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Surface ozone during a winter when coal fired heat was used in the urban area of Jinan, China

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ABSTRACT

Near surface ozone, NO, NO_2, and solar radiation levels were measured during late 2003 and early 2004, in the urban area of Jinan, China. Ozone levels were relatively low throughout the study period, while NO_x were relatively high compared with other cities in the world. Statistical analysis of ozone showed a typical diurnal variation pattern of urban areas in Jinan, with the maximum in the afternoon and minimum in the early morning before sunrise. Analysis of ozone levels and solar radiation data in the daytime 8 h (08:00-16:00) shows that the variation of ozone follows solar radiation with a delay of about two hours, with both showing similar behavior. Further studies indicate that solar radiation is a significant impact factor for the daytime variations of ozone concentrations in Jinan, especially in the morning hours. Regression analysis of ozone level and NO_2/NO concentration ratio shows that they correlated well with each other. In late winter and early spring days local effects play a major role in determining ozone concentrations in Jinan.

Keywords: Surface ozone; Photochemical pollution; Ozone variation; Pollutant monitor; Jinan.

INTRODUCTION

Ozone (O_3) is one of the important secondary air pollutants because it is harmful to humans and plants at high concentrations (e.g. Burnett et al., 1997; Manning, 2003; Kim et al., 2004). The study of ozone levels and development has become a popular topic. Ozone and related trace gases and meteorology have been systemically monitored (e.g. Croes and Fujita, 2003; Hidy 2000; Solomon; 2000); factors and processes affecting ozone formation and accumulation have been extensively analyzed (e.g. Kleinman, 2005; So and

Wang, 2003; Wang, et al., 2001; Bloomfield et al., 1996), and the adverse effects of ozone pollution on human health and plants has been comprehensively studied around the world (e.g. Rabl and Eyre, 1998; Arbaugh, et al., 1998; Musselman and Minnick, 2000). However, each geographical region has distinct characteristics that require independent interpretation of the ozone chemistry, precursor emissions, and meteorology.

In the lower troposphere, ozone is mainly produced by photochemical reactions, it is formed by the action of UV light from the sun on nitrogen oxides, and the reaction is influenced by VOCs, CO, etc. Its production and concentration depend on the UV light and the presence of primary pollutants (e.g. NO, NO_2, CO, VOCs, etc.), which are released from various natural and anthropogenic processes. Studies have shown that the ozone production does not increase linearly when the precursors are increased (e.g. Dimitriades, 1977). Urban ozone formation is a complex phenomenon, therefore, many researchers have tried to study urban ozone levels along with their related meteorological factors and ozone production precursors, as well as their interrelationships (e.g., Abdul-Wahab et al., 2005). In recent years, the rapid economic development in China has brought drastic atmosphere pollution incidents. Major cities, such as Beijing, Shanghai, Hong Kong, Tianjin, Lanzhou, and Jinan, are faced with photochemical threat, and high ozone concentrations are reported frequently (He and Huang, 1995; Cheung and Wang, 2001; Lu et al., 2002; Xu et al., 1999; Yin et al., 2006). However, there are limited reports on surface ozone observed data in the urban sites of China mainland.

Surface ozone is mainly monitored in industrialized and relatively highly-populated continental regions because of its potential impact on human health and vegetation (Duenas et al., 2004). Jinan is the capital of Shandong Province in East China, with an area of 8, 227 km^2 and a population of 5.53 million, including 2.57 million urban people. This city, like other big cities in China, faces air pollution problems. Coal is the leading fuel in Jinan, which accounts for more than 70% of the total energy. Coal burning discharges hundreds of kilotons pollutants every year and is widely used for power heating. In recent years, with the rapid increase in the number of motor vehicles, an evident change is taking place in the characteristic of air pollution in Jinan. Surface ozone has obviously increased, so some areas of the city present symptoms of photochemical smog (Yin et al., 2006). However, until recent years there have been scarce systematic simultaneous measurements of surface ozone and its precursor gases in this city.

In order to study the ozone tendency in the urban area in Jinan, it is necessary to have a complete understanding of the relationship between ozone tendency and variations of meteorological factors and ozone precursor concentrations.

We have reported high ozone concentrations of nearly 300μg/m^3 caused by favorable meteorological factors and precursor conditions (Shan et al, 2006). In this study, we present measurements of ozone, NO, NO_2, and solar radiation obtained in the urban area of Jinan during a period when coal fired heat was used from November 2003 to March 2004. These measurements will not only help to study the urban air quality but will also be useful in studying the changes in the chemical composition of the regional atmosphere and adopting efficient pollution control measures. The data from this study were used to characterize the levels and variations of ozone, NO, NO_2 and NO_x (NO + NO_2), and to analyze the correlation of ozone and NO_2/NO; ozone and solar radiation. On the basis of the above studies, a simple ozone prediction model was constructed and tested.

EXPERIMENTAL

Study site

Measurements were carried out on top of the building of the Environmental Science and Engineering college (36°42' N, 117°08' E, 34.5m a.s.l.), on the campus of Shandong University, which is in the eastern area of Jinan (Fig. 1), with a yearly average temperature of 14¡æ and an annual mean precipitation of 650-700 mm. Three directions of the city are surrounded by mountains, and in the north is the Yellow River embankment, making Jinan a basin location. The unfavorable local topography means pollutants easing accumulate in the urban area.

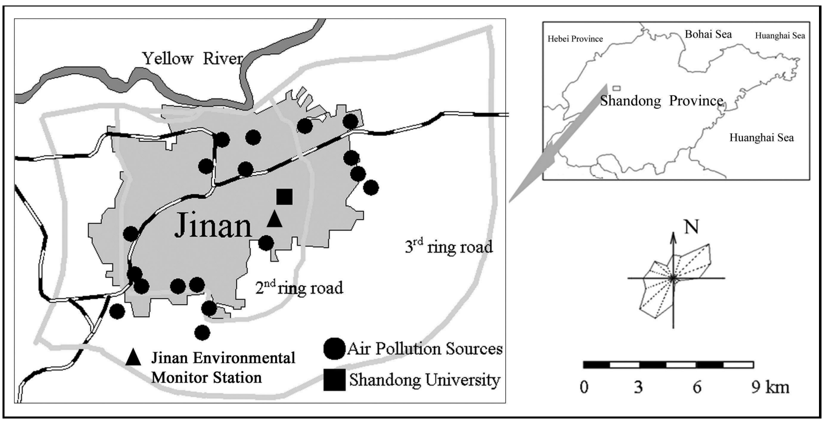

Fig. 1. A map showing Jinan, the observational sites, and major air pollution sources. Shandong University is the observation site of pollutants (O_3, NO, NO_2); Jinan Environmental Monitor Station is the observation site of solar radiation.

Measurement techniques

Ozone was continuously measured using a UV photometric analyzer (Thermo Environmental Instruments, model 49C). The limit of detection is 2 ppbv (signal-to-noise ratio (S/N) = 2, 2-min average) and the precision is ± 2 ppbv. NO and NO_2 were continuously determined with a chemiluminescence analyzer (Thermo Environmental Instruments, model 42S). These instruments were operated continuously operating every day for 24 h. The interval of each measurement was 1 min and the data presented in this paper are hourly averaged values. Air samples were collected through Teflon inlet tubes. The height of the air intake was 8.5 m above the ground. The solar radiation data were obtained from the Jinan Environmental Monitor Station, which is about 1.5km south-west of the pollutant monitoring site. Microsoft Excel 2003 and Microcal OriginPro 7.0 were used as statistics, analysis and graphing software.

RESULTS AND DISCUSSION

Temporal variation of ozone

The ozone levels in Jinan were analyzed statistically hourly, daily and monthly from November 2003 to March 2004 when coal-fired boilers are used for heating.

Table 1 shows some statistical characteristics of the hourly average ozone concentrations, these statistics reveal rather low ozone pollution in the monitoring period. The maximum hourly averaged ozone concentration in this period is 185.54g/m^3 (86.70ppbv), occurring on November 30, 2003, lower than the US National Ambient Air Quality Standards (NAAQS) for ozone (1-h 120 ppbv). Many previous studies have pointed out that, due to the influence of the Asian monsoon winter is the season with the lowest ozone levels in this region (e.g. Cheung and Wang, 2001), and since the monitoring was mainly done in winter, the obtained ozone values are relatively low. The low ozone levels also indicate that, the effect of heating in winter to ozone pollution is not as significant as meteorological factors.

There is a very clear variation of surface ozone concentration over the five months monitored. Ozone levels in November 2003 slowly decreased through December 2003, and then continuously increased (see Fig. 2). The minimum daily averaged ozone concentration in this period was 5.37μg/m^3, observed on December 5, 2003, while the maximum was 117.35μg/m^3, observed on March 15, 2004. A polynomial fit of the daily values of ozone concentration is shown in Fig. 2, the correlation coefficient is 0.76.

Table 1: Statistical results of measured ozone concentrations from November 2003 to March 2004 in Jinan

Parameter	Nov-03	Dec-03	Jan-04	Feb-04	Mar-04
Mean (g/m^3)	23.76	20.73	27.91	44.18	69.12
Median (g/m^3)	15.77	15.98	18.84	38.14	69.00
Min (g/m^3)	2.62	3.44	1.80	1.66	2.10
Max (g/m^3)	185.54	70.97	112.76	132.94	150.78
SD	22.76	15.22	24.44	34.61	37.38
n	720	737	741	696	742

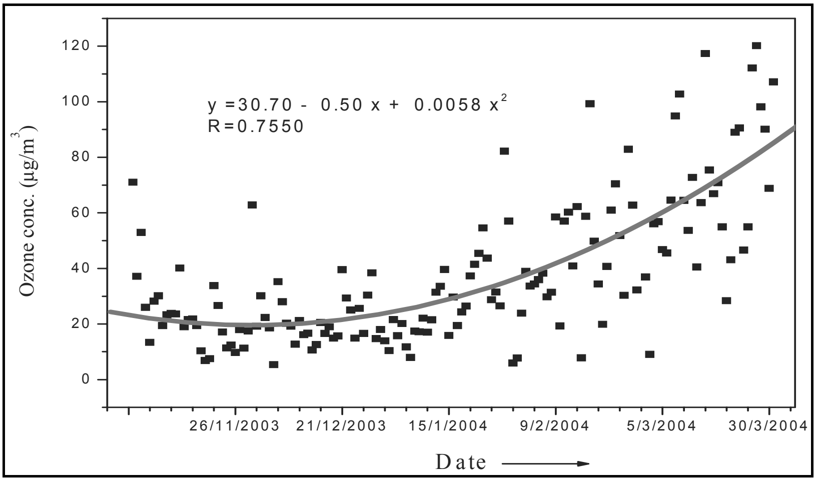

Fig. 2. Daily averaged ozone concentrations in November 2003-March 2004.

In general, ozone variation over the diurnal scale can provide insight into the interplay of emissions, and chemical and physical processes that operate on a diurnal cycle. Ozone levels tend to follow the solar radiation intensity, resulting in higher ozone concentrations during the daylight period (US EPA, 1996). A clear diurnal cycle of ozone can be seen from Fig. 3, which shows the diurnal statistics including maximum, mean, median, minimum, central 50%, 90%, and 98% of the data for each hour. The diurnal ozone variation observed in Jinan shows the typical pattern for urban areas, similar to that observed in other cities, such as Malaga (Duenas et al., 2002), Marseilles (Pont and Fontan, 2001), and Pune (Khemani et al., 1995). Except for 14:00, the median value in all hours of the day is lower than the mean value, which means that there is a larger percentage of data with low values in the monitoring period. The trends for each parameter were almost identical, increasing from 07:00 to afternoon, peaking at

13:00-15:00, and then decreasing until 19:00. Ozone levels are observed to be relatively steady at night, as there is no photoxidation of precursors causing ozone formation (Lal et al., 2000). However, from 02:00 the values continuously decreased at a low rate. This loss of ozone is attributed to in situ destruction of ozone by the well-known reaction between ozone and NO, and the surface deposition. Ozone maintained a relatively low value in nighttime, and the minimum value in the day was observed during early morning hours, near sunrise, although concentrations of NO and NO_2 were very high at that time (see Fig. 5). Chemical loss of ozone by nitrogen oxides is one reason for the low levels of ozone in the early morning, as well as meteorological factors (low temperature, light wind, development of a nocturnal inversion layer, etc.).

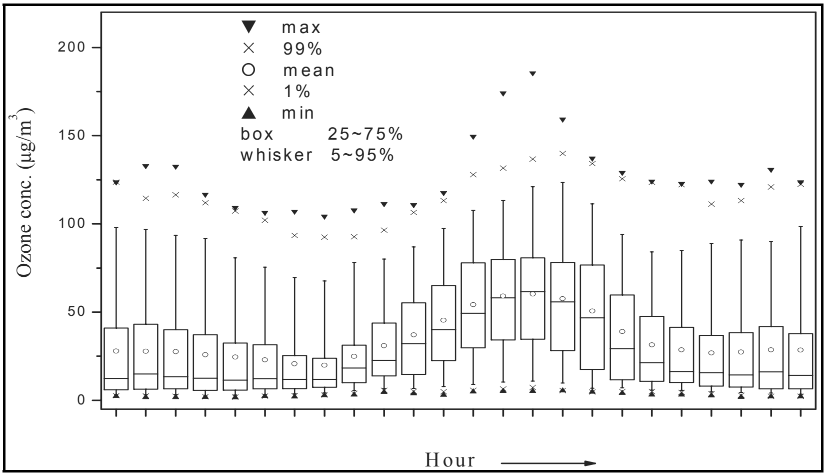

Fig. 3. Diurnal variations of ozone concentrations in November 2003-March 2004.

Correlation of ozone and solar radiation

Generally, ozone is formed primarily through the action of UV light, hence it would be expected that the variation of ozone concentrations follow solar radiation. Fig. 4 shows the varying concentration of ozone and intensity of solar radiation from 8:00 to 16:00 in February-March 2004. It can be seen that, the varying tendency of hourly averaged ozone concentrations was consistent with solar radiation, though it was delayed about two hours. So we calculated the correlation between solar radiation from 08:00 to 14:00 and ozone concentrations from 10:00 to 16:00, with the result that the correlation coefficient is 0.90. Solar radiation reaches its peak value at 12:00, and then decreases sharply in the afternoon, but ozone was almost constant from 14:00 to 15:00. In the afternoon, due to the higher temperature, variations of ozone precursors and other factors, ozone does not correlate with solar radiation as well as in the morning. When the correlation between solar radiation from 08:00 to 12:00 and ozone concentrations from 10:00 to 14:00 was calculated, a correlation coefficient of nearly 1.00 was obtained. The results suggest that solar radiation is one of the most significant influences for the daytime variations of ozone concentrations in Jinan, especially in the morning hours.

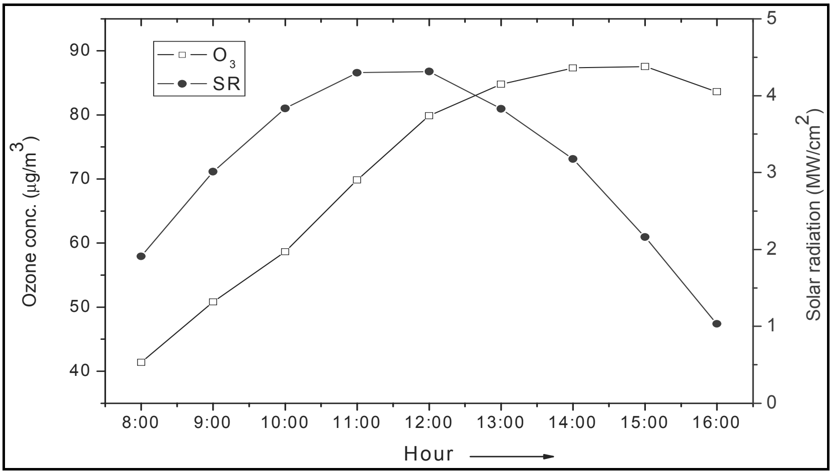

Fig. 4. Hourly variations of ozone and solar radiation from 8:00 to 16:00 in February-March 2004.

Correlation of ozone and nitrogen oxides

Ozone is a secondary pollutant, primarily influenced by two independent groups of factors: emission rates of its precursors and meteorological conditions. It is well known that the ozone generation rate relates to these factors in a complex way. Many studies have shown that NO and NO_2 are the principal precursors of ozone (e.g. Kleinman, 2005). According to the observed diurnal variation of ozone, the daytime data set brackets the photochemically active period of the day that affects the relationship between ozone and ozone precursors. Hence, the collected data for ozone, NO, NO_2, and NO_x (NO + NO_2) during the daylight 8 h (08:00-16:00) were compared.

The measurements showed that there were relatively low ozone levels accompanied by high NO_x levels during this period in Jinan. Fig. 5 shows the variations of hourly averaged ozone, NO, NO_2 and NO_x concentrations from 8:00 to 16:00 in February-March 2004. NO, NO_2 and NO_x all showed high levels in the morning hours, which was different from ozone, and NO reached its peak value earlier than NO_2 (09:00). High levels of primary pollutants (NO,

NO_2, NO_x) indicated the accumulation of locally emitted pollutants in the shallow boundary layer, especially pollutants emitted by the busy traffic in the morning. Ozone had an opposite trend mainly due to surface deposition and titration by NO in the night (Cheung and Wang, 2001). In the reaction of NO with ozone, NO is oxidized to NO_2 by ozone, so the maximum value of NO_2 appeared later than NO. Due to the photochemical processes in the daytime, ozone levels continuously increased from morning to afternoon, while NO_x levels continuously decreased from 09:00 to 15:00. Again there were obvious differences in the variations of ozone, NO, NO_2 and NO_x levels between clear days and cloudy days, variations in clear days were smooth, but on cloudy days were fluctuating. These differences also indicate that solar radiation is an important factor for the variation of ozone levels.

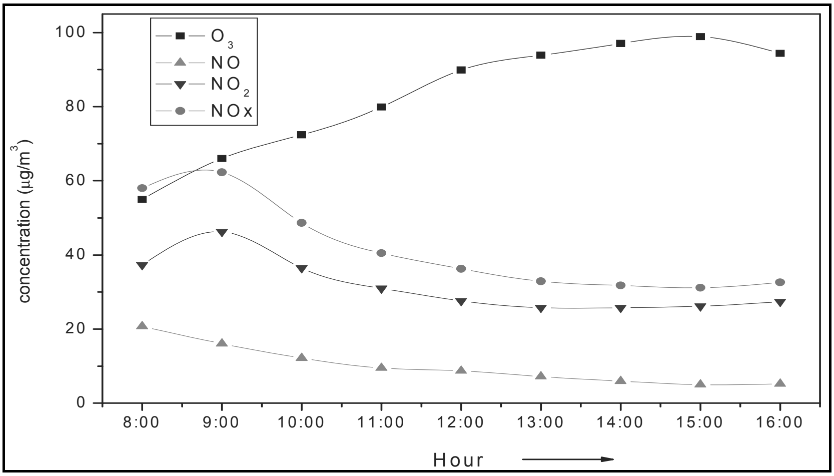

Fig. 5. Hourly variation of ozone, NO, NO_2 and NO_x concentrations from 8:00 to 16:00 in February-March 2004.

In the presence of solar energy, hν, NO_2 molecules may dissociate into NO and O, then the recombination of O and O_2 will occur when any other gaseous molecule (M, usually N_2, O_2, or H_2O in air) collides with them. On the other hand, the NO molecule may undergo a rapid reaction with ozone molecules. The balance of these reactions is dependent on the solar energy and NO_x available. The well-known photooxidation cycle can be expressed as follows:

$$NO_2 + h\nu \rightarrow NO + O(^3P) \quad (1)$$

$$O(^3P) + O_2 + M \rightarrow O_3 + M \quad (2)$$

$$NO + O_3 \rightarrow NO_2 + O_2 \quad (3)$$

In the absence of other processes to convert NO to NO_2 and assuming steady-state conditions, equations (1)-(3) can be written as:

$$[O_3] = K \times [No_2]/[NO] \quad (4)$$

$$K = \text{Rate coefficient for reaction (1)/Rate coefficient for reaction (3)} \quad (5)$$

It is assumed that K is constant with height in the lower troposphere (Syri et al., 2001; Barrett et al., 1998). In this study, the relationship between ozone level and NO_2/NO concentration ratio was analyzed and discussed according to the observed data at Jinan from 08:00 to 16:00 in February- March 2004. The correlation coefficient of linear fitting analysis is 0.86, showing that ozone level and NO_2/NO ratio correlated well with each other. Further analysis showed that the correlation coefficient in clear days is higher than in cloudy days. The results indicate that, in late winter and early spring local effects, such as reactions with pollutants from nearby emission sources, play a major role in determining ozone concentrations in Jinan.

There are topographic and meteorological reasons for the air pollution of Jinan. This city is in a basin location, surrounded by mountains and the Yellow River embankment. The unfavorable local topography makes pollutants difficult to disperse from the urban area. Considering the meteorology, the frequency of light wind and nocturnal inversion is very high in the winter in Jinan, thus the air is steady, which is quite unfavorable for the diffusion of the pollutants. Unreasonable distribution of urban industry is also an important factor for the air pollution of the city. The dominant wind direction in Jinan is northeast and southwest, but some industrial parks and several industrial manufacturers are located in these parts of the city (see Fig. 1), which aggravates the air pollution of the urban area.

CONCLUSION

In this study, we present the observational data of near surface ozone, NO, NO_2, and solar radiation during late 2003 and early 2004, in the urban area of Jinan, China. The obtained ozone values are relatively low, which indicates that the impact of using coal fired in winter on ozone pollution is not significant compared to meteorological factors. There is a very clear variation in the surface ozone levels throughout this period, and the lowest average ozone value is in the December. Ozone exhibited a typical diurnal variation pattern in urban Jinan, with the maximum in the afternoon and minimum in the early morning before sunrise.

Analysis of the variation of ozone and solar radiation in the daytime 8 h (08:00-16:00) shows that they have a similar tendency with ozone levels delayed about two hours. Clear vs. cloudy day studies showed that solar radiation has a significant influence on the daytime ozone concentrations in Jinan, especially in the morning hours.

Measurements showed that there were relatively low ozone levels accompanied with high NO_x levels during this period in Jinan. NO, NO_2 and NO_x all showed high levels in the morning hours, which indicates the accumulation of locally emitted pollutants in the shallow boundary layer, especially pollutants emitted by the busy traffic in the morning. Ozone had an opposite trend with NO_x mainly due to surface deposition and titration by NO in the night. Regression analysis of ozone level and NO_2/NO concentration ratio shows that they correlate well with each other, which indicates that in late winter and early spring local effects, such as reactions with NO_x from nearby emission sources, play a major role in determining ozone concentrations in Jinan.

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الأوزون السطحي أثناء الشتاء عند استعمال حرارة الفحم في منطقة جنان الحضرية - الصين

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خلاصة

تم قياس تركيز أول أكسيد النيتروجين و ثاني أكسيد النيتروجين و مستويات الإشعاع الشمسي قرب الأوزون السطحي خلال أواخر عام 2003 وأوائل عام 2004 في منطقة جنان الحضرية – الصين. لقد كانت مستويات الأوزون منخفضة نسبيا خلال فترة الدراسة، بينما NOx كانت عالية نسبيا مقارنة بمدن العالم الأخرى. لقد أظهر التحليل الإحصائي للأوزون تغير شكلي يومي مثالي للمناطق الحضرية في منطقة جنان، مع أعلى نسبة في أوقات الظهيرة، وأدنى نسبة في الصباح الباكر قبل شروق الشمس.

كما تبين من تحليل مستويات الأوزون ومعلومات الإشعاع الشمسي أثناء 8 ساعات خلال اليوم أن التغير في الأوزون يتبع الإشعاع الشمسي مع تأخير حوالي ساعتين، مع أن كليهما يظهران تصرفاً مماثلاً.

كما دلت دراسات أخرى على أن الإشعاع الشمسي هو عامل تأثيري هام لاختلاف تركيز الأوزون اليومي في منطقة جنان، خاصة في ساعات الصباح. ودل التحليل التراجعي لمستوى الأوزون ونسبة تركيز ثاني أكسيد النيتروجين/ أول أكسيد النيتروجين إن كليهما مرتبطان ارتباطا جيدا مع بعضهما البعض. كما اتضح أنه توجد تأثيرات محلية في أواخر أيام الشتاء وأوائل أيام الربيع تلعب دورا رئيسيا في تحديد تركيز الأوزون في منطقة جنان – الصين.