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Influence of regional pollution and long range transport over western India: Analysis of ozonesonde data

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ABSTRACT

Four years (April 2003 – July 2007) of ozonesonde observations over Ahmedabad have been studied for the first time using ten days backward trajectories in the boundary layer (lower 2 km) and lower troposphere (2.5–4 km). Ozone data are classified according to the residence times of air-masses over the North-Western India (NWI), marine and Northern Africa/Southern Europe (NASE) regions. Ozone increases linearly with increasing residence days over the NWI region for about six days with maximum increase rate (boundary layer ~4.5 ± 1.1 ppbv/day, lower troposphere ~3.4 ± 0.8 ppbv/day) during spring and minimum during winter (boundary layer ~0.7 ± 0.8 ppbv/day, lower troposphere ~0.8 ± 0.7 ppbv/day). The analysis of surface ozone over Ahmedabad confirms that ozone increase with residence days is largely due to photochemical build up. The estimated background ozone corresponding to zeroth residence day is found to be significantly lower during summer-monsoon (~26.3 ± 3.3 ppbv) than winter (~47.7 ± 3.2 ppbv) within the boundary layer. The air masses mainly influenced by NWI region, marine and NASE regions are termed as regionally polluted, marine and long range transport (LRT) respectively. The regionally polluted ozone is found to be higher than the average ozone during spring and summer-monsoon by 22–41% within the boundary layer and by 9–12% in the lower troposphere. The marine air shows lower ozone by 38% and 10% during spring and summer seasons respectively in the boundary layer. LRT plays a significant role in the lower troposphere during spring and summer seasons with an ozone enhancement of 9% and 27% respectively. The present work suggests that regional pollution and long range transport have significant influence on the seasonal distribution of ozone in the lower troposphere whereas the background ozone levels in summer-monsoon are mainly influenced by marine air mass over this region.

1. Introduction

Ozone is an important precursor of oxidizing hydroxyl radical that decides the life time of several trace gases and plays key role in the tropospheric chemistry. At elevated levels, it is a pernicious pollutant having detrimental effects on human health and crop productivity (e.g. Ellingsen et al., 2008; Fuhrer, 2009). In addition, it is an effective greenhouse gas, which plays a crucial role in the earth's radiation budget (Gauss et al., 2003). Its photochemical production is nonlinearly dependent on the levels of precursors – nitrogen oxides (NO_x), non-methane hydrocarbons (NMHCs), carbon monoxide (CO) and methane (CH_4) through linked chemical reactions (Seinfeld and Pandis, 1998). The burden of tropospheric ozone is increasing particularly over the Asian region (Ding et al., 2008; Wang et al., 2009) due to large scale emission of these precursor gases over this region (Akimoto, 2003; Streets et al., 2006; Ohara et al., 2007).

Several studies have documented the intercontinental transport of air pollution (Huntrieser et al., 2005; Chin et al., 2007). The large scale advection can transport the pollutants to great distances, far away from their local sources. Ozone lifted to levels above the boundary layer can be transported over long distances and can contribute to the background at the downwind regions (e.g. Fiore et al., 2003; Jaffe et al., 2003; Cooper et al., 2010). From the Asian perspective, 40% of the tropospheric ozone over East Asia is shown to be influenced by European sources (Newell and Evans, 2000). The model studies suggest that European emission may contribute about 4 ppbv of ozone over Central Asia in spring season (Wild

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et al., 2004). Contribution of African/European outflow to surface ozone is found to be about 8–11 ppbv during January–March at Nainital, a high altitude site in Northern India (Kumar et al., 2010). However, the contribution of Indian pollution and long range transport on the vertical distribution of ozone over the Indian region has not been investigated so far.

The back-trajectories are found to be an important tool to interpret the ozone levels in relation to transport from source regions in several studies (e.g. Delcloo and De Backer, 2008; Davis et al., 2009). A refined trajectory based method has been introduced by Pochanart et al. (2001) in which various features of surface ozone distribution are explained on the basis of residence time of air-masses over different regions. This method is used to interpret the contributions of long range transport and regional production of ozone over Europe. This approach was used for the first time in analyzing ozonesonde data within the boundary layer and lower troposphere over Europe (Naja et al., 2003) and Japan (Naja and Akimoto, 2004). However, such study using ozonesonde data simultaneously within the boundary layer and lower troposphere is not made over the Indian region.

To delineate the contributions of various processes like boundary layer dynamics, local photochemistry, regional emissions of precursors, long range transport etc. in the distribution of ozone in the lower atmosphere up to 4 km, investigation of ozone is made for the first time using ozonesonde observations. This work presents seasonal variations of ozone within the boundary layer and in the lower troposphere over Ahmedabad. The back trajectories are used as a tool to quantify the contributions of various processes in ozone distribution.

2. Site description, observational details and general meteorology

The observational site, Physical Research Laboratory (PRL), is situated in the western part of Ahmedabad (23.03°N, 72.54°E, 49 m amsl) which is an urban city in north western India having several industries and a power plant. The observation site is located about 10–15 km away from the industrial area. The balloon flights were conducted from the rooftop of PRL from April 2003 to July 2007. The ozonesondes (EN SCI Corporation, USA) and radiosondes (model RS-80, Vaisala, Finland) were launched fortnightly in the time window of 0930 h and 1030 h IST (Indian Standard Time, IST = UT + 5.5 h) in coordination with local Air Traffic Controller (ATC). A total of 79 balloonsondes were launched each with the help of a 1.2 kg rubber balloon to obtain high resolution vertical profiles of ozone, temperature, humidity and pressure. However, there were breaks in balloon flights due to technical problems and field campaigns in between. The ascent rates of the balloons were typically 3–5 m s^{-1} and ceiling altitudes were in the range of 30–35 km. The transmitted data were received by a ground based 403 MHz receiver at every 1.2 s interval. Each ozonesonde consists of a teflon pump, an ozone sensing electrochemical concentration cell (ECC) and an electronic interface board. The ECC sensor is made up of teflon cathode and anode chambers containing platinum electrodes immersed in KI solutions of different concentrations. This sensor yields a precision better than ±(3–5)% and an accuracy of about ±(5–10)% up to 30 km altitude (Smit et al., 2007). The RS-80 Vaisala radiosonde consists of capacitance based temperature, humidity and pressure sensors having accuracies of ±0.2 °C, ±2% and ±0.5 mb respectively below 5 km (Vaisala Inc., 1989).

The measurements of surface ozone have been made over Ahmedabad using ozone analyzers based on UV absorption technique (Dasibi model RS 1008; Thermo model 49i). The full year ozone data are available for 2002 and 2009. A teflon inlet is used to suck the ambient air inside the analyzers from the open field away from the building. These analyzers have a minimum detection limit of 1 ppbv, response time of 10 s and absolute accuracy of about 5% (Kleinman et al., 1994).

The weather over the western Indian region is mostly influenced by seasonal winds. The southwest monsoon occurs from June to August (summer-monsoon) when the Inter-Tropical Convergence Zone moves towards northern India (up to 30°N). The low pressure system over the Indian region causes the inflow of moist air from the Indian Ocean and the Arabian Sea resulting into heavy rainfall. This region experiences drier northerly and north-easterly winds during December to February months (winter). Spring (March to May) and Autumn (September to November) seasons are referred to as the transition periods, when wind reversal takes place from north-easterly to south-westerly and vice versa. Ahmedabad experiences about 750 mm rainfall during summer-monsoon. The relative humidity varies from 70–80% during summer-monsoon to 20–30% during winter. The surface temperature showed maximum in May (31.6 ± 2.1 °C) and minimum in December (20.1 ± 2.3 °C) during the observational period over this location at the balloon launch time (Srivastava et al., 2010).

3. Results and discussion

3.1. Vertical distributions of ozone

Fig. 1 shows the average vertical distributions and individual ozone profiles of ozone during winter (DJF), spring (MAM), summer-monsoon (JJA) and autumn (SON) up to 8 km over Ahmedabad. Ozone levels are higher during winter below 2.5 km while these are higher during spring and autumn in the altitude range of 2.5 to 4 km. At higher altitudes (above 6 km) ozone mixing ratios are maximum during summer-monsoon. A layer of enhanced ozone is observed in the 0.5–2 km altitude range during winter which is absent during other seasons. The ozone mixing ratios are found to be minimum within the boundary layer (<40 ppbv below 2 km and 20 ppbv below 1 km) in summer-monsoon. The vertical distributions of ozone are found quite similar in spring and autumn in the lower 2 km. Minimum ozone values are seen in winter and maximum in summer-monsoon season above the boundary layer in the altitude range of 2–3 km. Therefore, it appears that different meteorological processes play important roles in the ozone distributions within and above the boundary layer. In view of this, ozone distribution in the lower 4 km is further analyzed using back trajectories utilizing spatial and temporal classification of air-masses.

3.2. Backward trajectory analysis

The backward trajectories can provide important information about the origin of air-mass arriving at the observation location and their effects on the distribution of ozone. The trajectories simulation has been done using the METeorological data EXplorer (METEX) trajectory model developed at the National Institute of Environmental Studies, Japan and Global Environmental Forum, Japan (Zeng et al., 2008). The METEX uses the NCEP/NCAR reanalysis data having spatial resolution of 2.5° × 2.5° at 17 different pressure levels archived at every six hours. This trajectory model has been used in several research publications (Naja et al., 2003; Naja and Akimoto, 2004; Kumar et al., 2010).

The spatial distributions of ten days isentropic backward trajectories are investigated over Ahmedabad at different altitudes. These observations are used to classify preliminary sectors which might be affecting the observations over Ahmedabad. The air masses of three different regions influence this site during

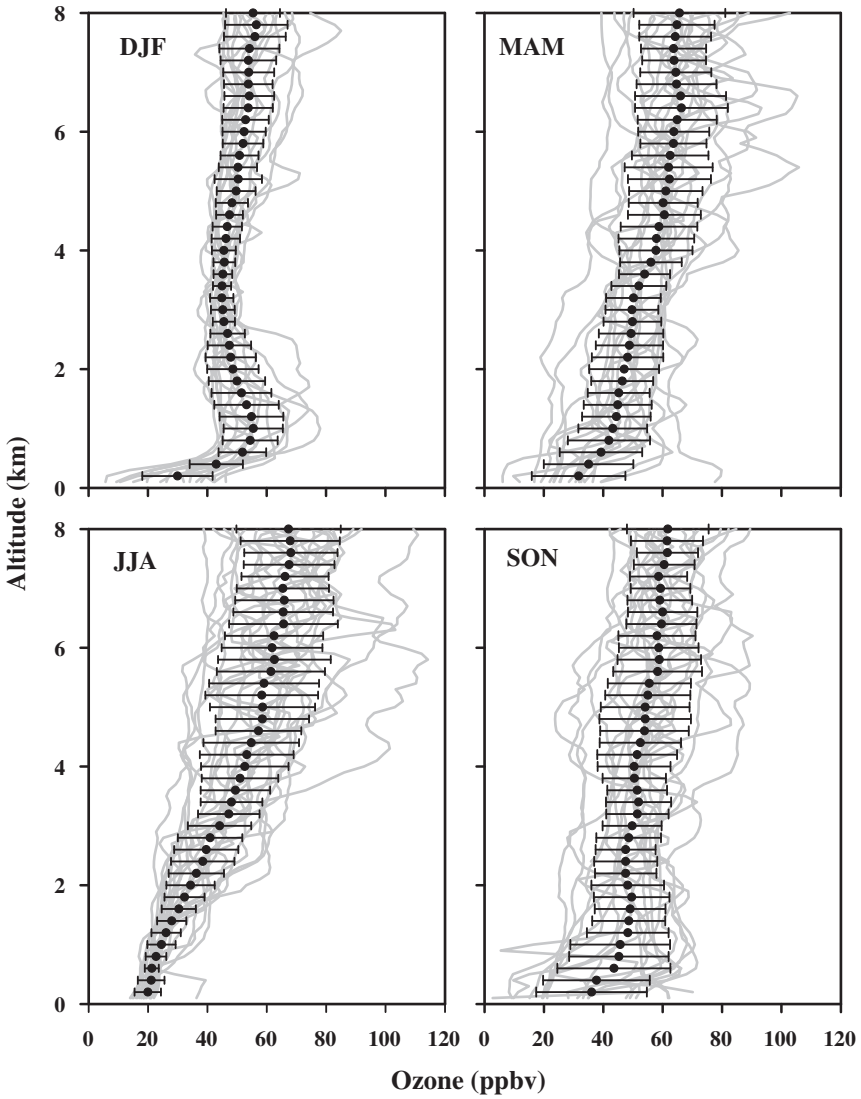

Fig. 1. Seasonal variation of vertical distribution of average ozone up to 8 km in different seasons namely Winter (DJF), Spring (MAM), Summer-Monsoon (JJA) and Autumn (SON). The horizontal bars are $\pm$ 1σ variation. Individual profiles used for the averaging are also shown.

different months (Fig. 2). These regions are defined to classify the trajectories:

(1) The region from 18°N to 30°N and from 65°E to 80°E is designated as North-Western Indian (NWI) region.
(2) The Arabian Sea, Bay of Bengal and Indian Ocean are designated as the marine regions.
(3) The region west of 65°E and excluding marine region is defined as Northern Africa/Southern Europe (NASE) region (trajectories originate mostly from northern Africa and southern Europe).

NWI region is the region of large anthropogenic emissions including vehicular and industrial emissions. The air-mass circulates over this region during late autumn and early winter in lower altitudes. The air masses reach at observation site from marine region during late spring, entire summer-monsoon and early autumn. The third sector mainly influences the air masses of higher altitude (>2 km) round the year. There are very few trajectories (<1%) from East Asian region and are not classified explicitly.

The ten days backward trajectories are simulated over Ahmedabad at seven initial altitudes (1 km, 1.5 km, 2 km, 2.5 km, 3 km, 3.5 km and 4 km) at 0930 IST (4 GMT) for each balloon flight day. In addition to that, four additional trajectories are calculated at each mentioned altitude at the corners of the 0.5° × 0.5° grid made surrounding the observation site (A total of 7 × 5 trajectories per ozonesonde flight). These additional trajectories are used to filter out non coherent trajectories as per the criteria given by Naja et al., (2003). The average residence time (number of hours spent) of trajectories are calculated for each day and seven predefined altitudes over three defined sectors i.e., NWI, marine and NASE regions. To check the performance and sensitivity, the residence times of trajectories from METEX and HYSPLIT trajectory models are compared for different seasons over three different regions and the difference is found to be less than 2% when the air parcel has spent significant time (>3 days) over a particular region.

The average mixing ratios of ozone are calculated at these seven altitudes for each balloon flight day. The ozone mixing ratio corresponding to 1 km is calculated by averaging its values between 0.75 km and 1.25 km (±0.25 km). This procedure is followed for 1.5 km, 2.0 km, 2.5 km, 3.0 km, 3.5 km and 4.0 km. The ozone values below 0.75 km region are not considered in the present analysis to avoid the effects of immediate local emission of

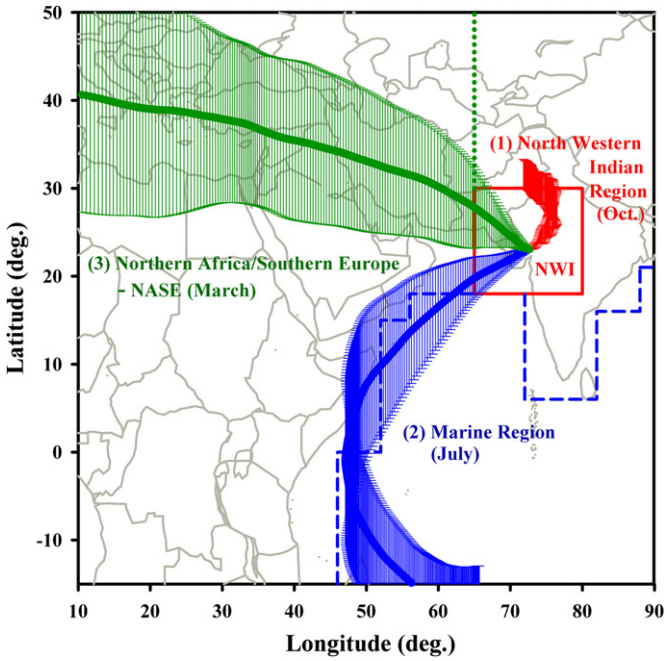

Fig. 2. The spatial coverage of 10 days back ward trajectories originated from (1) North Western Indian (NWI) region, (2) marine region, and (3) Northern Africa/Southern Europe (NASE) region. The vertical bars represent ± 1σ variation.

precursors of ozone, dry deposition and variation in balloon launch times during entire study period. For further analysis, these ozone data and corresponding residence times in seven layers are regrouped in the two regions i.e., boundary layer (1 km, 1.5 km and 2 km) and the lower troposphere (2.5 km, 3 km, 3.5 km and 4 km).

3.3. Dependence of ozone on residence time over north-western India

In order to understand the influence of different air masses having varying residence times over three defined geographical regions, mixing ratios of ozone and corresponding residence times are analyzed. The effects of Indian regional pollution are examined by investigating the dependence of average ozone on corresponding residence time over NWI region. Data of cloudy days and events of long range transport are not considered in this analysis. About 6% data in the boundary layer and 11% data in the lower troposphere have been filtered out accordingly. Figs. 3 and 4 show the dependence of ozone mixing ratios on the residence time over NWI region for different seasons (DJF, MAM, JJA and SON) in the boundary layer and in the lower troposphere respectively. The residence time is assigned 1 day if air-mass has spent less than 1.5 days, 2 days if it has spent between 1.5 days to 2.5 days and so on for the other days. Further, average ozone values in the boundary layer and lower troposphere during each sounding have been marked with the corresponding residence times of air-masses.

A correlation between average ozone values and corresponding residence times show that the ozone values increase with increasing residence time up to 3–6 days mainly in spring. Such increasing tendency can be seen in summer-monsoon and autumn. The mixing ratios of ozone increase with a rate of 4.5 ± 1.1 ppbv/day and 3.4 ± 0.8 ppbv/day (from 1 day to 6 day) during spring in the boundary layer and lower troposphere respectively. During summer-monsoon, the boundary layer air mass spends several days over the marine region before arriving at Ahmedabad, therefore data are not available for more than 4 residence days in the boundary layer region (Fig. 3). Ozone mixing ratios show increase with residence time by 3.2 ± 0.6 ppbv/day in the lower troposphere during this season. This observed growth in ozone with increasing residence time is due to the accumulation of ozone and its precursors in the air parcel over this region during the residence period. The availability of sufficient

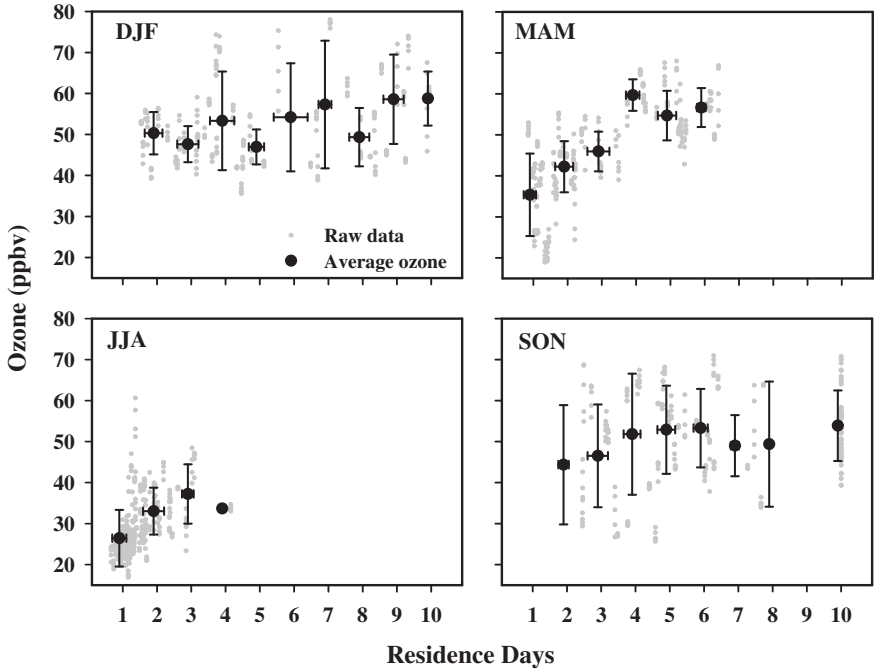

Fig. 3. Variation of ozone mixing ratios with residence days in the boundary layer during winter (DJF), spring (MAM), summer-monsoon (JJA) and autumn (SON) over regionally polluted sector. The vertical bars show ±1σ variation in ozone mixing ratio and horizontal bars show ±1σ variation in residence time of corresponding air masses used in the averaging.

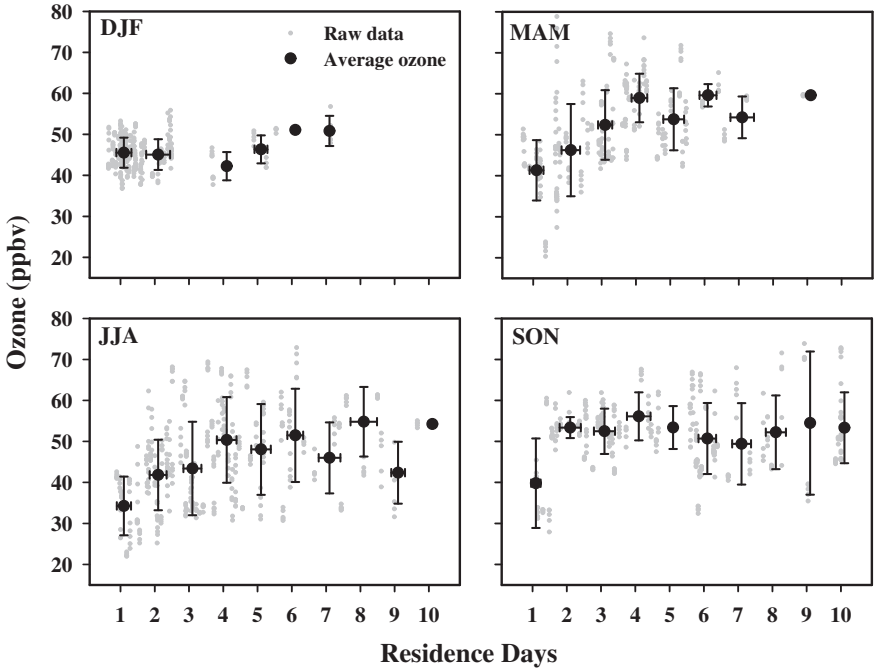

Fig. 4. Variation of ozone mixing ratios with residence days in the lower troposphere during winter (DJF), spring (MAM), summer-monsoon (JJA) and autumn (SON) over regionally polluted sector. The vertical bars show $\pm 1\sigma$ variation in ozone mixing ratio and horizontal bars show $\pm 1\sigma$ variation in residence time of corresponding air masses used in the averaging.

solar radiation induces ozone formation through the photo-oxidation of the accumulated precursors. Solar radiation is highest in spring over this region (Lal et al., 2000). A small ozone increase rate is observed during winter (about 0.7–0.8 ppbv/day in the boundary layer and lower troposphere), but a clear systematic increase is absent during this season. The accumulation of local pollution is very high due to shallow boundary layer in the winter accompanied with sufficient solar flux during this season. Thus, the average ozone values are already high (~50 ppbv). Mixing ratios of NO are also reported to be maximum during this season at this site (Lal et al., 2000). Simultaneous photochemical production and titration with NO (during the extended period of more than 2 days) may be the possible causes for the absence of ozone build up in winter.

Generally, it is observed that the ozone mixing ratio achieves its saturation levels after about 5–6 days during spring and autumn seasons within the boundary layer. Its mixing ratio remains almost constant or some time decreases with increasing residence days further (Pochanart et al., 2001; Naja and Akimoto, 2004). This decreasing feature after 6 days may arise due to unfavourable weather conditions (like clouds and rain, lesser amount of solar radiation etc) for the photochemical production of ozone over the NWI region. The other possible limiting phenomenon for further enhancement in ozone levels in air parcel of extended stay may be the titration of ozone by high level of NO over this region. Therefore, it is observed that generally air-masses which have spent 3–6 days over this region are found to be sufficiently photoxidised and can be taken as the representative of polluted air of this region.

In order to estimate the ozone value before the air-masses enter the NWI region, its values from six to one day are linearly extrapolated back-ward to zero day condition as in the study of Naja et al. (2003). The ozone values corresponding to zero day will be equivalent to "background" ozone levels in the air-masses which are just entering the boundaries of NWI sector. Thus, the annual background mixing ratio of ozone are found to be 28.0 ± 3.6 ppbv for the boundary layer and 42.5 ± 2.3 ppbv for the lower troposphere. Estimated background ozone in the boundary layer may be affected by local photochemistry; however in the lower troposphere it should be more representative of regional background levels. The background mixing ratios of ozone are also calculated for different seasons. The background ozone and rate of increase of ozone with residence days within the boundary layer and in the lower troposphere during different seasons are given in Table 1. The background ozone (~26.3 ± 3.3 ppbv) obtained for JJA within the boundary layer represents the ozone level in the cleaner air of the Arabian Sea.

Table 1
The background mixing ratios of ozone and its increase rates (±standard error) in the boundary layer and in the lower troposphere are provided here for different seasons.

Season	Boundary layer		Lower troposphere	
	Background O_3 (with six days regression) (ppbv)	Rate of increase of O_3 with residence days (ppbv/day)	Background O_3 (with six days regression) (ppbv)	Rate of increase of O_3 with residence days (ppbv/day)
Winter	47.7 ± 3.2	0.7 ± 0.8	43.1 ± 3.0	0.8 ± 0.7
Spring	33.8 ± 4.3	4.5 ± 1.1	39.6 ± 3.3	3.4 ± 0.8
Summer	26.3 ± 3.3	2.6 ± 0.9	33.3 ± 2.6	3.2 ± 0.6
Autumn	40.3 ± 1.4	2.4 ± 0.4	44.9 ± 5.1	1.7 ± 1.2
Annual	28.0 ± 3.6	5.0 ± 0.9	42.5 ± 2.3	1.9 ± 0.6

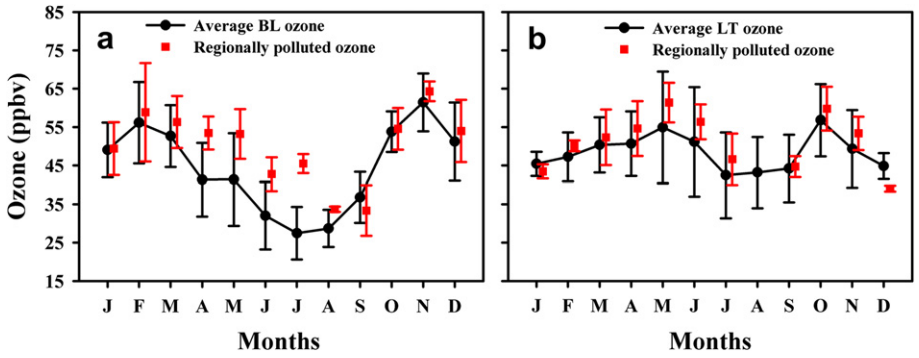

Fig. 5. The monthly distribution of average ozone with variation of ozone in regionally polluted air in the (a) boundary layer (BL) and (b) lower troposphere (LT). Vertical bars show $\pm 1\sigma$ variation.

4. Contributions of different air-masses

4.1. Regionally polluted air-mass

An air-mass is considered as the regionally polluted boundary layer air mass if it has spent 3 to 6 days in the NWI sector (significantly photo-chemically processed – based on ozone-residence time correlation plot) and its mean altitude remains lower than 3 km over this region. Air-mass within 3 km will restrict the contribution from higher heights. Similarly, an air-mass is considered as the regionally polluted lower tropospheric air mass if it has spent 3 to 6 days in the NWI sector, less than 5 days over NASE region with its mean altitude over NWI region less than 4.5 km. Criteria of "less than 5 days over NASE region" will remove probable influence of long range transport in the lower tropospheric region. Considering these criterion, the average monthly variations of the regionally polluted ozone with monthly average ozone obtained from all the data are shown in Fig. 5. The average ozone obtained from all the data show minimum during summer-monsoon with mixing ratios of about 30 ppbv in the boundary layer (Fig. 5a). This is largely due to south westerly wind which brings the pristine maritime air-mass to the observation site. Ozone levels in the regionally polluted air-mass are higher than average ozone by 22% during spring and 41% during summer-monsoon in the boundary layer. The average distribution of ozone in the lower troposphere is different compared to the variations in the boundary layer. Unlike the boundary layer, the two peaks are identified in average ozone variation during May and October months. The regionally polluted ozone levels are found to be higher (9–12%) than average ozone during spring and summer-monsoon. Some influence of regional pollution can also be seen in October and November months as regionally polluted ozone levels are higher by 5–7 ppbv than average monthly values. The difference between regionally polluted ozone and background ozone may provide information on photochemical build up of ozone that is contributed from Indian regional pollutants. Considering this aspect, the build up is maximum (~20.5 ppbv) during spring season in the boundary layer due to availability of intense solar flux and high levels of regional pollutants accumulated in shallow boundary layer during winter. Ozone build up is found to be 14.3 ppbv in summer-monsoon in the boundary layer (Fig. 6). The build up is nearly similar during spring and summer-monsoon in the lower troposphere. As expected, this build up is lowest during winter, both in the boundary layer and lower troposphere due to higher background ozone in this season as mentioned earlier.

4.2. Marine air-mass and influence of long range transport

During April to August period, the marine originated air-masses dominate particularly in the boundary layer as shown in Fig. 7a. The air-masses are categorized as the marine air-mass if they have spent more than 3 days over the marine region and less than 2 days over the NWI region. The mixing ratios of ozone are found to be 38% and 10% lower than the average ozone mixing ratios during spring and summer respectively. There were very few trajectories in the lower troposphere from the marine region and their residence times were also not sufficient to identify the contribution in the lower tropospheric region. Average mixing ratios of ozone in the marine air during this period was 26.5 ± 3.0 ppbv, which is closely

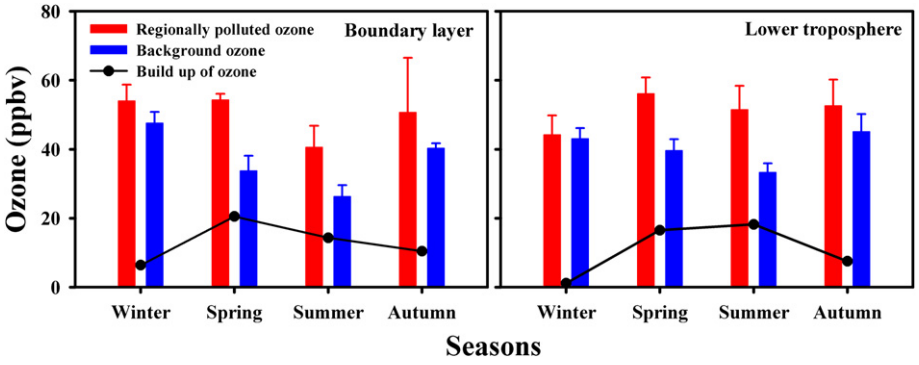

Fig. 6. The seasonal variation of regionally polluted and background levels of ozone. Vertical bars show $\pm 1\sigma$ variation in regionally polluted ozone and ±1 standard error in background ozone. The circles show the build up of ozone contributed from regional pollution.

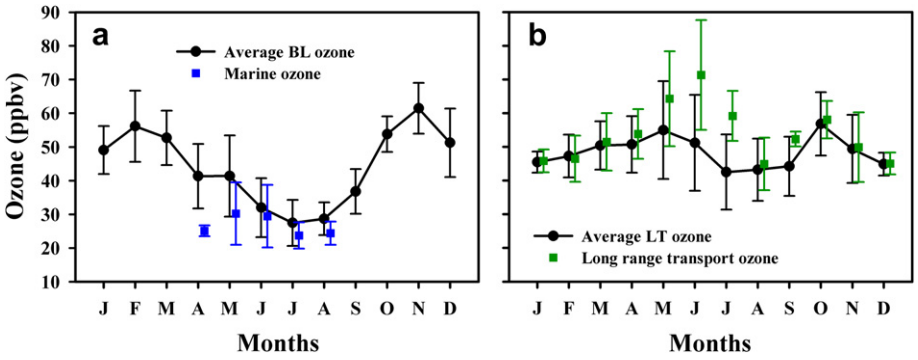

Fig. 7. The monthly distribution of (a) average ozone with monthly variation of marine ozone in boundary layer (BL) and (b) average ozone with monthly variation of long range transport ozone in the lower troposphere (LT). Vertical bars show $\pm 1\sigma$ variation.

comparable to the background level of ozone during summer season (Table 1).

Influence of long range transport (LRT) is expected not to be very significant in the boundary layer and this analysis was limited to the lower tropospheric region. The LRT air-masses are classified on the basis of three factors. They should have spent minimum 3 days over NASE region and less than 1.5 days over marine sector with their average altitude over NWI region greater than 3 km. This altitude criterion will restrict the contribution from NWI region. These air-masses show higher ozone levels by 9% and 27% than average ozone during spring and summer seasons respectively (Fig. 7b). It appears that both regionally polluted air mass and LRT play major role in variations of the lower tropospheric ozone.

5. Surface ozone at Ahmedabad

Surface level ozone data over Ahmedabad are also used in association with back-air trajectories to perform the residence time analysis. The surface ozone over Ahmedabad shows a typical variation of an urban location (Lal et al., 2000). The seasonal averaged diurnal variation of surface ozone during 2002 is shown for the reference (Fig. 8). The average diurnal variation shows a broad maximum during day time, a sharp decrease after about 1700 h and lower levels throughout the night. This day time photo-chemically processed ozone and night time background ozone are used in the residence time analysis of surface ozone during years 2002 and 2009. The backward trajectories are calculated at 500 m over Ahmedabad at 1430 h and 0230 h and tagged with the average ozone mixing ratios of daytime (1400–1500 h) and night time (0200–0300 h) for further analysis similar to that of ozone sounding data. Fig. 9 shows the dependence of surface ozone during daytime and night time on residence time over the NWI sector.

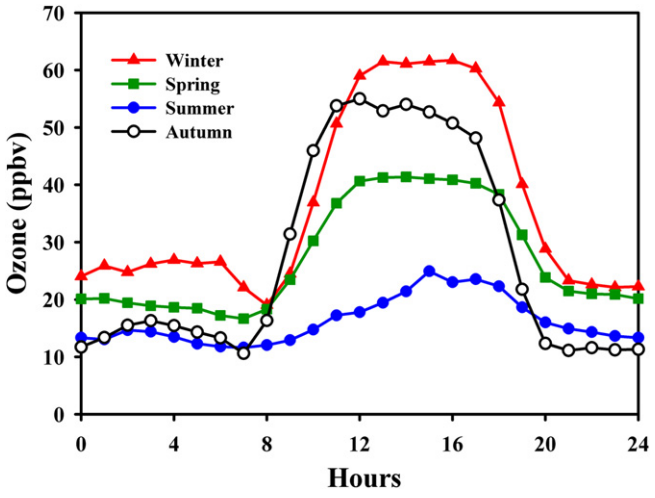

Fig. 8. The typical seasonal averaged diurnal variation of surface ozone over Ahmedabad during year 2002.

The surface ozone mixing ratios are found to increase with increasing residence days during daytime but not in the night time. This feature confirms that the availability of solar radiation during daytime is playing an important role and ozone build up with extended residence days is largely due to involvement of daytime photochemistry. Ozone growth rate is observed rather higher (Spring 5.6 $\pm$ 0.7 ppbv/day, Autumn 6.5 $\pm$ 0.5 ppbv/day, Winter 2.6 $\pm$ 0.7 ppbv/day,) for surface ozone analysis as compared with ozone sounding analysis. As expected, air-mass is largely from marine region during summer-monsoon period and therefore no data corresponding to residence time more than 4 days over this region.

It is seen that the ozone production rate is maximum during spring ($\sim$4.5 ppbv/day) in the boundary layer (ozonesonde data) and during autumn in the surface ozone ($\sim$6.5 ppbv/day). This is to observed that the boundary layer data from ozonesonde are regional representative whereas the surface ozone has more contribution from the local pollution over Ahmedabad.

6. Comparison with European sites

Naja et al. (2003) have done trajectory assisted ozonesonde data analysis over the two European sites, Hohenpeissenberg and Payerne. In contrast to present observations, ozone show a broad maximum during spring-summer-monsoon and minimum during autumn–winter for both boundary layer and lower troposphere. Similar variations are reported for several surface ozone sites over Europe (e.g. Bronnimann et al., 2000; Schuepbach et al., 2001). This is due to the influence of cleaner marine air-mass over Ahmedabad during late spring and summer, which is not the case over European locations.

In the present analysis of ozonesonde observations in the lower troposphere, the levels of ozone mixing ratios are found to be 20 ppbv higher in the air-mass arriving from NASE region (LRT) during summer-monsoon. The levels of ozone in the lower tropospheric LRT air-mass over Ahmedabad and European regionally polluted air-mass are almost comparable (60–70 ppbv) in summer-monsoon. During this season, the mean altitude of back-trajectories is found to be lower in the NASE region. Previous trajectory based and model based studies have suggested that European sources can have significant contribution in ozone levels over Asian region

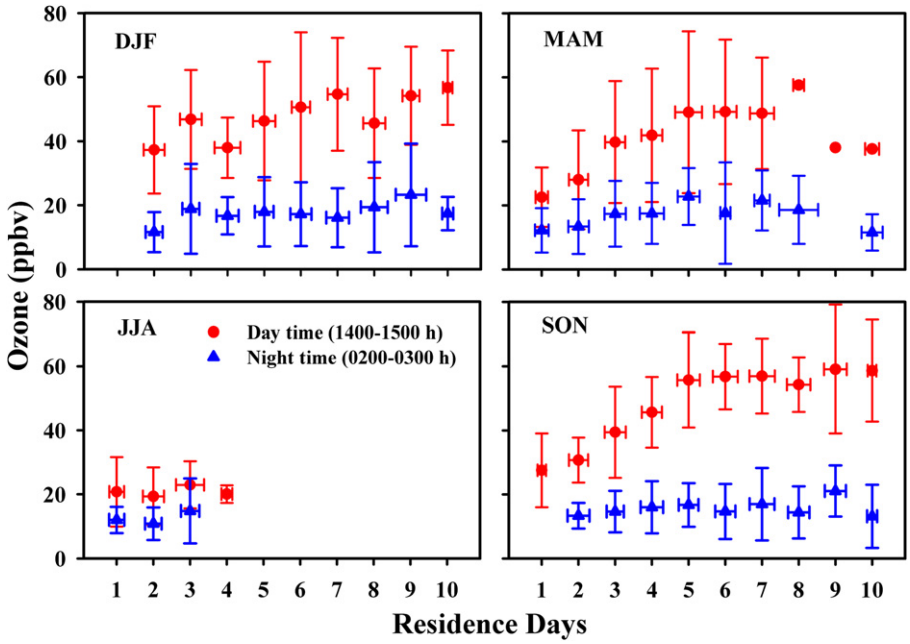

Fig. 9. The variation in surface ozone mixing ratios with residence days over NWI region in daytime (1400–1500 h) and night time (0200–0300 h) during winter (DJF), spring (MAM), summer-monsoon (JJA) and autumn (SON). The vertical and horizontal bars show $\pm 1\sigma$ variation in ozone mixing ratio and residence time respectively.

(Newell and Evans, 2000; Wild et al., 2004). However contribution from higher altitudes cannot be neglected in these air masses.

7. Conclusions

Four years of ozonesonde observations from Ahmedabad have been analyzed using ten days backward trajectories. The residence times have been estimated in the boundary layer and lower troposphere over NWI, marine and NASE regions. These residence times and associated observed ozone levels are studied to understand the effects of different air masses on the distribution of ozone. The ozone levels increase with increasing residence time of air parcels over the NWI region for about four to six days during all the four seasons in the boundary layer as well as in the lower troposphere. The rate of increase in ozone mixing ratios are highest (boundary layer $\sim 4.5 \pm 1.1$ ppbv/day, lower troposphere $\sim 3.4 \pm 0.8$ ppbv/day) during spring and lowest (boundary layer $\sim 0.7 \pm 0.8$ ppbv/day, lower troposphere $\sim 0.8 \pm 0.7$ ppbv/day) during winter. The background levels of ozone are estimated using six days regression line extrapolated to zeroth day in ozone-residence time correlation plots. The background levels of ozone are found minimum during summer-monsoon ($\sim 26.3 \pm 3.3$ ppbv) and maximum during winter ($\sim 47.7 \pm 3.2$ ppbv) within the boundary layer. Analysis of surface ozone data at Ahmedabad during noontime shows increase in ozone with residence time (Spring 5.6 ± 0.7 ppbv/day, Autumn 6.5 ± 0.5 ppbv/day and Winter 2.6 ± 0.7 ppbv/day) similar to that found from the balloon soundings. This confirms that increase in ozone with residence days is largely due to involvement of daytime photochemistry.

The regionally polluted ozone levels are estimated by restricting the contribution from other regions. This air mostly dominates within the boundary layer during winter season. The difference in average levels of ozone and regionally polluted ozone is small during this season due to large contribution of regionally polluted air itself. However, this difference is larger during spring ($\sim 22\%$) and summer-monsoon ($\sim 41\%$) when the contribution from the marine air drags down the average levels of ozone. The photochemical build up of ozone (difference of regionally polluted and background ozone) due to local pollutants is found to be maximum during spring season (~ 20.5 ppbv) within the boundary layer. The ozone in the marine air is found to be lower than average ozone by 38% and 10% during spring and summer respectively within the boundary layer. In the lower troposphere, a significant contribution of regionally polluted air is found during spring and summer-monsoon seasons (~ 9–12%). LRT is found to be important in the lower troposphere with contribution of 9% and 27% during spring and summer seasons respectively.

This work suggests that mixing ratios of surface ozone are more influenced by local pollution of the city whereas boundary layer ozone is the regional representative of NWI region. In addition to that, the boundary layer ozone distribution is affected by marine air advection and regional emission whereas lower troposphere is more influenced by regional pollution and the long-range transport.

The outflow of air pollution from European region to downwind locations is not well studied yet. The intercontinental transport of ozone and other trace species from Europe to Asian countries can enhance the background levels of ozone in the lower troposphere (Newell and Evans, 2000). It would be rather more useful to have regular and frequent balloon flights from this region to better quantify contributions to ozone levels from different regions especially from the European and African regions.

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