during the months of September to December the mixing ratio of O₃ increases to ~50 ppbv.

In the monsoon season, due to cloudy and rainy weather, the observed high levels of RH is typically associated with the low mixing ratios of O₃ provided by the less intense in-situ

Fig. 8 Monthly mean tropospheric (0–5 and 5–10 km of altitudes) ozone, water vapor, and rainfall per month over Delhi for year 1996–2001

photochemical production (Lelieveld and Crutzen 1990). The depletion of O_3 due to the reactions with radical in the clouds could be another factor in the observed low mixing ratios (Kley et al. 1996). The importance of any process, whether simple dilution or presence of additional sinks mainly due to high H_2O need further investigation. As discussed in Section 3.0, both NE and NW winds prevailed during the dry season can transport O_3 -rich continental air from the local sources and also from the continents of Africa and Middle East particularly in the lower and middle troposphere over Delhi. On the other hand, the transport of O_3 -rich dry air from stratosphere influences the upper troposphere during winter and spring.

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) have shown the role of deep convection associated with the summer monsoon leading to low O_3 and high H_2O mixing ratios in the upper troposphere over Asia (Gettelman et al. 2004).

4.3 Average seasonal profiles over Delhi during 1996–2001

The mean vertical profiles of O_3 , RH and temperature calculated for the different seasons of year 1996–2001 are shown in Fig. 9(a–c). In the months of JFM, the mixing ratio of O_3 increases with height in the boundary layer and exhibits little variability at around 50 ppbv in the free troposphere. On the other hand, the mixing ratio of O_3 varies between 50 ppbv to 60 ppbv in the boundary layer during the spring and early summer (AMJ) and increases linearly with a rate of about 4 ppbv/km in the free troposphere. The mixing ratio of O_3 increases very slowly after attaining high values of 70–75 ppbv between 7 and 10 km of altitudes. During the summer monsoon (JAS), the mixing ratio of O_3 increases rapidly from less than 10 ppbv to 30 ppbv in the boundary layer. In the early winter, the mixing ratio of O_3 decreases in the lower troposphere and increases slowly in the mid and upper troposphere. The vertical gradients in O_3 (increasing) and RH (decreasing) are significant only up to 7 km of altitude. The gradients in the vertical profiles of temperature do not show significant variation but their levels vary with the season over Delhi. As expected, the ambient temperature is highest in summer, while lowest during winter season with a difference of $\sim 10^\circ\text{C}$ throughout the troposphere.

Fig. 9 Mean vertical profiles (1996–2001, MOZAIC data) of **a** ozone, **b** relative humidity and **c** temperature grouped in different months of the year over Delhi

4.4 Horizontal distribution of O₃ near airport

The MOZAIC aircrafts cover a large area around Delhi airport before the landing and after take off therefore, the data can provide information on local variations of O₃ mixing ratio in the lower atmosphere. Figure 10 shows the gridded mean mixing ratios of O₃ between 0–2 km of altitudes for the dry (winter and spring) and wet seasons. Figure 10(a) reveals that the high O₃ mixing ratio generally appeared in the southeastern and southwestern areas of Delhi during winter and spring. As shown in Fig. 9 and discussed in Section 3 that the southeastern and southwestern areas are very industrialized regions of namely Faridabad and Gurgaon, respectively. Moreover, Faridabad and Gurgaon are in the downwind of Delhi urban centers during the dry season. Therefore, the observed elevated mixing ratios (above 80 ppbv) could result from the local photochemical production and also from the transport of urban polluted air. However, the mixing ratios of O₃ were not so high in the northwest region of airport which suggests that the role of local production over Gurgaon and Faridabad could be the major factors for high O₃. On the other hand MOZAIC measurements are limited to get insightful information about the local distributions during summer, in any case the mixing ratios of O₃ were generally below 40 ppbv over the airport

Fig. 10 Map of Delhi showing horizontal distributions of mean O₃ mixing ratio around Delhi airport between 0–2 km of altitudes during **a** dry season and **b** summer monsoon

and surrounding regions. Interestingly, the spots of exceptionally high O_3 (above 80 ppbv) can also be observed in the summer (see Fig. 10(b)), however, right over the places of intense industrial activities in Gurgaon. Overall, this seasonality in spatial distribution of O_3 is expected as the cities of Gurgaon and Faridabad are in the upwind of Delhi urban centers during the summer circulation hence were not affected by the transport of pollutants from Delhi city and other local activities like biomass burning. Ding et al. (2008) have reported the studies of local O_3 in the PBL near Beijing metro city using MOZAIC data for year 1995–2005. The mixing ratio of O_3 at Beijing also shows strong seasonality caused by the local pollution and long-range transport. However, the climatology of O_3 distribution exhibits opposite seasonal cycles than over Delhi as the tropospheric mixing ratio is maximum in summer and minimum in winter over Beijing.

5 Case studies of O_3 profiles over Delhi

There can be large day-to-day variability in the vertical distributions of trace constituents and meteorological parameters exhibiting stratification, multiple peaks, uniform profiles and absence of structures in the troposphere.

The observed profile of O_3 can be a unique response to photochemical and dynamic processes. Therefore, study of variability in terms of its structure, particularly the magnitude and altitudes of maxima and minima, vertical gradient should provide the insights about the underlying processes. Usually, the mean profiles averaged by season or year together with some measure of variability have been analyzed (Kirchhoff et al. 1991; Diab et al. 1996; Fortuin and Kelder 1998; Thouret et al. 1998b; Logan 1999; Thompson et al. 2003). Case studies have been widely used to gain an understanding of the underlying atmospheric processes for specific events. Studies focusing on dynamic processes, particularly the 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). While, studies in which impacts of photochemical processes were targeted include Fujiwara et al. (1999) and Longo et al. (1999).

In the present study, we first describe the observed features in the profiles of O_3 , RH and temperature in the troposphere and then the back trajectories to investigate the impact of long-range transport. The 7-day isentropic back trajectories were calculated using the Hybrid Single-Particle Lagrangian Integrated Trajectories (HYSPLIT) program (Draxler and Hess 1998).

5.1 May 28, 1999

Vertical profiles of O_3 , RH and temperature observed on May 28, 1999 over Delhi are shown in Fig. 11(a). The lower troposphere (below 5 km) seems to be well mixed as O_3 remained nearly constant at 70 ppbv after a sharp increase from 30 ppbv in PBL. On the other hand, constantly low RH values of about 15% are observed over this region. In the mid-troposphere at ~5 km of altitudes, an enhancement in O_3 mixing ratio from 70 ppbv to 130 ppbv is marked also the by decline in RH to ~5%. At higher altitudes, RH remains extremely low and constant. We can see a layer of O_3 between 5 and 7.5 km of altitudes with a peak value of 140 ppbv at 6 km. Overall, the profile looks like a superimposition of enhanced O_3 layer with a background value of 70 ppbv in the free troposphere. This layer of O_3 is isolated by a very strong inversion as the gradient in temperature is almost negligible for the altitude range corresponding to this layer.

Fig. 11 Vertical profiles of ozone, relative humidity and temperature obtained from MOZAIC measurements over Delhi

To investigate the causes of observed O₃-rich layers we have calculated back trajectories at 3, 6 and 9 km of altitudes as shown in Fig. 12(a). The monthly fire count data from Along Track Scanning Radiometer (ASTR) have also been shown to demonstrate the emission activities. The trajectories arriving below and above the O₃-rich layer (3 and 9 km of altitudes) were originated in the free troposphere over the continental regions of southern Europe and Africa and passed over the Middle East before descending over Delhi. In other words, these trajectories pass over the regions of biomass burning and anthropogenic activities hence could have transported O₃ and its precursor gases. However, the high background of ~70 ppbv could be attributed to the enhanced regional emission activities of biomass or forest fire over northern India (see Fig. 12). However, the trajectories at 6 km altitude passed through lower stratospheric O₃-rich region at ~45° N latitude over Atlantic Ocean resulting in elevated mixing ratio of O₃ over Delhi. Study of some profiles shows the influence of stratospheric originated air mass in the upper troposphere over Delhi in the spring season. The potential vorticity (PV) values of 2 km² kg⁻¹ s⁻¹ or more can be attributed to the stratospheric origin of air masses (Appenzeller and Davies 1992). The Japan Meteorological Agency (JMA) data were used to estimate the PV. Figure 13 (a) shows a contour plot of PV at 330 K potential temperature which corresponds to 8–10 km over the mid-latitude zone. Very high PV of ~12 can be seen at the origin of trajectory corresponding to the enhanced O₃ layer over Delhi. The stratospheric exchange of O₃-rich air can occur due to the filamentation around the subtropical jet (STJ). Shear-induced differential advection can cause tracer surface to stretch which leads to quasi-horizontal laminae-like tracer structure (Appenzeller and Holton 1997).

The presence of layers with O₃ and H₂O mixing ratios significantly different from the background atmosphere introduces changes in the radiative cooling and heating rates, which are strongly dependent on these greenhouse gases (Stoller et al. 1999). In one such layer between 5–7 km (see Fig. 11(a)), we noticed that the vertical gradient in temperature profile is less than the values in other regions of the troposphere. Almost constant value of about -10°C suggest heating due to elevated mixing ratio of O₃. The presence of such layers has implications in the atmospheric stability and hence can impact the vertical mixing of other trace constituents in the troposphere.

5.2 March 14, 2000

Vertical profiles of O₃, RH and temperature observed on March 14, 2000 over Delhi are shown in Fig. 11(b). The mixing ratio of O₃ increases rapidly in the boundary layer and its value is ~60 ppbv near 1 km of altitudes and RH decreases from 55% near the surface to 10% at 2 km. From the study of a set of profiles we estimated that the background mixing ratios of O₃ were 60–70 ppbv over Delhi during the month of March. In the mid-troposphere, several sharp peaks in mixing ratio of O₃ appear like superimposed in the background level. In the upper troposphere, occurrence of such peaks are rare as the mixing ratio of O₃ shows constant value of 50–55 ppbv and slightly higher RH level compared to the dry mid-troposphere. The peaks in O₃ can be observed at around 2, 4, 5 and 7 km of altitudes, however among these the one at 5 km with 115 ppbv is the most prominent. The back trajectories analyses for 3, 5.5 and 9 km of altitudes over Delhi are shown in Fig. 12 (b). The trajectories arriving in the lower mid-tropospheric altitudes over Delhi have all originated over North America and passed over the continents of Europe and the Middle East. The path of a back trajectory for O₃ peak at 5.5 km of altitude originated in the free troposphere while source regions of trajectories arriving at lower altitudes were located in the lower troposphere over the regions of mid-latitude. Thus, the transport of O₃-rich dry air

Fig. 12 The 7-day back trajectory plots indicating the source and transport history of air masses at different altitudes for five selected dates of observation over Delhi. The monthly fire counts are shown to locate the hot spots

Fig. 13 Latitude–Longitude cross section of potential vorticity (PV) in unit of $\text{km}^2 \text{kg}^{-1} \text{s}^{-1}$ at 330 K on **a** May 28, 1999 and **b** March 14, 2000

from mid latitudes could have resulted in the layers with high O_3 and low RH over Delhi. The air masses originated over the tropical Atlantic Ocean and Africa influenced the upper troposphere as we observed moderate values of O_3 and RH. Figure 13(b) shows the contour plot of PV at 330 K for this day of observation. We have selected this level because the trajectories correspond to the peaks in O_3 originated between 8 km and 10 km over the mid-

latitudes. The influence of stratospheric air can be confirmed, as the PV values were significantly higher than 2 over the regions from where the trajectories originated and yielding peaks in O₃ profile over Delhi.

5.3 Nov 29, 2000

Vertical profiles of O₃, RH and temperature observed on November 29, 2000 over Delhi are shown in Fig. 11(c). This is a distinct example in a sense that the mixing ratio of O₃ does not show any clear relations with RH. In the PBL, the mixing ratio of O₃ varies between 20 ppbv to 30 ppbv, but the RH remained elevated at 70–90%. The profile of temperature shows an inversion at ~2 km of altitude, due to this mixing barrier the RH decreases drastically from above 85% to less than 20%. On the other hand, the profile of O₃ does not show such sharp changes near the inversion. The distributions in the middle and upper troposphere exhibit fairly constant O₃ values of 30–40 ppbv, however, relatively large variability of 15–90% can be noticed in the profile of RH. In the lower troposphere, a layer with ~20% of RH and 40 ppbv of O₃ was observed between 2 and 4 km of altitudes. Another layer with 90% RH and 35 ppbv of O₃ was observed in the middle troposphere between 6 and 8 km of altitudes. With no appreciable change in the mixing ratio of O₃, the profile of RH shows increasing trend in the upper troposphere. The back trajectories at 3, 6 and 9 km of altitudes for this day of observation are shown in Fig. 12(c). The air mass originated over the southern region of Europe and passed over the Middle East, this transport pathway could have resulted in a dry layer at 3 km altitude however O₃ is relatively low over Delhi. The trajectory arriving in the middle troposphere at 6 km (layer with very high RH values) originated over the tropical Atlantic Ocean and passed over the continent of central Africa.

5.4 October 03, 2001

Vertical profiles of O₃, RH and temperature observed on October 3, 2001 over Delhi are shown in Fig. 11(d). The mixing ratio of O₃ was observed to be very low in the lower troposphere as it varies between 10–20 ppbv except a sharp peak (42 ppbv) at 3 km of altitude. On the other hand, the level of RH was very high between 70–100% in the lower troposphere. A minimum can be observed in the mixing ratio of O₃ just below 4 km of altitude, however, accompanied by RH of about 100%. The sharp changes in the profiles of both O₃ and RH were recorded simultaneously at different altitudes. As can be observed from the profiles between 4–5 km, the mixing ratio of O₃ increases from 10 ppbv to 80 ppbv and RH decreases from 75% to 5%. Such transitions were also marked by the strong inversion in the profile of temperature which could act as barrier for the vertical mixing of lower tropospheric air masses with middle and upper troposphere. The mid-tropospheric region was influenced by extremely dry air with RH of 5% and O₃-rich of 80 ppbv between altitudes of 5 and 7.5 km. In the upper troposphere, the profiles of O₃ and RH show increasing and decreasing trends, respectively. Figure 12(d) shows the back trajectories at 2, 6 and 9 km of altitudes for this day of the measurements over Delhi. In the lower troposphere, the origins of trajectories were located over the Indian Ocean and confined within the boundary layer during the transport. This flow could have transported humid marine air with low O₃ in the lower troposphere over Delhi. The trajectory analysis at 6 km suggests the origin in the upper troposphere over continental Africa and transport via the Middle East. On the other hand, the trajectory arriving in the upper troposphere at 9 km altitude originated over the Bay of Bengal and passed through the Arabian Peninsula.

Consequently, the transport of the free tropospheric tropical marine air resulted in the observed decreasing and increasing trends of O_3 and RH, respectively. Such profiles in O_3 and RH can be attributed to the advection processes without any significant role of convection. The low levels of O_3 in the middle and upper troposphere are frequently observed in the tropical marine air (Fujiwara et al. 2000). In the early winter month, the strong temperature inversion in the mid-troposphere also indicates the role of convection free dynamical process in the distributions of O_3 and H_2O over Delhi.

5.5 August 24, 2000

Vertical profiles of O_3 , RH and temperature observed on August 24, 2000 over Delhi are shown in Fig. 11(e). This day of MOZAIC measurements is taken 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 ~30 ppbv, but RH was very high ~70–80%. In the upper troposphere, O_3 increases from 30 ppbv to 60 ppbv, while RH decreases with height in the upper troposphere. Such low levels of O_3 in the lower and middle tropospheric regions are frequently observed in the tropical marine air (Fujiwara et al. 2000).

For this day of observation, the back trajectories calculated at three different altitudes of 0.5, 3 and 7 km are shown in Fig. 12(e). All these trajectories in the lower troposphere were tracked to have originated in the marine boundary layer (MBL) of southern Indian Ocean. The paths of these trajectories also suggest that the air masses were mostly within the MBL of Arabian Sea during the transport. The NCEP wind streamlines for summer season (Fig. 4) also show strong southwest wind flow from Arabian Sea. However, some enhancements in O_3 associated with low RH in the upper troposphere could be due to the transport of air from high latitudes as suggested by the trajectory at 7 km. The low mixing ratio of O_3 observed in the month of August is expected due to the negligible activities of biomass burning and also due to the transport of cleaner air over Indian subcontinent.

6 Conclusions

The seasonality in the tropospheric mixing ratios of O_3 and H_2O over Delhi, India has been presented based on the MOZAIC aircraft profile data measured during the year 1996–2001. The NCEP wind, rainfall data, HYSPLIT back trajectory calculation, and fire count data were used to investigate the role of long-range transport of various air masses from different regions of the world. Frequent rainfalls are recorded during the period of summer monsoon circulation, from the months of July to September over Delhi, and dry weather persists during rest of the months. The vertical distribution mixing ratios of both O_3 and H_2O show strong seasonal variations. The elevated mixing ratio of O_3 of 110–120 ppbv observed in the middle and upper troposphere are attributed to the long-range transport of pollutants from continents of Africa and Arabian Peninsula and intrusions of stratospheric air. On the other hand, the mixing ratios of 20–30 ppbv during the months of July–September coincide with the summer monsoon circulation over the Indian subcontinent. Overall, the monthly rainfall and vertical distributions of H_2O show just opposite seasonal variations to that of 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 .

Case studies of some selected profiles of O_3 , RH and temperature were also presented to investigate the impacts of long-range transport of air masses from different source regions. Profiles on May 28, 1999 and March 14, 2000 show layers of enhanced O_3 and dry air in

the lower and middle troposphere over Delhi. These elevated layers of O₃ appear to be superimposed in the stable backgrounds of 60–70 ppbv. The back trajectories and studies of hot spots suggest that transports of pollutants from northwest of India, Arabian Peninsula and Africa play important role in the observed elevated O₃ backgrounds in the middle troposphere. However, the long-range transport of O₃-rich air from high PV regions resulted in the enhanced mixing ratios of O₃ of 100–130 ppbv and dry air in the upper troposphere. Case studies of profiles on August 24 2000, November 29 2000 and October 3 2001 reveal the influences of oceanic air in the upper troposphere over Delhi.

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