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Simultaneous Measurements of Surface Ozone at Two Sites over the Southern Asia: A Comparative Study

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ABSTRACT

This article presents variations of simultaneous measurements of near surface ozone (O_3) at two sites namely Anantapur [14.62°N, 77.65°E], a semi arid rural location in India and Xi'An [34.20°N, 108.98°E], a semi arid urban location in China during January-July 2009. The results showed a clear diurnal cycle of O_3 with a minimum at sunrise and a maximum at noon for both the sites. The monthly average diurnal variation shows that the maximum/minimum ozone was observed in March/July whereas in Xi'An maximum/minimum ozone was observed in July/February because of different climatic zones and rainfall activity. The average diurnal variation of O_3 for different seasons (summer and winter) shows higher ozone concentration at Anantapur than at Xi'An. This may be due to slower titration of NO in the evening hours at Anantapur. But in Xi'An, the highest ozone levels recorded in noon hours for some days in June and July months. This is mainly due to strong emissions of NO_x, VOC and high solar radiation and this implies significant negative effects on vegetation and regional air quality around Xi'An. The rate of increase of ozone is almost the same at two sites but the rate of decrease of ozone is more at Xi'An than at Anantapur which is due to the higher NO_x concentration from vehicular emission and also due to the fast titration of O_3. The maximum 54% of frequency distribution of ozone lies between 20–45 ppbv at Anantapur whereas in Xi'An 34% lies in the range of 0–5 ppbv, 32% of Ozone lie between 5–20 ppbv and 24% of all O_3 lie in the range of 20–45 ppbv.

Keywords: Surface ozone; Photoxidation; NO_x; Asian winter monsoon.

INTRODUCTION

Ozone (O_3) is a protective component in stratosphere, which absorbs the major ultraviolet ray and prevents the solar radiation with wavelengths less than 280 nm from reaching the earth's surface. However, ozone is considered as a pollutant at ground level and the overall effect of ozone exposure leads to the decrease of lung capability to perform normal function. Hence, ozone can be regarded as a beneficial ultraviolet shield in the stratosphere but a harmful matter to human being at the ground level. The major primary pollutants emitted from motor vehicles include nitrogen oxides NO_x (nitric oxide NO and nitrogen dioxide NO_2), carbon monoxide CO, respirable suspended particulates RSP, sulphur dioxide SO_2, and volatile organic compounds VOC, etc. The reduction of surface ozone which is harmful to human, animal, and plant health, is an important objective of air quality policy for many governments. Surface ozone is produced through a complex set of photochemical reactions involving NO_x (= NO + NO_2) and volatile organic compounds (VOCs). NO_x and VOCs are emitted from anthropogenic sources such as fossil fuel power plants, industrial activities and transportation, as well as natural sources such as lightning and soil (NO_x), and vegetation (biogenic VOCs such as isoprene). The resulting ozone concentrations depend sensitively upon meteorological parameters such as temperature, sunlight, wind speeds and the mixed layer depth. Therefore, changes in these meteorological

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parameters due to climate change will necessarily impact surface ozone concentrations. However, the direction of change itself is often unclear because of multiple competing effects.

In recent years, O_3 chemistry has received considerable attention in Asia owing to rapid economic and industrial development, accompanied by its increasing air pollutant emissions. India and China are most populous countries in the world and also they two are one of the top energy consuming countries. The temporal variations of O_3 have been reported at many sites including rural, urban, coast and mountain sites in India (Lal *et al*., 2000; Nair *et al*., 2002; Debaje *et al*., 2003; Naja *et al*., 2003; Satsangi *et al*., 2004., Reddy *et al*., 2010), China (Shan *et al*., 2008., Ran *et al*., 2009), and Thailand (Pochanart *et al*., 2001; Zhang and Oanh, 2002). These studies also showed great spatial variability. For instance, higher O_3 values did not appear during daytime at a mountain site in India (Naja *et al*., 2003), while significant increasing O_3 values during daytime were observed at an urban site, Ahmedabad, in tropical India (Lal *et al*., 2000). The seasonal variation in O_3 at Ahmedabad showed that O_3 concentrations in autumn and winter months were higher; while those in summer months were lower (Lal *et al*., 2000). On the contrary, the seasonal variation of O_3 at an urban area, Nagoya, Japan, showed that higher concentrations were observed from late spring to summer, while lower concentrations were observed in winter (Saito *et al*., 2002).

The tropospheric ozone changed dimensions about fifteen years ago, when it was realized that the increased surface ozone is not only a local urban problem but also a global increase of surface rural ozone concentration, especially in the northern hemisphere. This was observed during the 20th century, which has been attributed to photochemical production (Volz and Kley 1988; Sillman *et al*., 1990; Lin *et al*., 1998; Crutzen *et al*., 1999). Measurements during the second half of the last century at many sites show that the present levels of ozone have raised more than double their levels (Anfossi *et al*., 1997; Volz and Kley 1998). Estimations made using the chemical transport model showed that on increasing the anthropogenic emissions, ozone production efficiency was maximum over the Indian region followed by Japan and China. This can be explained on the basis of an increase in OH peroxy radicals (Berntsten *et al*., 1996). Over the globe, especially in southern Asian countries, a few scared surface measurements of ozone are being carried out based on ozone sensors (Lal *et al*., 2000; Reddy *et al*., 2010). These measurements are restricted to a few rural and urban locations in southern Asia countries. However, no simultaneous measurements using the same technique were carried out in Asian countries, for a comparative study. Hence in this article, we presented the simultaneous measurements of surface ozone at Anantapur, India and Xi'An, China over the Asian continent. A comparison of variations in surface ozone on diurnal, monthly and seasonal scales over the observation sites was also discussed.

MEASUREMENT SITE, METEOROLOGY AND EXPERIMENTAL TECHNIQUE

The measurements were carried out simultaneously at Anantapur [14.62°N, 77.65°E], a semi-arid rural site in India and at Xi'An [34.20°N, 108.98°E], a semi-arid urban location in China (Fig. 1). The Anantapur site is about 200 km and 350 km from the two nearby major cities, Bangalore [12.5°N, 77.4°E] to the southwest and Hyderabad [17.4°N, 78.4°E] to the northeast, respectively. Both contribute to the enhancement of O_3 precursor gases in these areas. And Anantapur is geographically situated on the boundary of a

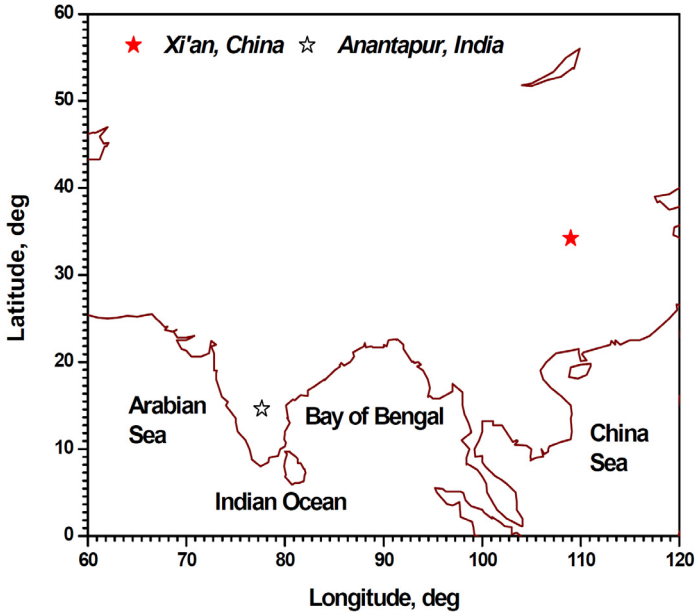

Fig. 1. Geographical location of sampling sites of Anantapur [14.62°N, 77.65°E] (a semi arid rural zone in India is indicated by Black star) and Xi'An [34.2°N, 108.98°E] (an urban scale zone in China is indicated by Red star).

semi arid and rain shadow region. The climate is hot and dry in the summer (March–May), hot and humid during monsoon (June–November) and dry during the winter (December–February). The normal rainfall during the southwest monsoon period is 350 mm, which forms more than 60–70% of the total rainfall, whereas for the northeast monsoon period is it only ~150 mm, which forms 40–30% of annual rainfall. The remaining months of summer are warm and dry with a normal daily maximum temperature that ranges between 29°C to 42°C. The winter months are cooler when the temperature falls about 20°C which is fair weather. This condition prevails with low wind speed which is of the order of 4–5 m/s with southeasterly direction and moderate relative humidity that exists during this season. Surface ozone is measured using an analyzer (O_3 41 M; Environnement S.A., France) based on absorption of Ultraviolet (UV) radiation at 253.7 nm by ozone molecules. Contribution by other species in the absorption and scattering of the radiation in the cell is eliminated by comparing the measurement with ozone free air in reference mode. Systematic and regular measurements of surface ozone have been made at observation site with the ozone analyzer Xi'An is situated in the southern part of the Guan Zhong Plain in Shaanxi province, with the Qinling Mountains to the south and the Weihe River to the north. It is in a warm temperate zone, and has a continental monsoon climate with four distinct seasons; Xi'An is hot and wet in summer, dry and seldom snowy in winter and is prone to extended spells of rain in spring and autumn. Summer in Xi'An (June to August) becomes both hot and humid. The climate of Xi'An is dominated by large variations in East Asian monsoon. During the winter half a year, the northerly Asian winter monsoonal winds blow, and they are associated with the transport of Asian dust from Chinese deserts and nearby loess areas (Zhang *et al*., 1993, 1997). In summer, the monsoonal winds bring moisture to the region from the south. Xi'An is one of the major 'stove' cities of the whole country with the hottest month of the year, i.e. July, with the highest average temperature i.e. 32°C and the maximum temperature is often above 40°C. Half of Xi'An's annual rainfall occurs during the summer. In summer, the southeast monsoon from the western Pacific Ocean and the southwest monsoon from the equatorial Indian Ocean blow onto the Chinese mainland. These monsoons are the main cause of rainfall. The annual rainfall is occured between 558–750 mm. Ozone was continuously measured by using a UV photometric analyzer (TEI, model 49C). The instrument was checked automatically every day by scrubbed ambient air and with a span standard generated in a multi-gas calibrator (TEI, model 146). The height of the air intake was 8.5 m above the ground.

RESULTS AND DISCUSSION

Ozone Variations on a Diurnal Scale

Diurnal variations have a major influence on exposure levels at sites nominally exposed to the same regional ozone distribution. The physical and chemical mechanisms which give rise to diurnal variations are detailed so that sites can be screened for different diurnal behavior characteristics (Derwent and Kay 1988).

Ozone concentration slowly decreases during nighttime due to the chemical loss by NO and deposition process. However, it started rapidly coinciding with the increase of solar radiation after sunrise by photochemical production. Therefore, the time of sunrise is a turning point of diurnal ozone variation trend. The average diurnal variation of O_3 over the two observation sites has been studied for the study period is shown in Fig. 2. Ozone diurnal variation of each site showed a similar pattern, but the magnitudes of variations were different. Fig. 2 depicts that minimum values of O_3 appeared in the early morning 06:00–08:00 LT (Local Time) and the increases in ozone concentrations were then observed. The highest levels occurred approximately at

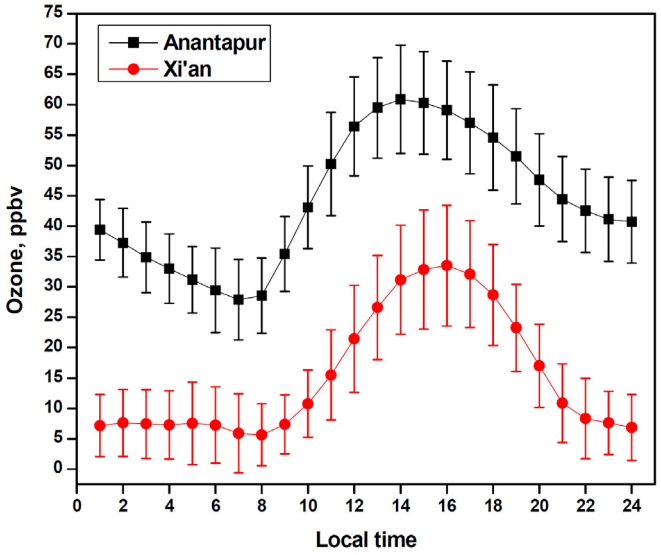

Fig. 2. A clear diurnal cycle of hourly mean ozone for the study period measured at Anantapur, India and Xi'An, China respectively.

14:00–16:00 LT. Thereafter, ozone concentrations decreased steadily. This diurnal pattern is similar to that found in several rural sites in China and India (Cheung and Wang 2001; Naja *et al*., 2003; Debaje *et al*., 2006). An increase in ozone levels during the day is attributed to photochemical processes of ozone production in the mixing layer and transport from the upwind site and layer, both favored by solar radiation, and a decrease at night is due to in situ destruction of ozone by deposition and/or the reaction between O_3 and NO (Duenas *et al*., 2002). It can also be found that the daily amplitude of surface ozone is highest in summer at both the sites. Meanwhile, the maximum hourly mean concentration observed at Anantapur (Xi'An) in the summer is 73.25 ppbv, 14:00 LT (64.39 ppbv, 14:00 LT), and the minimum 20.34 ppbv, 07:00 LT in the month of July during monsoon (1 ppbv, 06:00 LT in the month of February during winter) for the sampling period. Daytime high ozone concentration in summer, due to favorable meteorological conditions has aroused great interest in the study of photochemical pollution in urban areas. After sunrise the NO concentration quickly increases to its peak value at 07:00–10:00 LT and then started to decrease. Lower levels appear at night due to the titration of NO by ozone and cessation of photolysis of NO_2 after sunset. NO_x can react with ozone in atmosphere as following reactions:

$$NO_2 + h\nu \rightarrow NO + O \quad (1)$$
$$O + O_2 + M \rightarrow O_3 + M \quad (2)$$
$$O_3 + NO \rightarrow NO_2 + O_2 \quad (3)$$

In reaction (1) and (2), NO_x is a positive effect for ozone formation, whereas reaction (3) is a negative effect. It shows that NO_x can not only form but also consume ozone. Excessive NO_x in urban areas is unbeneficial for the accumulation of ground-level ozone. Besides the photochemical reactions affected by solar radiation and variations in anthropogenic emissions, boundary layer processes and meteorological parameters also play important roles in the process of diurnal variations in O_3 and its precursors.

Daily/Monthly/Seasonal Variations in Surface Ozone

Regional ozone pollution has become one of the top environmental concerns in Asia, especially in those economically vibrant and densely populated regions, such as India and China. To address this issue, surface ozone and ancillary data over Anantapur and Xi'An were analyzed during the study period. Some detailed statistics of O_3 including the monthly-averaged diurnal minima/maxima with their appearing time, diurnal amplitudes, and monthly mean concentrations in each months are listed in Table 1. Variation tendency of daily mean, daily maximum and monthly mean ozone concentrations in over the two observation sites (Anantapur and Xi'An) during the study period (Jan-July 2009) is also shown in Fig. 3. The highest diurnal maximum (73 ppbv/64 ppbv for Anantapur/Xi'An) was observed in summer whereas minimum (21 ppbv/1 ppbv for Anantapur/Xi'An) observed in monsoon/winter at two observation sites. This variation pattern is almost the typical mode that observed sites in the Northern Hemisphere. But the summer maximum of ozone in both the observation sites are just contrary to some cities under the impact of summer monsoon, such as Hong Kong and Thumba, where are dominated by summer minimum (Wu and Chan 2001; Nair *et al*., 2002). In general, ozone variation on diurnal scale can provide insight into the interplay of emissions, and chemical as well as physical processes that operate on a diurnal cycle. It can also be noticed that averaged diurnal variation during seasons for the study period (January–June 2009) shows high ozone concentrations at the rural site Anantapur than the urban site, Xi'An. In most Chinese cities, ozone levels were higher in summer and lower in winter (Shan *et al*., 2008).

The ozone concentrations of Anantapur and Xi'An in the temperate zone have one peak value during study period around occurring in March, while Xi'An it is gradually increasing order form June–July 2009, among which the highest occurs in July 2009. This is related to the difference of rainy seasons in different climatic zones. The maximum ozone concentration was observed in the month of March, 56.08 ± 9.24 ppbv (July, 32.66 ± 6.32 ppbv) and minimum in July, 27.26 ± 3.74 ppbv (February, 2.90 ± 1.52 ppbv) at Anantapur (Xi'An) respectively. It is also seen from the figure that the O_3 concentration at Anantapur were higher than those at Xi'An except in July because of cloudy sky condition and rainfall activity. Tropospheric O_3 maximum in June at Xi'An is a result of strong photochemical production, transport of regional pollution, and possibly also more intense burnings of biomass in Central- Eastern China.

Table 1. Detailed statistics of O_3 including the monthly-averaged diurnal minima/maxima with their appearing time, diurnal amplitudes, and monthly mean concentration in each month for the study period.

Month	Anantapur, India				Xi'An, China			
	Mean ozone ppbv	Diurnal Max, ppbv (time)	Diurnal Min, ppbv (time)	Diurnal Amplitude	Mean ozone ppbv	Diurnal Max, ppbv (time)	Diurnal Min, ppbv (time)	Diurnal Amplitude
January–09	43.56	62 (14:00)	21 (08:00)	41	3.99	12 (15:00)	1 (06:00)	11
February–09	43.71	62 (15:00)	20 (07:00)	42	2.90	10 (15:00)	1 (07:00)	09
March–09	56.08	73 (15:00)	31 (06:00)	42	7.72	20 (15:00)	2 (08:00)	18
April–09	53.85	72 (16:00)	34 (07:00)	38	13.55	29 (16:00)	5 (07:00)	24
May–09	46.48	63 (15:00)	32 (08:00)	31	18.52	36 (15:00)	8 (07:00)	28
June–09	34.47	44 (16:00)	26 (07:00)	18	28.48	64 (14:00)	7 (06:00)	57
July–09	27.26	31 (14:00)	23 (07:00)	08	32.66	62 (13:00)	12 (06:00)	50

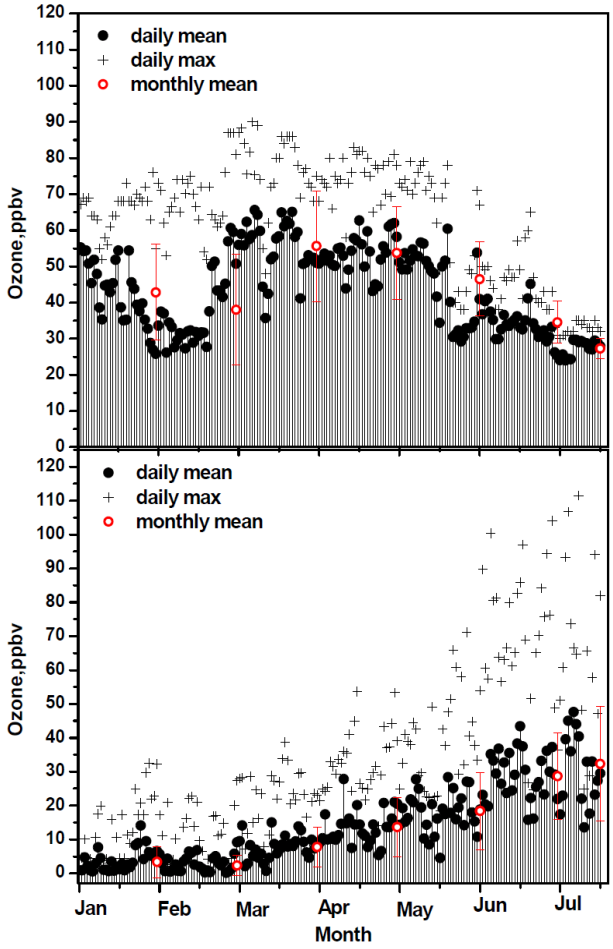

Fig. 3. Variation tendency of daily mean, daily maximum and monthly mean ozone concentrations over the two observation sites, Anantapur (top panel) and Xi'An (bottom panel) during the study period (Jan–July 2009).

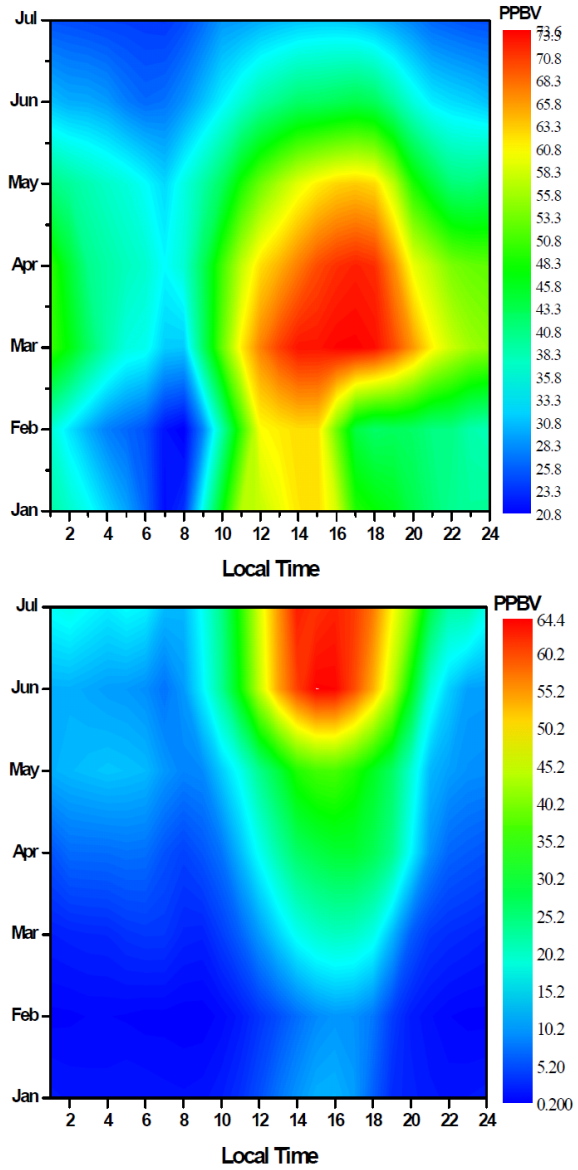

Fig. 4. Seasonal variation of surface ozone on a diurnal scale over the observation sites, Anantapur (top panel) and Xi'An (bottom panel).

O_3 shows a well-defined seasonal variation at these sites. The low values appear in winter, while the high values appear in early summer. Fig. 4 reveals that ozone presents a maximum (52.14 ± 9.43 ppbv for Anantapur and for Xi'An 30.56 ± 9.56 ppbv) in summer but minimum (30.86 ± 3.65 ppbv for Anantapur and for Xi'An 3.44 ± 1.25 ppbv) in monsoon at Anantapur and in winter at Xi'An. This is mainly due to different regional, climatic zones and also the contribution of photochemically generated O_3 from sunshine and anthropogenic and natural precursors (Jo and Park, 2005).

Daytime/Nighttime Variations and Rate of Change of Surface Ozone

The monthly average daytime/night time ozone for the study period is shown in Fig. 5. We have divided total 24 h into daytime (08:00 h–20:00 h) and night time (20:00 h–08:00 h) changes of zone for the total study period. The corresponding statistical data is presented in Table 2. The monthly average daytime/night time maximum ozone occurred in the month of March, 67.07 ± 9.85/45.09 ± 9.85 ppbv (July, 48.47 ± 9.96/16.48 ± 3.04 ppbv and minimum in July, 29.74 ± 1.55/24.77 ± 0.73 ppbv (February 5.26 ± 3.39/0.53 ± 0.14 ppbv) at Anantapur (Xi'An). The daytime increase in ozone is due to photoxidation of precursor gases in presence of NO_x. Whereas in nighttime decrease in ozone is due to dry deposition and titration effect due to NO (see Eq. (3)).

Monthly average rate of change of ozone in the morning hours (08:00–11:00 h) and evening hours (17:00–19:00 h) at Anantapur and Xi'An during January–July 2009 is presented in Table 3. The total average rate of change of ozone during morning hours (evening hours) is 4.54 ppbv/h (–2.59 ppbv/h). This is mainly due to fast production of O_3 by freshly emitted NO. Whereas in Xi'An, the total rate of change of ozone in morning hours is 4.67 ppbv/h and evening hours is about –5.35 ppbv/h. The rate of change of ozone during evening hours is more at Xi'An compared to Anantapur.

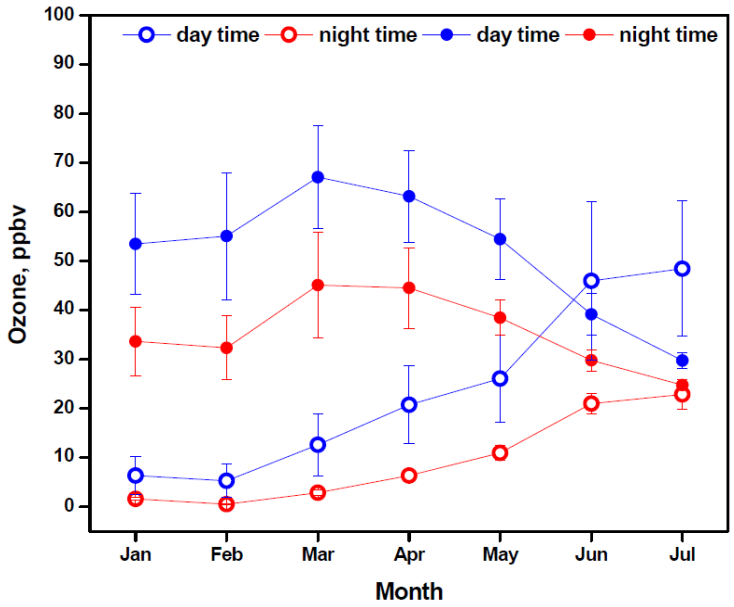

Fig. 5. Daytime/ Night time cycle of ozone for different months at sampling sites (Solid circle indicates Anantapur and hallow circle indicates the Xi'An).

Table 2. Monthly average ozone concentrations with ± 1σ standard deviation observed at Anantapur, India and Xi'An, China during day time (08:00–20:00h) and night time (20:00h–08:00h) for the study period.

Month	Anantapur, India		Xi'An, China	
	Daytime	Nighttime	Daytime	Nighttime
January–09	53.48 ± 9.26	33.65 ± 6.95	6.36 ± 3.84	1.61 ± 0.18
February–09	55.08 ± 9.56	32.33 ± 6.48	5.26 ± 3.39	0.53 ± 0.14
March–09	67.07 ± 9.85	45.09 ± 9.85	12.60 ± 6.31	2.83 ± 0.55
April–09	63.16 ± 9.25	44.53 ± 8.65	20.73 ± 7.19	6.35 ± 0.98
May–09	54.48 ± 8.22	38.49 ± 3.65	26.07 ± 8.92	10.95 ± 1.54
June–09	39.15 ± 4.15	29.78 ± 2.21	45.97 ± 9.65	10.98 ± 2.02
July–09	29.74 ± 1.55	24.77 ± 0.73	48.47 ± 9.96	16.48 ± 3.04

Table 3. Monthly rate of change of ozone during morning (08:00–11:00 h) and evening hours (17:00–19:00 h) for the study period.

Month	Anantapur, India		Xi'An, China	
	Rate of Ozone at (08:00–11:00 h) ppbv/h	Rate of Ozone at (17:00–19:00 h) ppbv/h	Rate of Ozone at (08:00–11:00 h) ppbv/h	Rate of Ozone at (17:00–19:00 h) ppbv/h
January–09	8.51	–4.39	1.27	–2.03
February–09	7.23	–3.81	1.08	–2.95
March–09	6.19	–2.83	2.58	–2.82
April–09	4.52	–2.42	3.77	–5.57
May–09	2.57	–1.83	4.80	–5.64
June–09	1.98	–1.29	9.62	–10.62
July–09	0.81	–1.62	9.63	–7.84

This is attributed to the higher NO_x concentration from vehicular emission and also due to the fast titration of O_3. This feature of ozone variations distinguishes the urban and rural sites.

Frequency Distribution of Ozone

Fig. 6 shows the frequency distribution of Ozone in different ranges with interval of five ppbv at two easurement sites for the study period. It is observed that the Anantapur site has 54% of O_3 lie in the range of 20–45 ppbv and 35% lie in the range of 45–65 and also the lowest 4% lie in the range 80–90 ppbv but whereas in Xi'An 34% of all O_3 lie in the range of 0–5 ppbv (32% of Ozone lie between 5–20 ppbv) and 24% of all O_3 lie in the range of 20–45 ppbv and the lowest 1% is observed lie in the range of 90–160 ppbv. Remaining 9% of ozone lies between 45–90 ppbv.

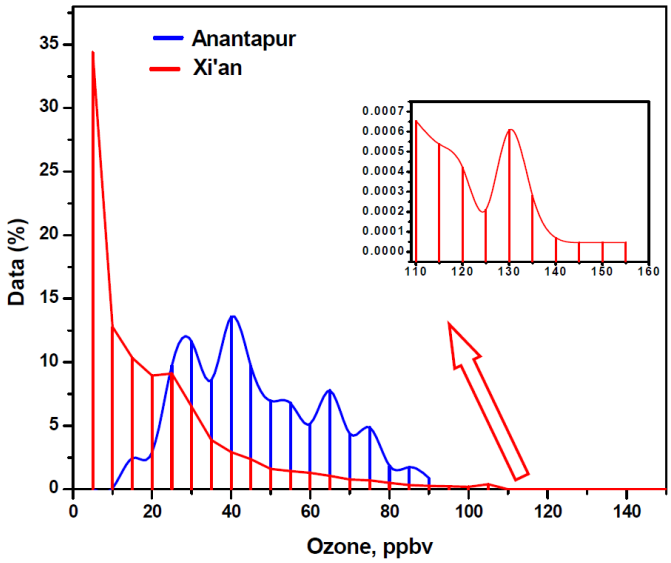

Fig. 6. Frequency distribution of ozone concentration for the measurement period at Anantapur, India and Xi'An, China.

SUMMARY AND CONCLUSIONS

In this study we presented the simultaneous measurements of surface ozone at Anantapur, India and Xi'An China over the Asian continent. The major conclusions are given below.

1. The total average diurnal variation of O_3 shows maximum/minimum 58.14 ± 6.54 ppbv/26.71 ± 3.82 ppbv and 33.28 ± 4.36 ppbv/5.14 ± 1.53 ppbv at noon/early morning at Anantapur and Xi'An, respectively.
2. The highest ozone concentration was observed in summer 91 ppbv for Anantapur and 157 ppbv for Xi'An) but lowest was observed in monsoon at Anantapur (11 ppbv) in winter at Xi'An (~1 ppbv).
3. The very high levels of O_3 suggest strong emissions of NO_x and VOC from Xi'An and imply significant negative effects on vegetation and regional air quality around Xi'An.
4. High ozone at Anantapur due to combined effect slower titration by NO during evening hours and the greater ozone formation due to pollutants transported from the near by polluted locations.
5. The rate of increase of ozone from morning to noon time was more or less the same in two observation sites, but the rate of decrease of ozone during evening hours was less in the Anantapur than at Xi'An. This is attributed to the higher NO_x concentration from vehicular emission and also due to the fast titration of O_3.
6. The maximum 54% of frequency distribution of ozone lies between 20–45 ppbv at Anantapur whereas in Xi'An 34% lies in the range of 0–5 ppbv, 32% of Ozone lie between 5–20 ppbv and 24% of all O_3 lie in the range of 20–45 ppbv.

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