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

Z. Q. Ma[1,2], J. Xu[1,2], W. J. Quan[2], Z. Y. Zhang[2], W. L. Lin[3], X. B. Xu[4]

[1] Institute of Urban Meteorology, China Meteorological Administration, Beijing, China

[2] Environmental Meteorology Forecast Center of Beijing-Tianjin-Hebei, Chinese Meteorological Administration, Beijing, China

[3] Meteorological Observation Centre, China Meteorological Administration, Beijing 100081, China

[4] Key Laboratory for Atmospheric Chemistry of CMA, Chinese Academy of Meteorological Sciences, Beijing 100081, China

* Correspondence to: W. L. Lin (linwl@camscma.cn)

Abstract

Ozone pollution in eastern China has become one of the top environmental issues. Quantifying the temporal trend of surface ozone helps to assess the impacts of the anthropogenic precursor reductions and the likely effects of emission control strategies implemented. In this paper, ozone data collected at the Shangdianzi (SDZ) Regional Atmospheric Background Station from 2003 to 2015 are presented and analyzed to obtain the variation in the 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 was performed on the maximum daily average 8-h (MDA8) concentrations of ozone to separate the contributions of different factors from the variation of surface ozone and remove the influence 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 has undergone a significant increase in the period of 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 may be completely attributed to changes in 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 observed increase 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 the OH radical, hence changes in its abundance can exert indirect radiative forcing by altering the lifetimes of some other greenhouse gases. Tropospheric O_3 originates from photochemical production within the troposphere and the 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.

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 precursors 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 NO_X ($NO + NO_2$) emissions is found in China and appears to have continued into the 21st century based on 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, surface O_3 in different regions showed inconsistent trends. In the eastern US, surface O_3 was found to decrease strongly in summer, be largely unchanged in spring, and increase in winter, while O_3 increases in the western US were the strongest in spring (Monks et al., 2015). In East Asia, surface O_3 was generally found to be increasing (Cooper et al., 2014). For example, ground-level O_3 in the Northeast Asian region, e.g. Japan (Lee et al., 1998) and Hong Kong (Chan et al., 2004; Wang et al., 2009) increased significantly from the 1990s to 2000s. Enhanced variability of surface O_3, particularly an increasing trend in the monthly highest 5% of the ozone mixing ratios, was reported for the Yangtze River Delta region in eastern China (Xu et al., 2008).

Dynamic 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 to the variability in the 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 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 in the period of 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 the increase of O_3 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) has steadily increased (Ohara et al., 2007; Kurokawa et al., 2013). Studies by Lin et al. (2015b) indicate that mean springtime ozone level 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. 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 has likely being experiencing severe photochemical pollution. Due to his industrialization, increases are expected in some regions. Indeed,

limited studies have indicated this is indeed the case. 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 a 2%/yr rate of increase in 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 over the period 1979-2005 and found a significant upward trend in tropospheric O_3 over the North China Plain for all seasons except for winter, with a maximum rate of increase 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 they associated the trend with an increase in tropospheric NO_2.

Information regarding the trends in surface O_3 concentrations in the different regions of China, particularly those with high anthropogenic emissions rates, is urgently needed. Due to a lack of long-term observations, it is difficult to gain reliable results about the long-term trends of surface O_3 in various regions of China. Recently, the long-term trend of surface O_3 in western China was 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 trends in surface O_3 in the North China Plain, based on the measurements

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

2. Data and methods

2.1 Site and measurements

Surface O_3 and ancillary 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 northeast of suburban of Beijing. The 30 km radius surrounding the site contains only small villages with a sparse population and insignificant anthropogenic emission sources. The observation facilities are situated on the south slope of a hill, which is surrounded by mountainous areas except in the southwest sector. Fruit trees and corn are grown in the fields surrounding the site. Previous studies (Lin et al., 2008; Xu et al., 2009) suggest that the observations of pollutants at SDZ reflect the regional scale air quality of North China.

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 of the observations and the quality assurance and quality control

(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 transport. Ground-level ozone concentrations are strongly influenced by fluctuations of meteorological parameters. Therefore, it is difficult to distinguish the trend of ozone related to the change in emissions from that related to meteorological impacts. In order to filter out or minimize the influence of meteorology on ozone levels, a method called 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 method 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

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

where $m=2k+1$, and the calculated Y_{i} becomes the input for the second pass, and so on.

Data filtered by the KZ filter preserve information related to 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 the KZ filter method with several others, such as wavelet transform, anomalies, etc. and demonstrated that the KZ

filter has the same level of accuracy as the wavelet transform method. In addition, the magnitude of the long-term trend estimated by the KZ filter provides estimates with approx. 10 times higher confidence than the other methods. However, the width moving average (*m*) of the KZ filter with wide windows will dampen sharp breaks of variations. Based on the KZ filter, an adaptive filter was developed by Zurbenko et al. (1996), which dynamically adjusts the width of *m* according to the rate of change of the process. As the rate of change increases, the *m* decreases. The modified KZ filter method is applied in this paper. More details on this method can be found in Zurbenko et al. (1996).

Rao et al. (1997) developed a method to separate different phenomena present in time series of both meteorological and ozone data, which have 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 the meso-scale and synoptic-scale variation, S(t) is the seasonal change, e(t) is the long-term (trend) component. Rao et al. (1997) found that 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), S(t) and e(t) can be calculated using the following formulae.

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

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

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

The Rao et al. (1997) method was implemented in this work.

3. Results and discussion

3.1 General characteristics of yearly ozone distribution

Yearly MDA8 statistics were calculated for 2004-2015 and are presented in Fig. 1. Since the ozone observation at SDZ commenced in October 2003, no reliable yearly MDA8 statistics can be obtained. 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 MDA8 values in this year exceeded 160 ppb, only second to that in 2012. The yearly average of MDA8 varied from 49.3 ppb to 60.2 ppb during 2004-2014, with a highly significant positive trend (1.05 ± 0.14 ppb/yr, $R=0.93$, $P<0.0001$). We also observed 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 a maximum approx. 120 ppb. However, the annual maximum value exhibited a dramatic increase from 123 ppb in 2006 to 165 ppb in 2015. This increase coincided with an increase of in the size of the vehicle fleet

in eastern China. For example, in Beijing, the number of registered vehicles was 2.30 million in 2004, 2.88 million in 2006, 4.81 million in 2010, and 5.60 million in 2014 (http://www.bjjtgl.gov.cn/jgj/ywsj/index.html). Both the maximum O_3 value and the vehicle number increased dramatically in the period 2004-2015. Nevertheless, it is not possible to derive a reliable long-term trend in the median or maximum value solely from the data shown in Fig. 1. Nor can we directly attribute the observed changes in surface O_3 to the increase in vehicles.

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 exhibits a distinct seasonal variation, with an overlapping of high frequency noises (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 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 rainfall damps ozone formation and accumulation. Under the influence of the

summer Asian monsoon, rainfall in July and August at SDZ can amount to more than 40% of the annual rainfall. Fig. 2c also demonstrates some irregularities in the seasonal cycle, particularly the year-to-year changes in the levels of annual maximum, annual minimum, and the dip between the double peaks. 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 effectively removed 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 the 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 during 20004-2015 was 1.13±0.01ppb/yr (r^2 = 0.92). Previous work by Ding et al. (2008) using MOZAIC data obtained a yearly increase of 2% (about 1 ppb/yr) of O_3 in the boundary layer around Beijing in the period of 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 μg/m^3 (46.7 ppb) and 160 μg/m^3 (74.7 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 was 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 sum 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 the ozone concentration 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, apportionment of the O_3 trend to precursor emissions and other causes are worthy of further study.

Although many meteorological variables can influence the photochemical formation of O_3, temperature is the prevailing one. The increase of temperature can speed up the photochemical reactions, strengthen the emissions of biogenic VOCs, and reduce wind speed, etc. (Lin et al., 2001; NRC, 1991; Pusede et al., 2015). In certain regions, temperature is also closely related to the intensity of solar radiation,

which plays a critical role in the 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 initial step was 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 at 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 unfiltered 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 O_3 (from Fig. 2c). A similarity 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 a 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 the 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 increases in anthropogenic emissions 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 with 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, $\varepsilon(t)$ is the residual of the relationship. Here, $\varepsilon(t)$ reveals the 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 a 95% confidence interval band except for some special cases in the summer months. 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 O_3 (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 in the period of 2003-2015 could be mainly attributed to 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 in the period of 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 in the period of 2007-2010, and finally a gradual decrease in the period of 2011-2015. The large decrease of NO_2 in the period of 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, which was one of the largest industrial sources in Beijing. The NO_2 increase from 10.2 ppb to 13.5 ppb between 2007 and 2010 corresponded with the rapid increase in the number 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 NO_X 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 emissions and the ratio VOCs/NOx might have caused the increase of surface O_3 observed at SDZ. Unfortunately, no systematic VOC observations are available from the SDZ site so that we cannot prove this supposition conclusive. However, a 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 in the period of 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 in the period of 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 to 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 in the period of 2003-2015, at an average rate of 1.13±0.01 ppb/yr. Together with the reported yearly increase rate of 2% in the lower tropospheric O_3 around Beijing in the period of 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 found that the observed increase of surface O_3 in the period of 2003-2015 was mainly induced by the emission changes and the meteorological factors exerted only a tiny negative influence. Our result also indicates that the change of VOCs emissions might have played a more important role in the O_3 increase than the effect of NO_X.

Because fine particulate pollution has been very severe in eastern China, the central government of China has implemented several measures to control $PM_{2.5}$ Pollution, including reductions of both NOx and VOCs. This, however, risks further O_3 increases as a VOCs/NOx ratio more favorable to ozone production may be reached. Thus, further studies are needed to trace the ozone trend and its influence in eastern China.

Acknowledgements

The authors would like to thank the staff of the Shangdianzi Station for their excellent work. This research is supported by the National Science Foundation of China

(41475135, 41330422), Beijing Natural Science Foundation (8132025, 8152018), CCSF201505. We also thank the referees and editor for their constructive suggestions.

References

1. Brown-Steiner, B., Hess, P.: Asian influence on surface ozone in the United States: A comparison of chemistry, seasonality, and transport mechanisms, J. Geophys. Res., 116, D17309, 2011, doi:10.1029/2011JD015846.
2. Chan, C. Y., Chan, L. Y., and Harris, J. M.: Urban and background trend in 1984-1999 subtropical Hong Kong, South China, Ozone: Science and Engineering, 25, 513-522, doi:10.1080/01919510390481829, 2003.
3. Chou, C. C. K., Liu, S. C., Lin, C. Y., Shiu, C. J., and Chang, K. H.: The trend of surface ozone in Taipei, Taiwan, and its causes: Implications for ozone control strategies, Atmos. Environ., 40, 3898-3908, doi:10.1016/j.atmosenv.2006.02.018, 2006.
4. Cooper O. R., Parrish D. D., Ziemke J., Balashov N. V., Cupeiro M., Galbally I. E., Gilge S., Horowitz L., Jensen N. R., Lamarque J.-F., Naik V., Oltmans S. J., Schwab J., Shindell D. T., Thompson A. M., Thouret V., Wang Y., and Zbinden R. M.: Global distribution and trends of tropospheric ozone: An observation-based review. Elementa: Science of the Anthropocene, 2, 1-28, doi: 10.12952/journal.elementa.000029, 2014.
5. Derwent, R. G., Utembe, S. R., Jenkin, M. E., Shallcross, D. E.: Tropospheric ozone production regions and the intercontinental origins of surface ozone over Europe, Atmospheric Environment, 112, 216-224, 2015
6. Ding, A. J., Wang, T., Thouret, V., Cammas, J. P., Nédélec, P.: Tropospheric ozone climatology over Beijing: analysis of aircraft data from the MOZAIC program, Atmos. Chem. Phys., 8, 1-13, doi:10.5194/acp-8-1-2008, 2008.
7. Eskridge, R.E., Ku, J. Y., Rao, S. T., Porter, P. S., Zurbenko, I. G.: Separating Different Scales of Motion in Time Series of Meteorological Variables, Bulletin of the American Meteorological Society, 78, 1473-1483, doi: 10.1175/1520-0477(1997)078<1473:SDSOMI>2.0.CO;2, 1997.
8. Gao, Y., Liu, X., Zhao, C., Zhang, M., Wang, Y.: Emission controls versus meteorological conditions in determining aerosol concentrations in Beijing during the 2008 Olympic Games, Atmos. Chem. Phys., 11, 12437-12451, 2011.
9. Ge, B.Z., Xu, X.B., Lin, W.L., and Wang, Y.： Observational study of ozone production efficiency at the Shangdianzi Regional Background Station (in Chinese with English abstract). Environ. Sci. 31, 1444-1450, 2010.
10. Ge, B.Z., Xu, X.B., Lin, W.L., Li, J., Wang, Z.F.: Impact of the regional transport of urban Beijing pollutants on downwind areas in summer: ozone production efficiency analysis, Tellus B, 64, 17348, DOI: 10.3402/tellusb.v64i0.17348, 2012.
11. Granier, C., Bessagnet, B., Bond, T., D'Angiola, A., van der Gon, H. D., Gregory J. Frost, G. J., Heil, A., Kaiser, J. W., Kinne, S., Klimont, Z., Kloster, S.,

Lamarque, J. F., Liousse, C. Masui, T., Meleux, F., Mieville, A., Ohara, T., Raut, J. C., Riahi, K., Schultz, M. G., Smith, S. J., Thompson, A., Aardenne, J. V., Werf, G. R., Vuuren, D. P.: Evolution of anthropogenic and biomass burning emissions of air pollutants at global and regional scales during the 1980–2010 period. Climatic Change, 109, 163-190. doi:10.1007/s10584-011-0154-1,2011.

12. Hauglustaine, D. A., Brasseur, G.: Evolution of tropospheric ozone under anthropogenic activities and associated radiative forcing of climate, J. Geophys. Res., 106, 32337-32360, 2003.
13. Hess, P. G., Zbinden, R.: Stratospheric impact on tropospheric ozone variability and trends: 1990e2009. Atmos. Chem. Phys. 13, 649-674. http://dx.doi.org/10.5194/acp-13-649-2013, 2013.
14. Itano, Y., Bandow, H., Takenaka, N., Saitoh, Y., Asayama, A., Fukuyama, J.: Impact of NOx reduction on long-term ozone trends in an urban atmosphere, Sci. Total Environ, 379, 46-55, doi:10.1016/j.scitotenv.2007.01.079, 2007.
15. Jaffe, D., and Ray, J.: Increase in surface ozone at rural sites in the western US, Atmos. Environ., 41, 5452-5463, doi:10.1016/j.atmosenv.2007.02.034, 2007.
16. Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., Akimoto, H.: Emissions of air pollutants and greenhouse gases over Asian regions during 2000-2008: Regional Emission inventory in ASia (REAS) version 2, Atmos. Chem. Phys., 13, 11019-11058, 2013.
17. Lee, S.-H., Akimoto, H., Nakane, H., Kurnosenko, S.: Lower tropospheric ozone trend observed in 1989-1997 in Okinawa, Japan, Geophys. Res. Lett., 25, 1637-1640, 1998.
18. Lin, C., Jacob, D. J., Fiore, A.M.: Trends in exceedances of the ozone air quality standard in the continental United States, 1980-1998, Atmospheric Environment, 35, 3217-3228, 2001.
19. Lin, M., Fiore, A. M., Horowitz, L. W., Cooper, O. R., Naik, V., Holloway, J., Johnson, B. J., Middlebrook, A. M., Oltmans, S. J., Pollack, I, B., Ryerson, T. B., Warner, J. X, Wiedinmyer, C., Wilson, J., Wyman, B.: Transport of Asian ozone pollution into surface air over the western United States in spring. Journal of Geophysical Research, 2012, 117(D21):183-204.
20. Lin, M., Fiore, A.M., Horowitz, L.W., Langford, A.O., Oltmans, S. J., Tarasick, D., Reider H.E.: Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions, Nature Communications, 6, 7105, doi:10.1038/ncomms8105, 2015a.
21. Lin, M., Horowitz, L. W., Cooper, O.R., Tarasick, D., Conley, S., Iraci, L.T., Johnson, B., Leblanc, T., Petropavlovskikh, I., Yates, E. L.: Revisiting the evidence of increasing springtime ozone mixing ratios in the free troposphere over western North America, Geophysical Research Letter, 42, doi:10.1002/2015GL065311, 2015b.
22. Lin, M., Horowitz, L.W., Oltmans, S. J., Fiore, A. M., Fan S.: Tropospheric ozone trends at Manna Loa Observatory tied to decadal climate variability, Nature Geoscience, 7, 136-143, doi:10.1038/NGEO2066, 2014.

23. Lin, W., Xu, X., Zhang, X., Tang, J.: Contributions of pollutants from North China Plain to surface ozone at the Shangdianzi GAW Station, Atmos. Chem. Phys., 8, 5889-5898, doi:10.5194/acp-8-5889-2008, 2008.
24. Mijling, B., van der A, R. J., and Zhang, Q.: Regional nitrogen oxides emission trends in East Asia observed from space, Atmos. Chem. Phys., 13, 12003-12012, doi:10.5194/acp-13-12003-2013, 2013.
25. Monks P. S., Archibald A. T., Colette A., Cooper O., Coyle M., Derwent R., Fowler D., Granier C., Law K. S., Mills G. E., Stevenson D. S., Tarasova O., Thouret V., von Schneidemesser E., Sommariva R., Wild O., Williams M. L.: Tropospheric ozone and its precursors from the urban to the global scale from air quality to short-lived climate forcer. Atmos. Chem. Phys., 15, 8889–8973, 2015.
26. NRC (National Research Council), 1991. Rethinking the Ozone Problem in Urban and Regional Air Pollution. National Academy Press, Washington, DC.
27. Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period of 1980-2020, Atmos. Chem. Phys., 7, 3319-4444, 2007.
28. Oltmans, S. J., Lefohn, A. S., Shadwick, D., Harris, J. M., Scheel, H. E., Galbally, I., Tarasick, D.W., Johnson, B.J., Brunke, E.-G., Claude, H., Zeng, G., Nichol, S., Schmidlin, F., Davies, J., Cuevas, E., Redondas, A., Naoen, H., Nakano, T., Kawasato, T.: Recent tropospheric ozone changes-a pattern dominated by slow or no growth. Atmospheric Environment, 67(2), 331-351, 2013.
29. Oltmans, S., Lefohn, A.S., Harris, J.M., Galbally, I., Scheel, H.E., Bodeker, G., Brunke, E., Claude, H., Tarasick, D., Johnson, B.J., Simmonds, P., Shadwick, D., Anlauf, K., Hayden, K., Schmidlin, F., Fujimoto, T., Akagi, K., Meyer, C., Nichol, S., Davies, J., Redondas, A., Cuevaso, E.: Long-term changes in tropospheric ozone, Atmos. Environ., 40, 3156-3173, 2006.
30. Ordonez, C., Brunner, D., Staehelin, J., Hadjinicolaou, P., Pyle, J.A., Jonas, M., Wernli, H., Prevot, A.S.H.: Strong influence of lowermost stratospheric ozone on lower tropospheric background ozone changes over Europe, Geophys. Res. Lett., 34, L07805. http://dx.doi.org/10.1029/2006GL029113, 2007.
31. Parrish, D. D., Law, K. S., Staehelin, J., Derwent, R., Cooper, O. R., Tanimoto, H., Volz-Thomas, A., Gilge, S., Scheel, H.-E., Steinbacher, M., Chan, E.: Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes, Atmos. Chem. Phys 12: 11485–11504, 2012, doi:10.5194/acp-12-114852012.
32. Pausata, F. S. R., Pozzoli, L., Vignati, E., Dentener, F. J.: North Atlantic oscillation and tropospheric ozone variability in Europe: model analysis and measurements intercomparison, Atmos. Chem. Phys., 12, 6357-6376, 2012.
33. Penkett, S.A.: Indications and causes of ozone increase in the troposphere. In: Rowland, F.S., Isaksen, I.S.A. (Eds.), The Changing Atmosphere. J. Wiley&Sons, p. 91. 1988.
34. Pusede, S. E., Steiner, A. L., and Cohen R. C.: Temperature and Recent Trends in the Chemistry of Continental Surface Ozone, Chem. Rev., 115, 3898-3918, 2015.
35. Rao, S. T., and Zurbenko, I. G.: Detecting and Tracking Changes in Ozone Air

Quality, Air & Waste, 44, 1089-1092, doi:10.1080/10473289.1994.10467303, 1994.
36. Rao, S. T., Zalewsky, E., and Zurbenko, I. G.: Determining Temporal and Spatial Variations in Ozone Air Quality, Journal of the Air & Waste Management Association, 45, 57-61, doi:10.1080/10473289.1995.10467342, 1995.
37. Rao, S. T., Zurbenko, I. G., Neagu, R., Porter, P. S., Ku, J. Y., and Henry, R. F.: Space and Time Scales in Ambient Ozone Data, Bulletin of the American Meteorological Society, 78, 2153-2166, 1997.
38. Richter, A., Burrows, J. P., Nub, H., Granier, C., and Neimeier, U.: Increase in tropospheric nitrogen dioxide over China observed from space, Nature, 437, 129-132, doi:10.1038/nature04092, 2005.
39. Streets, D. G., Tsai, N. Y., Akimoto, H., and Oka, K.: Trends in emissions of acidifying species in Asia, 1985–1997, Water Air and Soil Pollution, 130, 187-192, doi: 10.1023/A:1013883628877, 2001.
40. Verstraeten, W. W., Neu, J. L., Williams, J. E., Bowman, K. W., Worden, J. R., Boersma, K. F.：Rapid increases in tropospheric ozone production and export from China，Nature Geoscience, 8, 690－695, 2015, doi:10.1038/ngeo2493.
41. Wang, T., Wei, X.L., Ding, A.J., Poon, C.N., Lam, K.S., Li, Y.S., Chan, L.Y., Anson, M.: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994-2007, Atmos. Chem. Phys., 9, 6217-6227, 2009.
42. Wang, M., Shao, M., Chen, W., Lu, S., Liu, Y., Yuan, B., Zhang, Q., Zhang, Q, Chang, C.-C., Wang, B., Zeng, L., Hu, M., Yang, Y., and Li, Y.: Trends of non-methane hydrocarbons (NMHC) emissions in Beijing during 2002–2013, Atmos. Chem. Phys., 15, 1489–1502, 2015.
43. Xu, X., Lin,W., Wang, T., Yan, P., Tang, J., Meng, Z., and Wang, Y.: Long-term trend of surface ozone at a regional background station in eastern China 1991–2006: enhanced variability, Atmos. Chem. Phys., 8, 2595–2607, 2008.
44. Xu, X.B., Liu, X.W., and Lin, W.L.: Impacts of air parcel transport on the concentrations of trace gases at regional background stations (in Chinese with English abstract), Journal of Applied Meteorological Science, 20(6), 656-664, 2009.
45. Xu, X. B., and Lin, W. L.: Trends of Tropospheric Ozone over China Based on Satellite Data (1979-2005), Advances in Climate Change Research, 2, 43-48, doi: 10.3724/SP.J.1248.2011.00043, 2011.
46. Xu, W.Y., Lin, W.L., Xu, X.B., Tang, J., Huang, J.Q., Wu, H., and Zhang, X.C.: Long-term trends of surface ozone and its influencing factors at the Mt. Waliguan GAW station, China – Part 1: Overall trends and characteristics, Atmos. Chem. Phys. Discuss., 15, 30987–31024, 2015.
47. Zhang, Q.H., Zhang, J.P., and Xue, H.W.: The challenge of improving visibility in Beijing, Atmos. Chem. Phys., 10, 7821-7827, 2010.
48. Zhang, X. Y., Zhang, P., Zhang, Y., Li, X. J., Qiu, H.: The trend, seasonal cycle, and sources of tropospheric NO2 over China during 1996-2006 based on satellite measurement, Science in China Series D, 50(12), 1877-1884, 2007.

49. Zurbenko, I., Chen, J., Rao, S.T., Ku, J. Y., Gui, R., and Eskridge, R. E.: Detecting discontinuities in time series of upper air data: Demonstration of an adaptive filter technique, Journal of Climate, 9, 3548-3560, 1996.

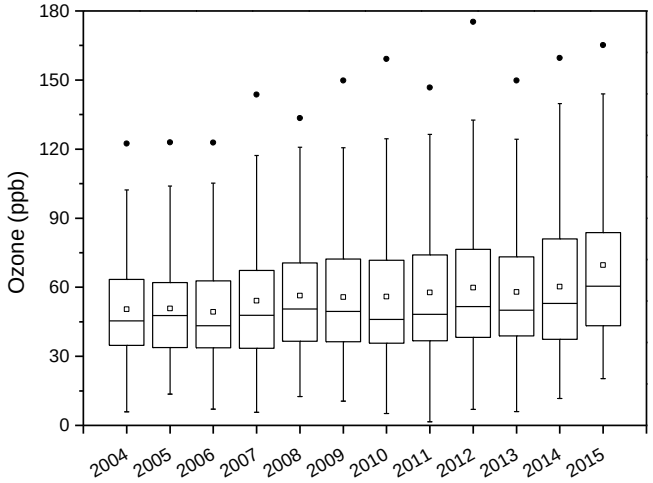

Fig 1. Percentile-box plot of yearly statistics of MDA8 values of ozone at SDZ, in the period of 2004-2015. Box depicts interquartile range and median; the square depicts the mean; whiskers depict 10th and 90th percentile; dot depicts maximum values.

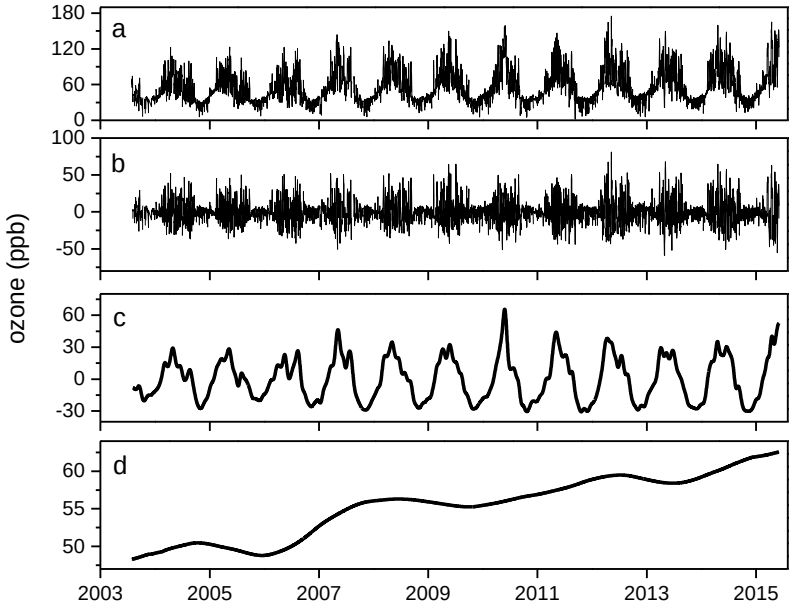

Fig 2. Separated time series of MDA8 values of ozone at SDZ: (a) the original data; (b) the short-term component, W(t); (c) the seasonal component, S(t); (d) the long-term component, e(t).

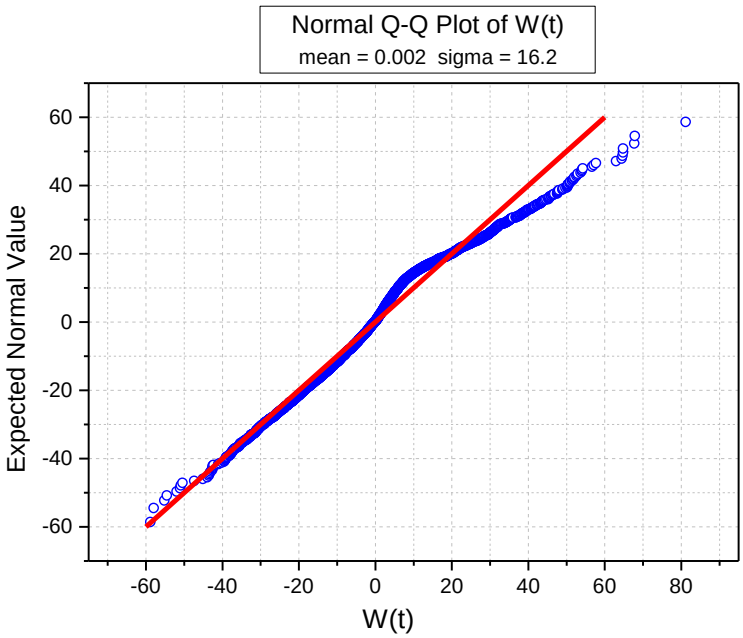

Fig 3. QQ plot of the short-term component W(t) for ozone.

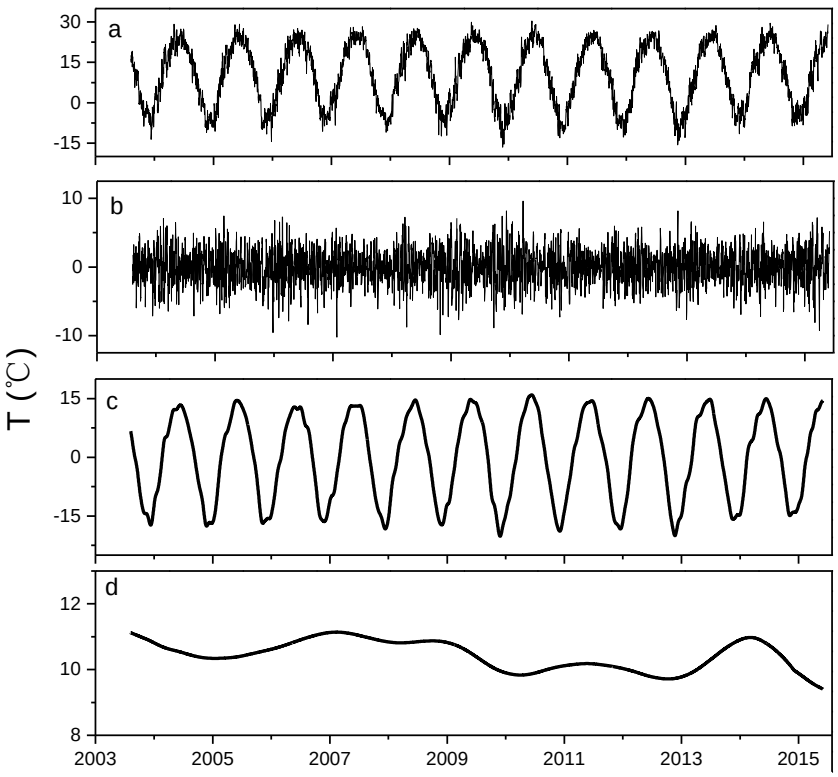

Fig 4. Separated time series of daily mean values of temperature at SDZ: (a) the original data; (b) the short-term component, W(t); (c) the seasonal component, S(t); (d) the long-term component, e(t).

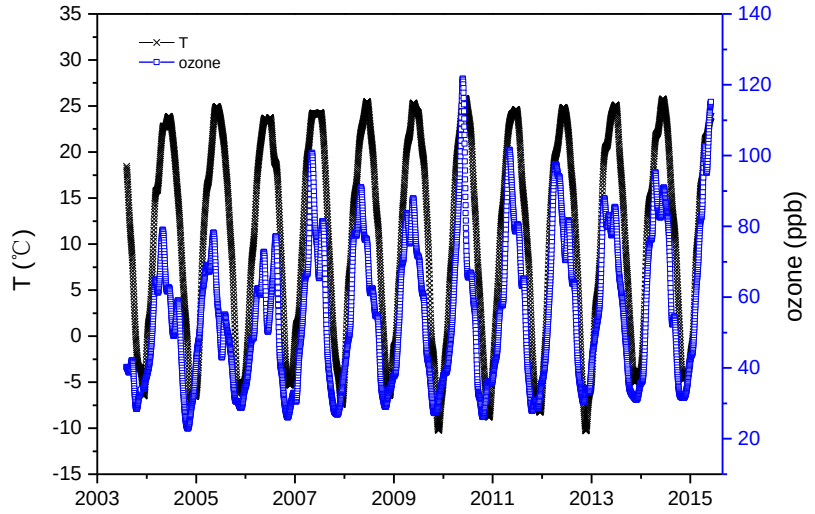

Fig 5. Results of the daily mean temperature and the MDA8 value of ozone after the application of $KZ_{15,5}$ filter to the original time series. The results indicate the sum of the seasonal and long-term components.

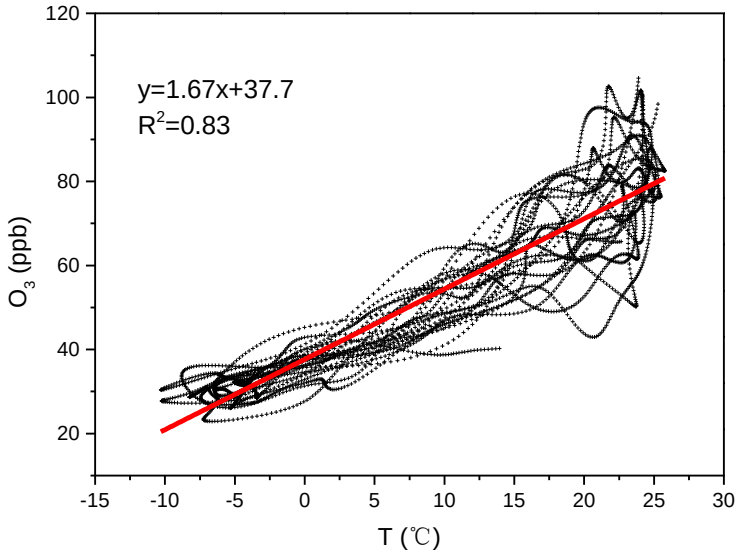

Fig 6. Linear regression fit on the filtered daily maxima of temperature and ozone. Temperature data are lagged by 17 days to ozone data.

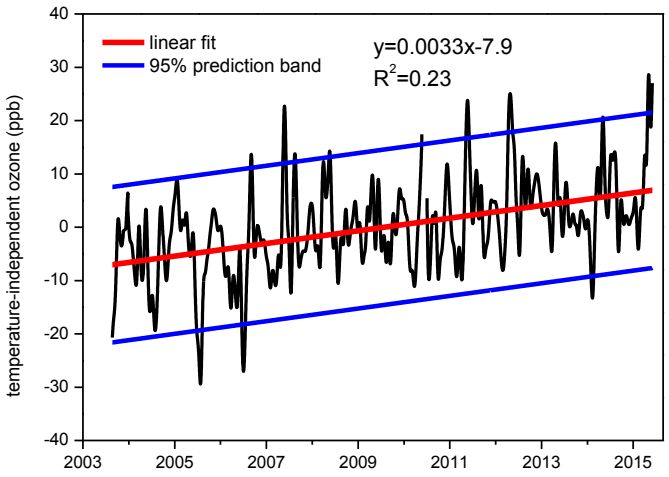

Fig 7. Time series of the noise-free and temperature-independent ozone. The red line is a linear fit and the blue lines are the 95% confidence prediction band.

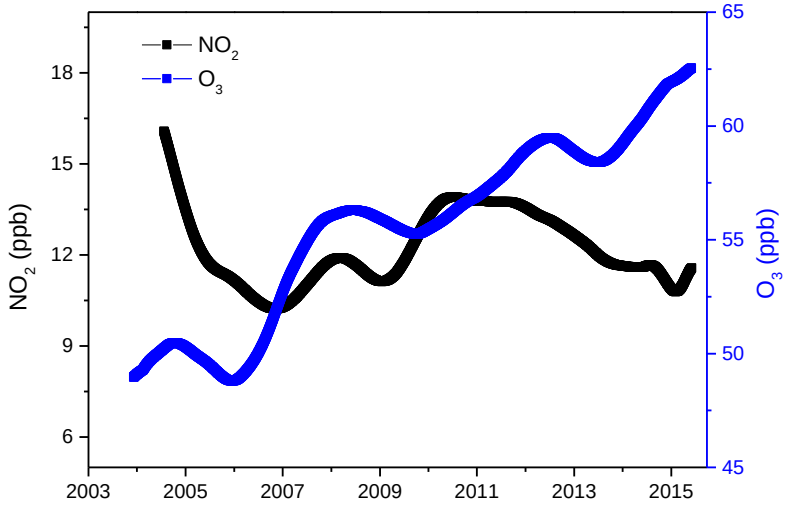

Fig 8. Long-term trends of NO_2 and the MDA8 ozone value calculated by $KZ_{365,3}$.