Significant increase of surface ozone at a rural site, north of eastern China

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Abstract

Ozone pollution in eastern China has become one of the top environmental issues and has been less studied. Quantifying the temporal trend of surface ozone helps to assess the impacts of the anthropogenic precursor reductions and the effects of emission control strategies. In this paper, ozone data collected at the Shangdianzi (SDZ) Regional Atmospheric Background Station from 2003 to 2015 are presented and analyzed in order to gain the variation trend of surface ozone in the most polluted region of China, north of eastern China or the North China Plain. A modified Kolmogorov–Zurbenko (KZ) filter method is performed on the maximum daily average 8-h (MDA8) concentrations of ozone to separate the contributions of different factors to the variation of surface ozone and remove the influences of meteorological fluctuations on surface ozone. Results reveal that the short-term, seasonal, and long-term component of ozone account for 36.4%, 57.6%, and 2.2% of the total

variance, respectively. The long-term trend indicates that the MDA8 undergone significant increase during 2003-2015, with an average rate of 1.13 ± 0.01 ppb/yr (R^2=0.92). It is found that meteorological factors did not significantly influence the long-term variation of ozone and the increase was completely resulted from the change of the emissions. Furthermore, there is no significant correlation between the long-term O_3 and NO_2 trends. This study suggests that emission changes in VOCs might have played a more important role in the increase trend of surface ozone at SDZ.

1. Introduction

Tropospheric Ozone (O_3) plays a key role in the oxidizing capacity of the atmosphere (Penkett, 1988) and acts as a greenhouse gas in terms of radiative forcing at the Earth's surface (IPCC, 2013). Moreover, it is an important precursor of OH radical, hence can exert indirect radiative forcing to the atmosphere by changing the lifetimes of some other greenhouse gases. Tropospheric O_3 originates from photochemical production in the troposphere and downward transport of stratospheric O_3 (Cooper et al., 2014; Monks et al., 2015). Ground-level O_3 is subject to in-situ chemical reactions and physical processes and is directly affected by precursor emissions, temperature, solar radiation and other meteorological factors.

Both observations (Oltmans et al., 2006) and model simulations (Hauglustaine and Brasseur, 2003) indicate that ground-level O_3 increased distinctly at northern mid-latitudes during the latter half of the 20th century, which is qualitatively in agreement with the increasing anthropogenic emissions of precursors. Anthropogenic emissions of O_3 precursor have been declining in Europe and North America while increasing in East Asia (Streets et al., 2001, Granier et al., 2011). The largest increase in NOx emissions is found in China and appears to have continued into the 21st century based on some emission inventories (Streets et al., 2001; Richter et al., 2005; Ohara et al., 2007; Mijling et al., 2013; Kurokawa et al., 2013). From 1990 to 2010, inconsistency occurred in the trends of surface O_3 in different regions. In the eastern US, surface O_3 decreased strongly in summer, was largely unchanged in spring, and increased in winter, while O_3 increases in the western US were the strongest in spring. Surface O_3 in East Asia was generally increasing (Cooper et al., 2014). It is found that ground-level O_3 in the Northeast Asian area, such as Japan (Lee et al., 1998) and Hong Kong (Chan et al., 2004; Wang et al., 2009) increased significantly from 1990s to 2000s. Enhanced variability of surface O_3, particularly the high level O_3, was reported for the Yangtze River Delta region in eastern China (Xu et al., 2008).

Dynamical factors may contribute to the long-term variations of

surface O_3. For example, the long-term increase of surface O_3 was found

to be related ~~with~~ the variability in stratosphere-to-troposphere transport

of O_3 (Ordonez et al., 2007; Hess and Zbinden, 2013; Lin et al., 2015a)

and changes in transport patterns (Pausata et al., 2012). Decadal

circulation shifts have played a key role in the autumnal ozone increase

and the absence of spring ozone change measured at Mauna Loa

Observatory (3.4 km altitude) over the subtropical Pacific Ocean during

1974-2012 (Lin et al., 2014). However, some studies (Brown-Steiner and

Hess 2011; Parrish et al., 2012; Lin et al., 2012; Oltmans, et al., 2013;

Derwent et al., 2015; Verstraeten et al., 2015) attribute O_3 increase in

some areas mainly to the transport of O_3 and related pollutants from the

continental China, where the emissions of O_3 precursors (NO_x and VOC)

had steadily increased (Ohara et al., 2007; Kurokawa et al., 2013). The

model results indicate that mean springtime ozone levels over western

North America in the most recent decade has increased by 5.9 ± 2.1 ppbv

compared to the 1980s, which could be attributed in part to rising Asian

ozone precursor emissions and global methane (Lin et al., 2015b).

Because of increasing emissions of O_3 precursors due to the sustained

economic growth in China (Zhang et al., 2007), fueled by favorable

photochemical conditions, China itself may have been suffering severe

photochemical pollution. Particularly over polluted regions of China, ~~the~~

long-term change of O_3 is expected. Indeed, limited ~~previous~~ studies

indicated that tropospheric O_3 ~~had been changing~~ significantly over some regions of China. Ding et al. (2008) analyzed O_3 data from the MOZAIC (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) program and obtained an ~~increase rate of~~ 2% per year ~~of the daytime~~ O_3 in the lower troposphere over Beijing and its surrounding areas for the period of 1995-2005. Xu et al. (2011) analyzed the TOR (Tropospheric Ozone Residue) data ~~during~~ 1979-2005 and found significant increasing trend~~s~~ of tropospheric O_3 over the North China Plain for all seasons except ~~for~~ winter, with a maximum rate of 1.10 DU per decade for summer. Wang et al. (2009) found that surface O_3 at a regional station in Hong Kong increased at an average rate of 0.58 ppb/yr from 1994 to 2007 and the ~~trend was~~ associated with ~~the~~ increase in tropospheric NO_2.

Information ~~about~~ the trends of surface O_3 in ~~China's different regions,~~ particularly those with high emissions of pollutants, is urgently needed. Due to the lack of long-term observations, it is ~~hardly possible~~ to gain reliable results about the long-term trends of surface O_3 in various regions of China. Recently, the long-term of surface O_3 in western China ~~is~~ reported by Xu et al. (2015), based on the observations at the Mt. Waliguan baseline station. So far, there has been no report of changes of surface O_3 levels in highly polluted eastern China during the recent decade. In this paper, we present the trend of surface O_3 in the north part of eastern China, based on the measurements from a rural site.

Furthermore, the relative contributions of meteorological factors and the change of anthropogenic emissions are investigated, which provide a further insight into potential causes of the observed trend of surface O_3.

2. Data and methods

2.1 Site and measurements

Surface O_3 and anscillary data were collected at the Shangdianzi (SDZ, 40.65 °N, 117.10 °E, 293.3m a.s.l.) station. SDZ is one of the regional Global Atmosphere Watch (GAW) stations, located about 100 km northeasterly to the urban area of Beijing. Within 30 km of the site, there are only small villages with sparse population and thus insignificant anthropogenic emission sources. The observation facilities of the station are situated on the south slope of a hill, which is surrounded by mountainous areas except the southwest sector. Fruit trees and corn are grown in the slope fields surrounding the site. It is shown that the observations of pollutants at SDZ could reflect regional scale air quality of North China (Lin et al., 2008; Xu et al., 2009).

The maximum daily average 8-h (MDA8) concentrations of O_3 were calculated from hourly averages of O_3 from October 2003 to June 2015 and are used in the following analysis. To facilitate the analysis, ambient NO_2 concentration and temperature measured at SDZ in the surface layer during the same time period were processed to obtain daily averages. Details about the observations and the quality assurance and quality

controll (QA/QC) procedures were described by Lin et al. (2008).

2.2   Analysis methods

It is well known that meteorology plays an important role in ozone formation and transportation. Ground-level ozone concentrations are strongly influenced by fluctuations of meteorological parameters. Thus, in the presence of fluctuations of meteorological parameters, it is difficult to distinguish the trend of ozone related to the change in emissions from meteorological impacts. In order to filter out or minimize the influence of meteorology on ozone levels, a method of Kolmogorov–Zurbenko (KZ) filter (Rao and Zurbenko, 1994) is used to separate data into short-term, seasonal, and long-term variations. The KZ filter is based on an iterative moving average that removes high frequency variations in the data. The procedure is briefly described below.

The KZ(m,n) filter is defined as n applications of a moving average of m points. The moving average can be expressed as

$$Y_i = \frac{1}{m} \sum_{j=-k}^{k} X_{i+j}, \qquad (1)$$

where m=2k+1, and the calculated Y_i becomes the input for the second pass, and so on.

Data filtered by KZ filter reserve information related with physical processes, whereas data treated by some other techniques may remove unwanted information but at the same time distort phenomena of interest. Eskridge et al. (1997) compared KZ filter with several methods, such as

wavelet transform, anomalies, etc. The results indicate that the KZ filter method has the same level of accuracy as that of the wavelet transform method. In addition, the magnitude of the long-term trend estimated by the KZ filter provides estimates with much higher (about 10 times) confidence than the other methods. However, the moving average of KZ filter with wide windows will dampen sharp breaks of variations. An adaptive filter based on KZ filter was developed that dynamically adjusted the length of moving according to the rate of change of the process (Zurbenko, 1996). As the rate of change increases, the length of modified KZ filter decreases. The detailed steps about the modified KZ filter applied in this paper were presented by Zurbenko (1996).

Rao et al. (1997) developed a method to separate different phenomena present in time series of both meteorological and ozone data having different characteristics such as long-term and short-term variations. Following the method, it is assumed that the time series of ozone can be partitioned as

$$O(t) = W(t) + S(t) + e(t), \qquad (2)$$

where $O(t)$ is the original time series, $W(t)$ is meso-scale and synoptic-scale variation, $S(t)$ is seasonal change, $e(t)$ is the long-term (trend) component. According to the results of Rao et al. (1997), when $KZ_{15,5}$ and $KZ_{365,3}$ filters are applied to the raw data, several influences could be removed and the actual variation of ozone at different scales

would be obtained.

$$W(t) = O(t) - KZ_{15,5}, \qquad (3)$$

$$S(t) = KZ_{15,5} - KZ_{365,3}, \qquad (4)$$

$$e(t) = KZ_{365,3} \qquad (5)$$

We followed the same method as Rao et al. (1997) in our filtering.

3. Results and discussion

3.1 General characteristics of yearly ozone distribution

The yearly statistics of MDA8 are presented in Fig. 1. Since the ozone observation at SDZ started in October 2003, the MDA8 statistics for 2003 are not showed in Fig. 1. It is noted that data from 2015 cover only the first 6 months. Although only the first 6 months records in 2015 are used for the statistics, the maximum of the MDA8s in this year exceeds 160 ppb, only second to that in 2012. The yearly average of MDA8 varies from 49.3 ppb to 60.2 ppb, with a very significant positive trend (1.05±0.14ppb/yr, R=0.93, P<0.0001) from 2004 to 2014. We also can find a similar fluctuation of the median value within the range of 43.3 ppb to 53.0 ppb, with a positive trend (0.62±0.20 ppb/yr, R=0.72, P<0.05)from 2004 to 2014. The MDA8 level was relatively stable during 2004-2006, with the maximum around 120 ppb. However, the maximum values exhibited a dramatic increase from 123 ppb in 2006 to 165 ppb in 2015, which seems to coincide with the increase of vehicles in eastern

China. For example, in Beijing, the vehicle fleet contained about 2.30 million in 2004, 2.88 million in 2006, 4.81 million in 2010, and 5.60 million in 2014 (data from: http://www.bjjtgl.gov.cn/jgj/ywsj/index.html). The changes of the maximum value of O_3 and vehicle numbers both have a dramatic increase trend during 2004-2015. Nevertheless, a clear long-term trend in the median or maximum value cannot be derived from the data shown in Fig. 1.

3.2 Ozone time series separated by KZ filter

Ozone time series (MDA8 value) from the SDZ site was separated using the method described in section 2.2. Fig. 2 shows the original time series of MDA8 values (Fig. 2a) and the time series of the separated short term, seasonal and long-term components (Figs. 2b-2d). The original MDA8 value exhibits a distinct seasonal variation, with overlapping of high frequency noise (Fig. 2a). Removing the short-term component (Fig. 2b) leads clearer seasonal cycles shown in Fig. 2c. As can be seen in Fig. 2c, there are evident double peaks of ozone during the summer in each year, which are not so obvious in the original time series (Fig. 2a). Generally, the double peaks occur in June and September respectively, and the dip in between occurs in July or August when relatively abundant rainfalls damps ozone formation and accumulation. Under the influence of the summer Asian monsoon, rainfalls in July and August at SDZ can amount to more than 40% of the whole year's rainfall. Fig. 2c

demonstrates also some irregularities in the seasonal cycle, particularly the year-to-year changes in the levels of annual maximum, minimum, and the dip. The seasonal fluctuations have to be accurately removed to get the long-term trend, as data for the trend analysis are required to be independent of season and normally distributed. The short-term component (Fig.2b) showed high frequency variations, ranging between -60 ppb and 70 ppb, which are composed of noise (or fluctuation) caused by meso-scale and synoptic-scale meteorological processes. Synoptic-scale events have a timescale from 2 days to 3 weeks, which could be removed by smoothing with the KZ filter for a window size of 15 days and 5 iterations. To further illustrate the short-term component, a quantile-quantile (QQ) plot of $W(t)$ is presented in Fig. 3. The QQ plot indicates that $W(t)$ basically obeys a normal distribution, with a mean value of 0.002 ppb, suggesting that the $KZ_{15,5}$ filter can effectively remove $W(t)$ from $O(t)$.

Through the previous steps and using the formulae (2)-(5), we obtained the long-term trend of MDA8 at SDZ, as shown in Fig. 2d. This long-term trend reveals a rapid increase of daily high value of surface ozone at the SDZ site in the last decade. It is noteworthy that the increase is not at a stable rate but with large inter-annual variations. Linear regression (not shown) indicates that the average increase rate is $1.13\pm$ 0.01ppb/yr ($r^2 = 0.92$). Previous study by Ding at al. (2008) using

MOZAIC data obtained a yearly increase of 2% (about 1 ppb/yr) of O_3 in the boundary layer around Beijing during 1995-2005, which agrees well with our result. Therefore, the greater Beijing area, probably the North China Plain, has been suffering a rapid ozone increase for the last two decades.

In view of the air pollution problems, the central government of China issued a revised National Ambient Air Quality Standard (CNAAQS, GB 3095-2012) in 2012, which has taken effect across the country since 1 January 2016 and sets the MDA8 O_3 limits to 100 $\mu g/m^3$ (51.0 ppb) and 160 $\mu g/m^3$ (81.6 ppb) for national reserve areas and residence/commercial areas, respectively. As can be seen in Fig. 2a, O_3 exceedance would be quite often in the warm seasons if the new CNAAQS had been implemented.

We also examined the contributions of different components to the total variance of MDA8, which is calculated from the unfiltered data. The contributions of the short-term and seasonal components to the total variance are about 36.4% and 57.6%, respectively. The long-term component accounts for only 2.2% of the total variance. The covariance terms sums to less than 4% of the total variance, indicating an effective separation of different components. The long-term component makes only a much smaller contribution than the other two components, confirming the necessity to clearly separate the short-term and seasonal

variations from the data to obtain the long-term trend.

3.3 Cause analysis

The long-term trend of ozone concentrations can be caused by the changes of both pollutant emissions and related meteorological variables. Climate variability and circulation shifts may lead to long-term changes of O_3 as discussed in Lin et al. (2014, 2015a, 2015b). To assess the influence of precursor emissions on the ozone trend, the meteorological and chemical impacts have to be separated. However, both meteorological and chemical impacts are complicated, not to mention the interactions among meteorology, precursor emissions, and photochemical reactions. Therefore, a clear separation of meteorological and chemical impacts is hardly possible purely based on observational data. Nevertheless, it is worthy to try to make attribution of the O_3 trend to precursor emissions and other causes.

Although many meteorological variables can influence photochemical formation of O_3, temperature is the prevailing one. The increase of temperature can increase reaction rates, emissions of biogenic VOCs, and reduce wind speeds, etc. (Lin et al., 2001; NRC, 1991; Pusede et al., 2015). In a certain region, temperature is also closely related with intensity of solar radiation, which plays a critical role in photochemical formation of O_3. Thus, we took temperature as a key meteorological parameter and investigated the relationship between O_3

and temperature, with the hope to obtain the influence of emission changes on the long-term trend of O_3. The first step of our effort is to divide the time series of temperature into three components in formula (2), just as done for that of MDA8 (Fig.2). The results of the different components of temperature are given in Fig. 4. Unlike the trend of MDA8 of O_3, the long-term component for temperature in SDZ shows a slight   decrease trend ($R^2=0.015$) (Fig. 4d) and this long-term component accounts only for 0.16% of the total variance of temperature.

The original data of O_3 and temperature are less correlated ($R^2=0.50$, P<0.0001), presumably due to the strong influence of the short-term component. Fig. 5 compares the derived seasonal cycles of the daily mean temperature (from Fig. 4c) and the MDA8 of ozone (from Fig. 2c). A coincidence is evident between both seasonal cycles. However, there is also a distinct phase lag of the seasonal cycle between O_3 and temperature, due to the influence of other processes on the O_3 level. Rao et al. (1995) found that similar phase lag of about 3 weeks in the data from the northeastern United States. In our case, the linear correlation between O_3 and temperature becomes strongest ($R^2=0.83$, P<0.0001) when the temperature data are lagged by 17 days (Fig. 6).

When only considering the influence of temperature, the seasonal- and long-term components of O_3 could account for 93% of the total variance at the Cliffside Park, New Jersey (Rao and Zurbenko, 1994).

While in our case, it just accounts for 83% (see R^2 in Fig. 6). We tried to add more meteorological factors that could affect O_3 production, such as solar radiation, relative humidity. However, the correlation was only improved by no more than 0.5%. This implies that the changes in emissions might have a more important influence on surface O_3 at SDZ than that at Cliffside Park. This view is consistent with the rapid increase of anthropogenic emission in China (particularly the North China Plain) during the last decade (Mijling et al., 2013).

Assuming that the residual of the total variance of O_3 after subtracting the contribution related temperature was all caused by pollutant emissions, the long-term trend of O_3, attributable to changes in emissions, can be determined by performing a linear regression between time and the noise-free, temperature-independent O_3 values ($\varepsilon(t)$), which are derived using function (6).

$$O_{kz}(t)=aT_{kz}(t+17)+b+\varepsilon(t) \qquad (6)$$

where $O_{kz}(t)$ is the filtered O_3 concentration, $T_{kz}(t+17)$ is the filtered temperature lagged by 17 days, a and b are fitted parameters, $\varepsilon(t)$ are the residuals of the relationship. Here, $\varepsilon(t)$ reveals changes in ozone attributable to changes in emissions.

Fig. 7 shows the time series of the noise-free and temperature-independent O_3, which is basically equal to the long-term component of O_3 only under the influence of emission changes. Most of

the data in Fig. 7 are within the range of 95% confidence prediction band

except for some special cases happened in summertime. In summer,

temperature is not the dominant restricting factor for O_3 production

compared to other factors, such as rainfall and precursor concentrations.

Substantial negative influences occurring in 2005 and 2006 can be

explained by stronger impact of Asian summer monsoon on surface

ozone (Lin et al., 2008). The results in Fig. 7 indicate that the influence

of emission has been varying substantially but with an average increase

rate of 1.19 ± 0.03 ppb/yr. This increase rate is very close to the average

long-term trend of MDA8 of O_3 (1.13 ± 0.01 ppb/yr) in Fig. 2d, implying

that the increase of O_3 during 2003-2015 was mainly resulted from the

emission changes and the meteorological factors had only a tiny negative

influence. Jaffe and Ray (2007) also found that the temperature change

had little influence on long-term ozone trends in the western US.

Some studies suggested that the trends of surface O_3 at the similar

latitude as SDZ could be attributed partly to the reduced titration by NO

(Chou et al., 2006; Itano et al., 2007). In order to assess the effect of

changing NO titration on the long-term trend of O_3, we examined the

long-term measurements of NO_2 at SDZ during 2004-2015. A

comparison of the long-term trend of O_3 with that of NO_2, which was

also extracted using the previous methods, is displayed in Fig. 8. The

evolution of the NO_2 trend can be divided into three stages, i.e., a

substantial decrease of NO_2 occurring during the first 3 years, followed

by a small increase during 2007-2010, and finally a gradual decrease

during 2011-2015. The large decrease of NO_2 during 2004-2006

corresponded to the control of coal consumption around Beijing,

especially for the Olympic Games in 2008 (Zhang et al., 2010; Gao et al.,

2011) and to the relocation of the Capital Steel and Iron Company, one of

the large industrial source. The NO_2 increase from 10.2 ppb to 13.5 ppb

between 2007 and 2010 corresponded the rapid increase in numbers of

vehicles in Beijing from 3.1 million to 4.8 million

(http://www.bjjtgl.gov.cn/jgj/ywsj/index.html). From 2011 to 2015, the

new standard for vehicle emissions and measures for reduction of NOx

emission from power plants were implemented, which may have helped

to reduce the NO_2 concentration. The long-term trends of O_3 and NO_2

given in Fig. 7 do not show any coincidence. Therefore, it is nearly

impossible that the reduced NO titration had led to the increase of surface

O_3 at SDZ. Previous studies (Ge et al., 2010; Ge et al., 2012) showed that

the ozone production efficiency at SDZ varied in from 0.2 to 21.1, with

an average of 4.9, implying that ozone formation at SDZ could be more

sensitive to VOCs than to NOx. Accordingly, we believe that the changes

of VOCs emission and the ratio VOCs/NOx might have caused the

increase of surface O_3 observed at SDZ. Unfortunately, no systematic

VOCs observations are available from the SDZ site so that we cannot

prove our view using measurements from SDZ. However, an large increase in the anthropogenic emissions of non-methane hydrocarbon (NMHC) can be inferred from the Multiresolution Emission Inventory for China (MEIC) (http://www.meicmodel.org) for Beijing during 2004-2012, which supports our view, although the emission data are questioned by a recent study (Wang et al., 2015).

4. Summary

We separated the time series of maximum daily average 8-h (MDA8) concentration of surface O_3 observed at SDZ during 2003-2015 into various spectral components using a modified KZ filter. This separation has led to a better understanding of the variation of surface O_3 at the site and its relationships with the meteorological and precursor variables, enabling us to unravel the trend of O_3 from the original data containing noises and seasonality, and estimate the contribution of changes of precursor emissions to the trend. Our analysis reveals that the short-term, seasonal, and long-term components of O_3 data from the SDZ site accounted for 36.4%, 57.6%, and 2.2% of the total variance, respectively.

It is found that the MDA8 of O_3 at the site north of eastern China has undergone a significant increase during 2003-2015, at a average rate of 1.13 ± 0.01 ppb/yr . Together with the reported yearly increase of 2% in the lower tropospheric O_3 around Beijing for 1995-2005 (Ding et al., 2008), we conclude that the north part of eastern China (i.e., the North

China Plain) may have been suffering a rapid increase in the O_3 level for at least two decades. By eliminating the influence of air temperature, we find that the observed increase of surface O_3 during 2003-2015 was mainly induced by the emission changes and the meteorological factors exerted only a tiny negative influence. Our result also indicates that changes of VOCs emissions might play a more important role in the O_3 increase than the effect of NOx.

Because fine particles pollution has been very severe in eastern China, the government has been implementing several measures to control $PM_{2.5}$, including reductions of both NOx and VOCs. This will have a potential risk to additional O_3 increase in case a VOCs/NOx ratio more favorable for ozone production is reached. Thus, further studies are needed to trace ozone trend and its influence in eastern China.

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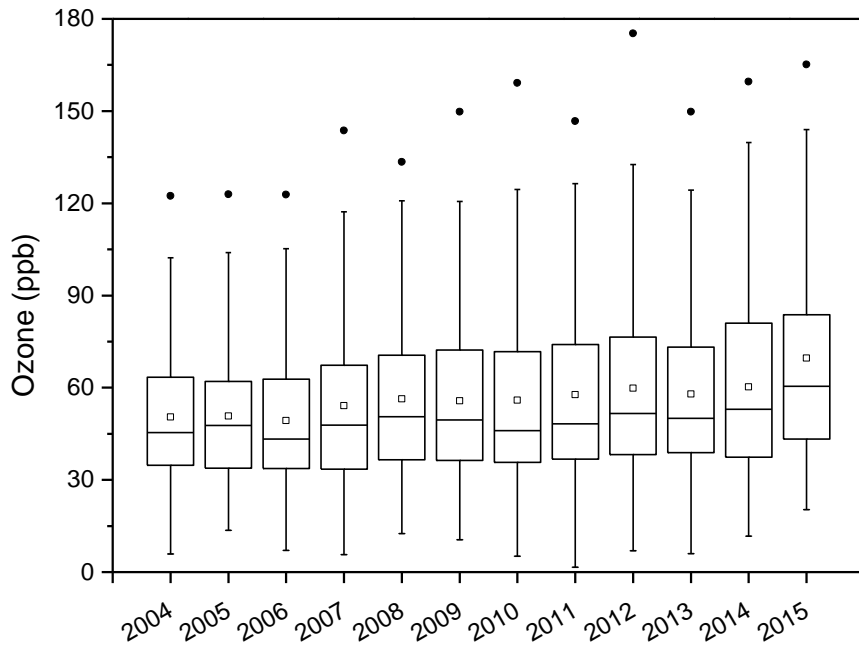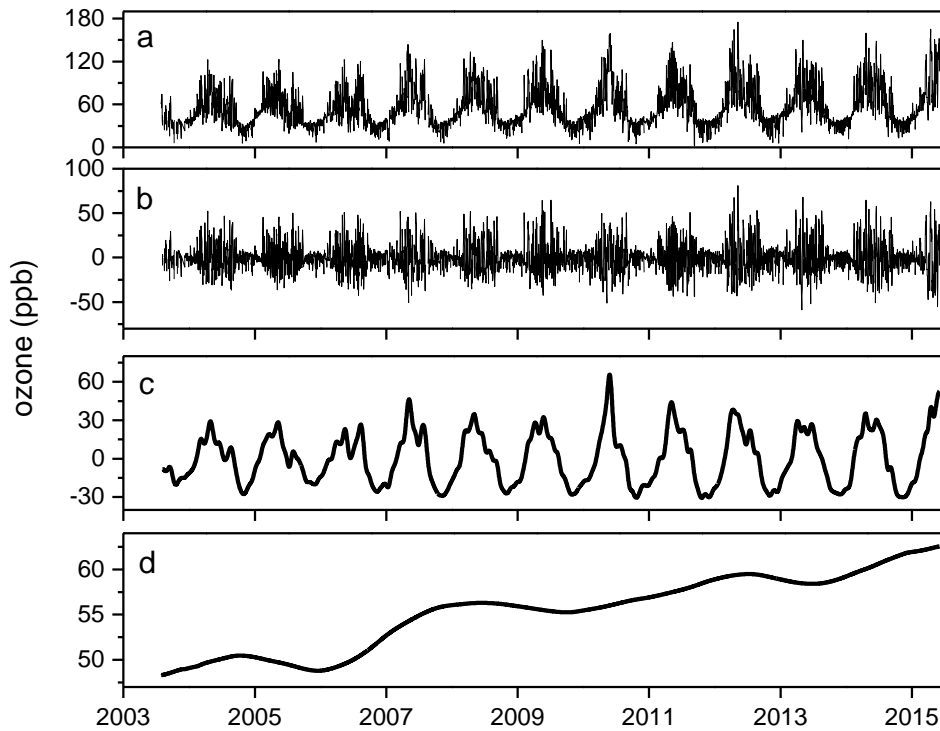
1

2　Fig 1. Percentile-box plot of yearly statistics of MDA8 values of ozone at SDZ,

3　2004-2015. Box depicts interquartile range and median; the square depicts the mean;

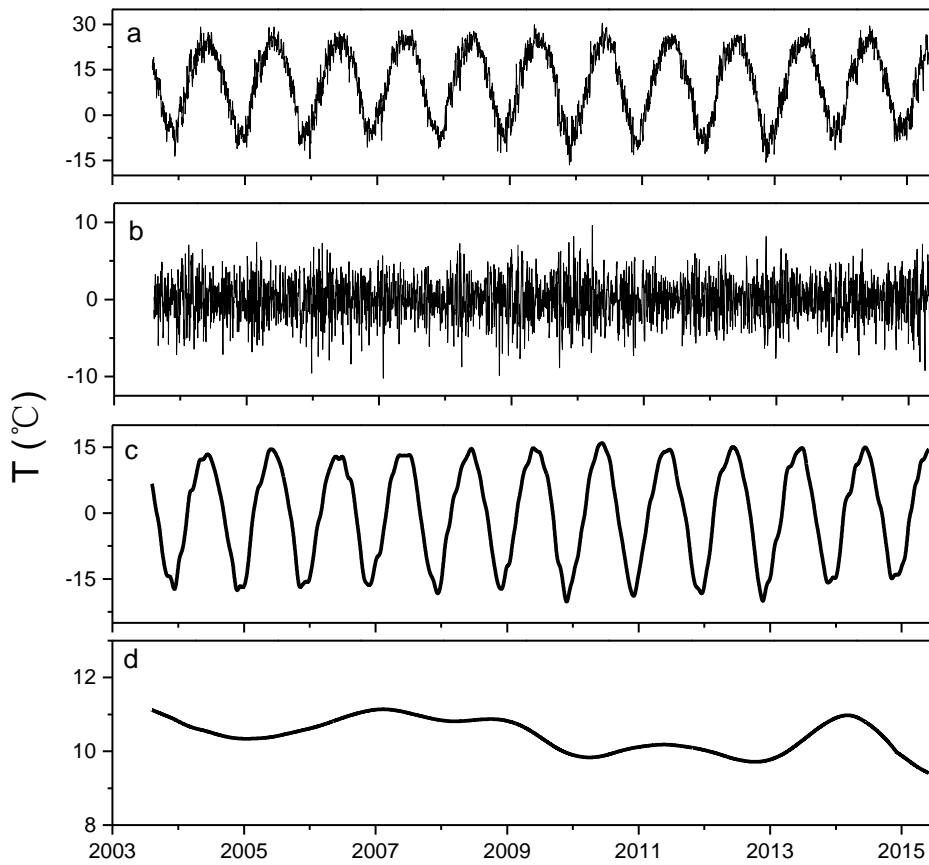4　whiskers depict 10th and 90th percentile; dot depicts maximum values.

5

6　Fig 2. Separated time series of MDA8 values of ozone at SDZ: (a) the original data;

7　(b) the short-term component, $W(t)$; (c) the seasonal component, $S(t)$; (d) the
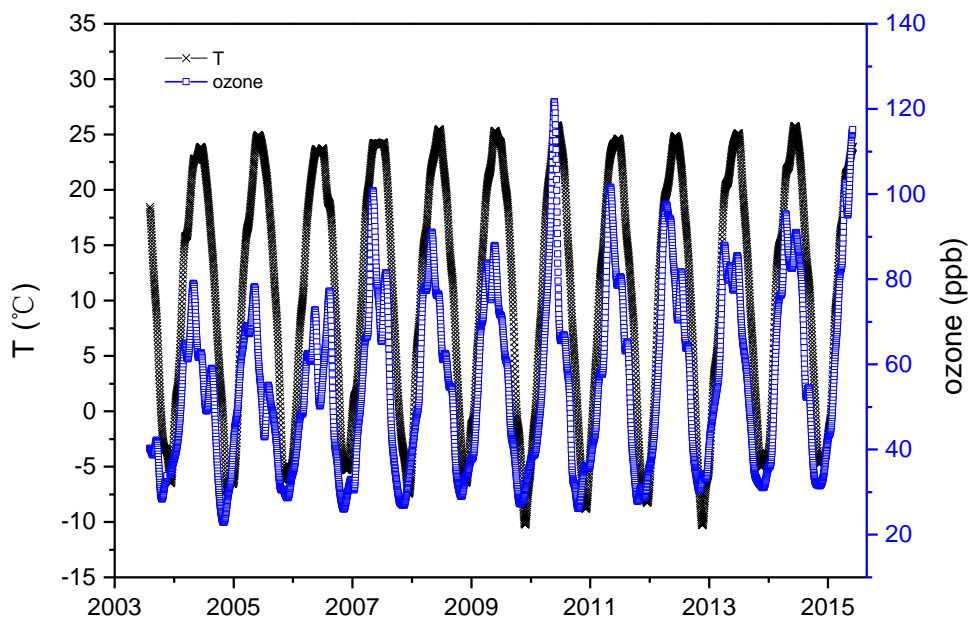
8　long-term component, $e(t)$.
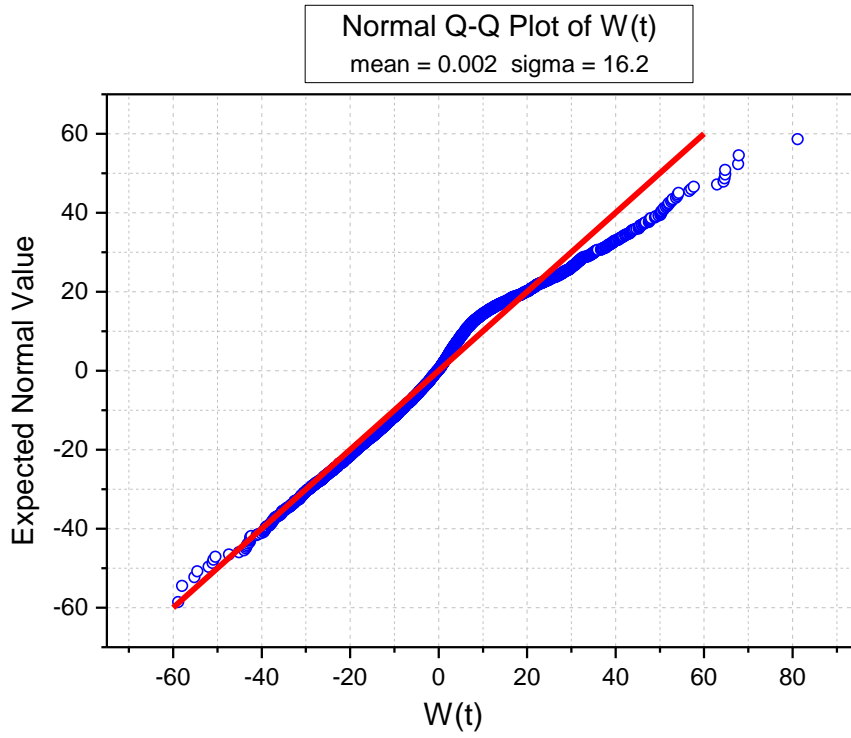
Fig 3. QQ plot of the short-term component W(t) for ozone.

Fig 4. Separated time series of daily mean values of temperature at SDZ: (a) the

original data; (b) the short-term component, W(t); (c) the seasonal component, S(t);
(d) the long-term component, e(t).

Fig 5. Results of the daily mean temperature and the MDA8 value of ozone after the application of $KZ_{15,5}$ filter to the original time series. The results indicate the sum of the seasonal and long-term components.
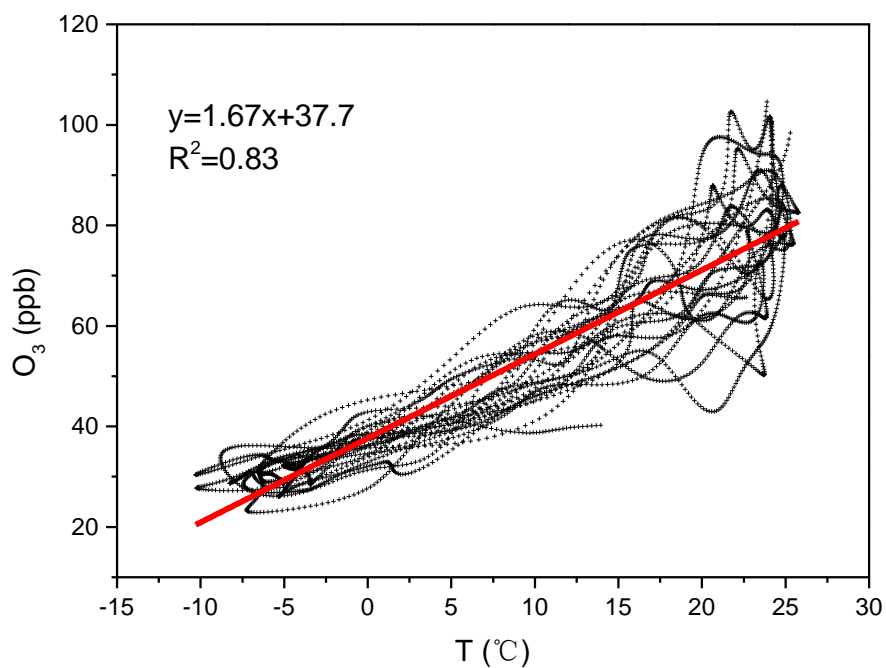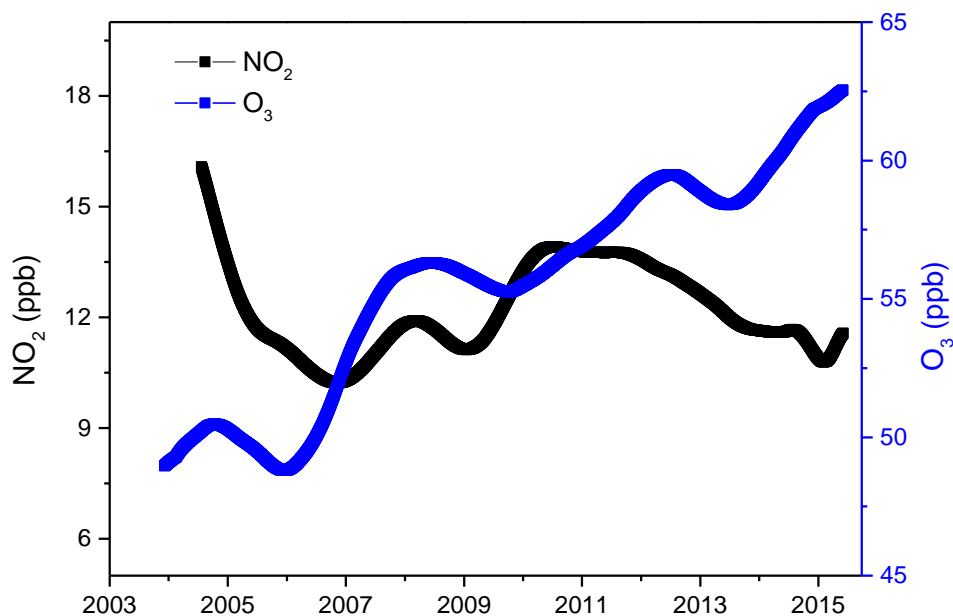

Fig 6. Linear regression fit on the filtered daily maxima of temperature and ozone.

1   Temperature data are lagged by 17 days to ozone data.
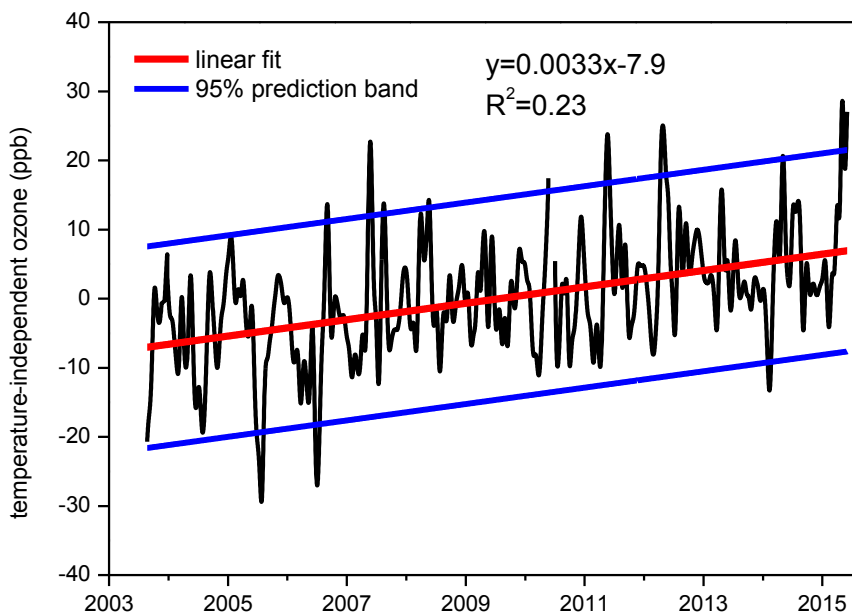
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3   Fig 7. Time series of the noise-free and temperature-independent ozone. The red line
4   is a linear fit and the blue lines are the 95% confidence prediction band.

5

6   Fig 8. Long-term trends of NO_2 and the MDA8 ozone value calculated by $KZ_{365,3}$.
7