

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

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13 **Abstract**

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

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

10

11 1. Introduction

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

1 Both observations (Oltmans et al., 2006) and model simulations
2 (Hauglustaine and Brasseur, 2003) indicate that ground-level O₃
3 increased distinctly at northern mid-latitudes during the latter half of the
4 20th century, which is qualitatively in agreement with the increasing
5 anthropogenic emissions of precursors. Anthropogenic emissions of O₃
6 precursor have been declining in Europe and North America while
7 increasing in East Asia (Streets et al., 2001, Granier et al., 2011). The
8 largest increase in NO_x emissions is found in China and appears to have
9 continued into the 21st century based on some emission inventories
10 (Streets et al., 2001; Richter et al., 2005; Ohara et al., 2007; Mijling et al.,
11 2013; Kurokawa et al., 2013). From 1990 to 2010, inconsistency
12 occurred in the trends of surface O₃ in different regions. In the eastern US
13 surface O₃ decreased strongly in summer, was largely unchanged in
14 spring, and increased in winter, while O₃ increases in the western US
15 were the strongest in spring. Surface O₃ in East Asia was generally
16 increasing (Cooper et al., 2014). It is found that ground-level O₃ in the
17 Northeast Asian area, such as Japan (Lee et al., 1998) and Hong Kong
18 (Chan et al., 2004; Wang et al., 2009) increased significantly from 1990s
19 to 2000s. Enhanced variability of surface O₃, particularly the high level
20 O₃, was reported for the Yangtze River Delta region in eastern China (Xu
21 et al., 2008).

22 Dynamical factors may contribute to the long-term variations of

1 surface O₃. For example, the long-term increase of surface O₃ was found
2 to be related with the variability in stratosphere-to-troposphere transport
3 of O₃ (Ordonez et al., 2007; Hess and Zbinden, 2013; Lin et al., 2015a)
4 and changes in transport patterns (Pausata et al., 2012). Decadal
5 circulation shifts have played a key role in the autumnal ozone increase
6 and the absence of spring ozone change measured at Mauna Loa
7 Observatory (3.4 km altitude) over the subtropical Pacific Ocean during
8 1974-2012 (Lin et al., 2014). However, some studies (Brown-Steiner and
9 Hess 2011; Parrish et al., 2012; Lin et al., 2012; Oltmans, et al., 2013;
10 Derwent et al., 2015; Verstraeten et al., 2015) attribute O₃ increase in
11 some areas mainly to the transport of O₃ and related pollutants from the
12 continental China, where the emissions of O₃ precursors (NO_x and VOC)
13 had steadily increased (Ohara et al., 2007; Kurokawa et al., 2013). The
14 model results indicate that mean springtime ozone levels over western
15 North America in the most recent decade has increased by 5.9±2.1 ppbv
16 compared to the 1980s, which could be attributed in part to rising Asian
17 ozone precursor emissions and global methane (Lin et al., 2015b).
18 Because of increasing emissions of O₃ precursors due to the sustained
19 economic growth in China (Zhang et al., 2007), fueled by favorable
20 photochemical conditions, China itself may have been suffering severe
21 photochemical pollution. Particularly over polluted regions of China, the
22 long-term change of O₃ is expected. Indeed, limited previous studies

1 indicated that tropospheric O₃ had been changing significantly over some
2 regions of China. Ding et al. (2008) analyzed O₃ data from the MOZAIC
3 (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft)
4 program and obtained an increase rate of 2% per year of the daytime O₃
5 in the lower troposphere over Beijing and its surrounding areas for the
6 period of 1995-2005. Xu et al. (2011) analyzed the TOR (Tropospheric
7 Ozone Residue) data during 1979-2005 and found significant increasing
8 trends of tropospheric O₃ over the North China Plain for all seasons
9 except for winter, with a maximum rate of 1.10 DU per decade for
10 summer. Wang et al. (2009) found that surface O₃ at a regional station in
11 Hong Kong increased at an average rate of 0.58 ppb/yr from 1994 to
12 2007 and the trend was associated with the increase in tropospheric NO₂.

13 Information about the trends of surface O₃ in China's different regions,
14 particularly those with high emissions of pollutants, is urgently needed.
15 Due to the lack of long-term observations, it is hardly possible to gain
16 reliable results about the long-term trends of surface O₃ in various
17 regions of China. Recently, the long-term of surface O₃ in western China
18 is reported by Xu et al. (2015), based on the observations at the Mt.
19 Waliguan baseline station. So far, there has been no report of changes of
20 surface O₃ levels in highly polluted eastern China during the recent
21 decade. In this paper, we present the trend of surface O₃ in the north part
22 of eastern China, based on the measurements from a rural site.

1 Furthermore, the relative contributions of meteorological factors and the
2 change of anthropogenic emissions are investigated, which provide a
3 further insight into potential causes of the observed trend of surface O₃.

4 2. Data and methods

5 2.1 Site and measurements

6 Surface O₃ and ancillary data were collected at the Shangdianzi
7 (SDZ, 40.65 °N, 117.10 °E, 293.3m a.s.l.) station. SDZ is one of the
8 regional Global Atmosphere Watch (GAW) stations, located about 100
9 km northeasterly to the urban area of Beijing. Within 30 km of the site,
10 there are only small villages with sparse population and thus insignificant
11 anthropogenic emission sources. The observation facilities of the station
12 are situated on the south slope of a hill, which is surrounded by
13 mountainous areas except the southwest sector. Fruit trees and corn are
14 grown in the slope fields surrounding the site. It is shown that the
15 observations of pollutants at SDZ could reflect regional scale air quality
16 of North China (Lin et al., 2008; Xu et al., 2009).

17 The maximum daily average 8-h (MDA8) concentrations of O₃ were
18 calculated from hourly averages of O₃ from October 2003 to June 2015
19 and are used in the following analysis. To facilitate the analysis, ambient
20 NO₂ concentration and temperature measured at SDZ in the surface layer
21 during the same time period were processed to obtain daily averages.
22 Details about the observations and the quality assurance and quality

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

2 2.2 Analysis methods

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

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

$$16 Y_i = \frac{1}{m} \sum_{j=-k}^k X_{i+j}, \quad (1)$$

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

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

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

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

$$17 \quad O(t) = W(t) + S(t) + e(t), \quad (2)$$

18 where $O(t)$ is the original time series, $W(t)$ is meso-scale and
19 synoptic-scale variation, $S(t)$ is seasonal change, $e(t)$ is the long-term
20 (trend) component. According to the results of Rao et al. (1997), when
21 $KZ_{15,5}$ and $KZ_{365,3}$ filters are applied to the raw data, several influences
22 could be removed and the actual variation of ozone at different scales

1 would be obtained.

$$2 \quad W(t) = O(t) - KZ_{15,5}, \quad (3)$$

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

$$4 \quad e(t) = KZ_{365,3} \quad (5)$$

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

6 3. Results and discussion

7 3.1 General characteristics of yearly ozone distribution

8 The yearly statistics of MDA8 are presented in Fig. 1. Since the ozone
9 observation at SDZ started in October 2003, the MDA8 statistics for
10 2003 are not showed in Fig. 1. It is noted that data from 2015 cover only
11 the first 6 months. Although only the first 6 months records in 2015 are
12 used for the statistics, the maximum of the MDA8s in this year exceeds
13 160 ppb, only second to that in 2012. The yearly average of MDA8
14 varies from 49.3 ppb to 60.2 ppb, with a very significant positive trend
15 (1.05 ± 0.14 ppb/yr, $R=0.93$, $P<0.0001$) from 2004 to 2014. We also can
16 find a similar fluctuation of the median value within the range of 43.3
17 ppb to 53.0 ppb, with a positive trend (0.62 ± 0.20 ppb/yr, $R=0.72$,
18 $P<0.05$)from 2004 to 2014. The MDA8 level was relatively stable during
19 2004-2006, with the maximum around 120 ppb. However, the maximum
20 values exhibited a dramatic increase from 123 ppb in 2006 to 165 ppb in
21 2015, which seems to coincide with the increase of vehicles in eastern

1 China. For example, in Beijing, the vehicle fleet contained about 2.30
2 million in 2004, 2.88 million in 2006, 4.81 million in 2010, and 5.60
3 million in 2014 (data from: <http://www.bjjtgl.gov.cn/jgj/ywsj/index.html>).
4 The changes of the maximum value of O₃ and vehicle numbers both have
5 a dramatic increase trend during 2004-2015. Nevertheless, a clear
6 long-term trend in the median or maximum value cannot be derived from
7 the data shown in Fig. 1.

8 **3.2 Ozone time series separated by KZ filter**

9 Ozone time series (MDA8 value) from the SDZ site was separated
10 using the method described in section 2.2. Fig. 2 shows the original time
11 series of MDA8 values (Fig. 2a) and the time series of the separated short
12 term, seasonal and long-term components (Figs. 2b-2d). The original
13 MDA8 value exhibits a distinct seasonal variation, with overlapping of
14 high frequency noise (Fig. 2a). Removing the short-term component (Fig.
15 2b) leads clearer seasonal cycles shown in Fig. 2c. As can be seen in Fig.
16 2c, there are evident double peaks of ozone during the summer in each
17 year, which are not so obvious in the original time series (Fig. 2a).
18 Generally, the double peaks occur in June and September respectively,
19 and the dip in between occurs in July or August when relatively abundant
20 rainfalls damps ozone formation and accumulation. Under the influence
21 of the summer Asian monsoon, rainfalls in July and August at SDZ can
22 amount to more than 40% of the whole year's rainfall. Fig. 2c

1 demonstrates also some irregularities in the seasonal cycle, particularly
2 the year-to-year changes in the levels of annual maximum, minimum,
3 and the dip. The seasonal fluctuations have to be accurately removed to
4 get the long-term trend, as data for the trend analysis are required to be
5 independent of season and normally distributed. The short-term
6 component (Fig.2b) showed high frequency variations, ranging between
7 -60 ppb and 70 ppb, which are composed of noise (or fluctuation) caused
8 by meso-scale and synoptic-scale meteorological processes.
9 Synoptic-scale events have a timescale from 2 days to 3 weeks, which
10 could be removed by smoothing with the KZ filter for a window size of
11 15 days and 5 iterations. To further illustrate the short-term component, a
12 quantile-quantile (QQ) plot of $W(t)$ is presented in Fig. 3. The QQ plot
13 indicates that $W(t)$ basically obeys a normal distribution, with a mean
14 value of 0.002 ppb, suggesting that the $KZ_{15,5}$ filter can effectively
15 remove $W(t)$ from $O(t)$.

16 Through the previous steps and using the formulae (2)-(5), we
17 obtained the long-term trend of MDA8 at SDZ, as shown in Fig. 2d. This
18 long-term trend reveals a rapid increase of daily high value of surface
19 ozone at the SDZ site in the last decade. It is noteworthy that the increase
20 is not at a stable rate but with large inter-annual variations. Linear
21 regression (not shown) indicates that the average increase rate is $1.13 \pm$
22 0.01 ppb/yr ($r^2 = 0.92$). Previous study by Ding et al. (2008) using

1 MOZAIC data obtained a yearly increase of 2% (about 1 ppb/yr) of O₃ in
2 the boundary layer around Beijing during 1995-2005, which agrees well
3 with our result. Therefore, the greater Beijing area, probably the North
4 China Plain, has been suffering a rapid ozone increase for the last two
5 decades.

6 In view of the air pollution problems, the central government of
7 China issued a revised National Ambient Air Quality Standard
8 (CNAAQs, GB 3095-2012) in 2012, which has taken effect across the
9 country since 1 January 2016 and sets the MDA8 O₃ limits to 100 µg/m³
10 (51.0 ppb) and 160 µg/m³ (81.6 ppb) for national reserve areas and
11 residence/commercial areas, respectively. As can be seen in Fig. 2a, O₃
12 exceedance would be quite often in the warm seasons if the new
13 CNAAQs had been implemented.

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

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

2 **3.3 Cause analysis**

3 The long-term trend of ozone concentrations can be caused by the
4 changes of both pollutant emissions and related meteorological variables.
5 Climate variability and circulation shifts may lead to long-term changes
6 of O₃ as discussed in Lin et al. (2014, 2015a, 2015b). To assess the
7 influence of precursor emissions on the ozone trend, the meteorological
8 and chemical impacts have to be separated. However, both
9 meteorological and chemical impacts are complicated, not to mention the
10 interactions among meteorology, precursor emissions, and photochemical
11 reactions. Therefore, a clear separation of meteorological and chemical
12 impacts is hardly possible purely based on observational data.
13 Nevertheless, it is worthy to try to make attribution of the O₃ trend to
14 precursor emissions and other causes.

15 Although many meteorological variables can influence
16 photochemical formation of O₃, temperature is the prevailing one. The
17 increase of temperature can increase reaction rates, emissions of biogenic
18 VOCs, and reduce wind speeds, etc. (Lin et al., 2001; NRC, 1991;
19 Pusede et al., 2015). In a certain region, temperature is also closely
20 related with intensity of solar radiation, which plays a critical role in
21 photochemical formation of O₃. Thus, we took temperature as a key
22 meteorological parameter and investigated the relationship between O₃

1 and temperature, with the hope to obtain the influence of emission
2 changes on the long-term trend of O₃. The first step of our effort is to
3 divide the time series of temperature into three components in formula
4 (2), just as done for that of MDA8 (Fig.2). The results of the different
5 components of temperature are given in Fig. 4. Unlike the trend of
6 MDA8 of O₃, the long-term component for temperature in SDZ shows a
7 slight decrease trend ($R^2=0.015$) (Fig. 4d) and this long-term
8 component accounts only for 0.16% of the total variance of temperature.

9 The original data of O₃ and temperature are less correlated ($R^2=0.50$,
10 $P<0.0001$), presumably due to the strong influence of the short-term
11 component. Fig. 5 compares the derived seasonal cycles of the daily
12 mean temperature (from Fig. 4c) and the MDA8 of ozone (from Fig. 2c).
13 A coincidence is evident between both seasonal cycles. However, there is
14 also a distinct phase lag of the seasonal cycle between O₃ and
15 temperature, due to the influence of other processes on the O₃ level. Rao
16 et al. (1995) found that similar phase lag of about 3 weeks in the data
17 from the northeastern United States. In our case, the linear correlation
18 between O₃ and temperature becomes strongest ($R^2=0.83$, $P<0.0001$)
19 when the temperature data are lagged by 17 days (Fig. 6).

20 When only considering the influence of temperature, the seasonal-
21 and long-term components of O₃ could account for 93% of the total
22 variance at the Cliffside Park, New Jersey (Rao and Zurbenko, 1994).

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

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

$$15 \quad O_{kz}(t) = aT_{kz}(t+17) + b + \varepsilon(t) \quad (6)$$

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

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

1 the data in Fig. 7 are within the range of 95% confidence prediction band
2 except for some special cases happened in summertime. In summer,
3 temperature is not the dominant restricting factor for O₃ production
4 compared to other factors, such as rainfall and precursor concentrations.
5 Substantial negative influences occurring in 2005 and 2006 can be
6 explained by stronger impact of Asian summer monsoon on surface
7 ozone (Lin et al., 2008). The results in Fig. 7 indicate that the influence
8 of emission has been varying substantially but with an average increase
9 rate of 1.19 ± 0.03 ppb/yr. This increase rate is very close to the average
10 long-term trend of MDA8 of O₃ (1.13 ± 0.01 ppb/yr) in Fig. 2d, implying
11 that the increase of O₃ during 2003-2015 was mainly resulted from the
12 emission changes and the meteorological factors had only a tiny negative
13 influence. Jaffe and Ray (2007) also found that the temperature change
14 had little influence on long-term ozone trends in the western US.

15 Some studies suggested that the trends of surface O₃ at the similar
16 latitude as SDZ could be attributed partly to the reduced titration by NO
17 (Chou et al., 2006; Itano et al., 2007). In order to assess the effect of
18 changing NO titration on the long-term trend of O₃, we examined the
19 long-term measurements of NO₂ at SDZ during 2004-2015. A
20 comparison of the long-term trend of O₃ with that of NO₂, which was
21 also extracted using the previous methods, is displayed in Fig. 8. The
22 evolution of the NO₂ trend can be divided into three stages, i.e., a

1 substantial decrease of NO₂ occurring during the first 3 years, followed
2 by a small increase during 2007-2010, and finally a gradual decrease
3 during 2011-2015. The large decrease of NO₂ during 2004-2006
4 corresponded to the control of coal consumption around Beijing,
5 especially for the Olympic Games in 2008 (Zhang et al., 2010; Gao et al.,
6 2011) and to the relocation of the Capital Steel and Iron Company, one of
7 the large industrial source. The NO₂ increase from 10.2 ppb to 13.5 ppb
8 between 2007 and 2010 corresponded the rapid increase in numbers of
9 vehicles in Beijing from 3.1 million to 4.8 million
10 (<http://www.bjjtgl.gov.cn/jgj/ywsj/index.html>). From 2011 to 2015, the
11 new standard for vehicle emissions and measures for reduction of NO_x
12 emission from power plants were implemented, which may have helped
13 to reduce the NO₂ concentration. The long-term trends of O₃ and NO₂
14 given in Fig. 7 do not show any coincidence. Therefore, it is nearly
15 impossible that the reduced NO titration had led to the increase of surface
16 O₃ at SDZ. Previous studies (Ge et al., 2010; Ge et al., 2012) showed that
17 the ozone production efficiency at SDZ varied in from 0.2 to 21.1, with
18 an average of 4.9, implying that ozone formation at SDZ could be more
19 sensitive to VOCs than to NO_x. Accordingly, we believe that the changes
20 of VOCs emission and the ratio VOCs/NO_x might have caused the
21 increase of surface O₃ observed at SDZ. Unfortunately, no systematic
22 VOCs observations are available from the SDZ site so that we cannot

1 prove our view using measurements from SDZ. However, an large
2 increase in the anthropogenic emissions of non-methane hydrocarbon
3 (NMHC) can be inferred from the Multiresolution Emission Inventory
4 for China (MEIC) (<http://www.meicmodel.org>) for Beijing during
5 2004-2012, which supports our view, although the emission data are
6 questioned by a recent study (Wang et al., 2015).

7 4. Summary

8 We separated the time series of maximum daily average 8-h (MDA8)
9 concentration of surface O₃ observed at SDZ during 2003-2015 into
10 various spectral components using a modified KZ filter. This separation
11 has led to a better understanding of the variation of surface O₃ at the site
12 and its relationships with the meteorological and precursor variables,
13 enabling us to unravel the trend of O₃ from the original data containing
14 noises and seasonality, and estimate the contribution of changes of
15 precursor emissions to the trend. Our analysis reveals that the short-term,
16 seasonal, and long-term components of O₃ data from the SDZ site
17 accounted for 36.4%, 57.6%, and 2.2% of the total variance, respectively.

18 It is found that the MDA8 of O₃ at the site north of eastern China has
19 undergone a significant increase during 2003-2015, at a average rate of
20 1.13 ± 0.01 ppb/yr . Together with the reported yearly increase of 2% in
21 the lower tropospheric O₃ around Beijing for 1995-2005 (Ding et al.,
22 2008), we conclude that the north part of eastern China (i.e., the North

1 China Plain) may have been suffering a rapid increase in the O₃ level for
2 at least two decades. By eliminating the influence of air temperature, we
3 find that the observed increase of surface O₃ during 2003-2015 was
4 mainly induced by the emission changes and the meteorological factors
5 exerted only a tiny negative influence. Our result also indicates that
6 changes of VOCs emissions might play a more important role in the O₃
7 increase than the effect of NO_x.

8 Because fine particles pollution has been very severe in eastern China,
9 the government has been implementing several measures to control PM_{2.5},
10 including reductions of both NO_x and VOCs. This will have a potential
11 risk to additional O₃ increase in case a VOCs/NO_x ratio more favorable
12 for ozone production is reached. Thus, further studies are needed to trace
13 ozone trend and its influence in eastern China.

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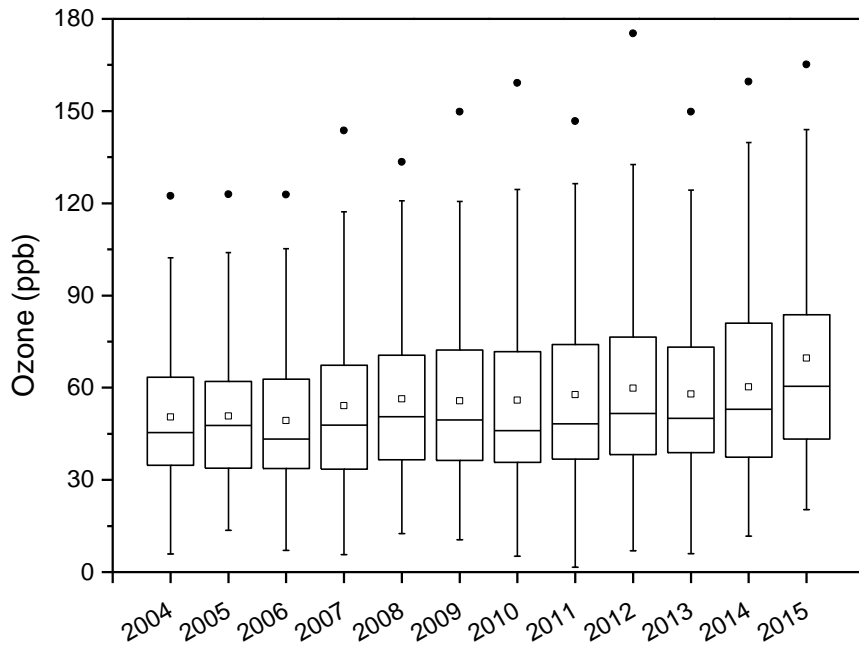
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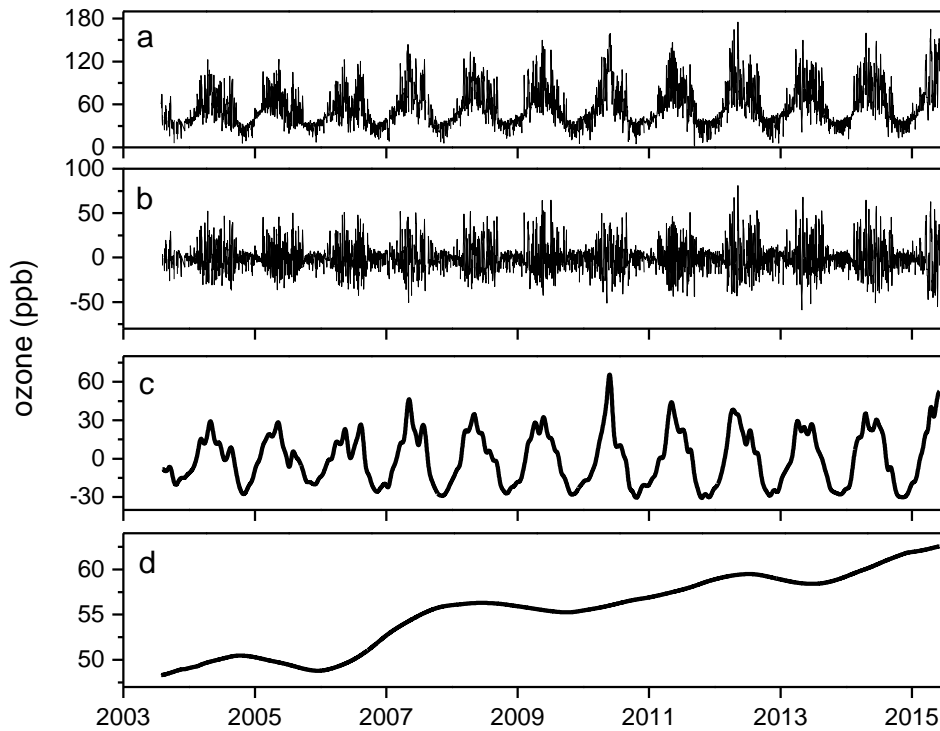
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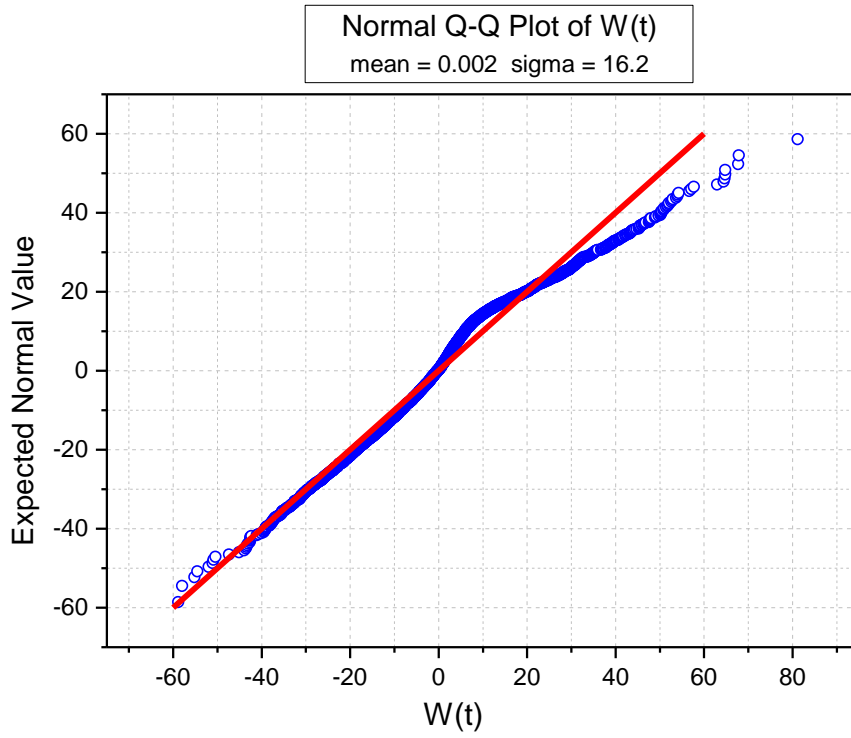
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1
 2 Fig 1. Percentile-box plot of yearly statistics of MDA8 values of ozone at SDZ,
 3 2004-2015. Box depicts interquartile range and median; the square depicts the mean;
 4 whiskers depict 10th and 90th percentile; dot depicts maximum values.

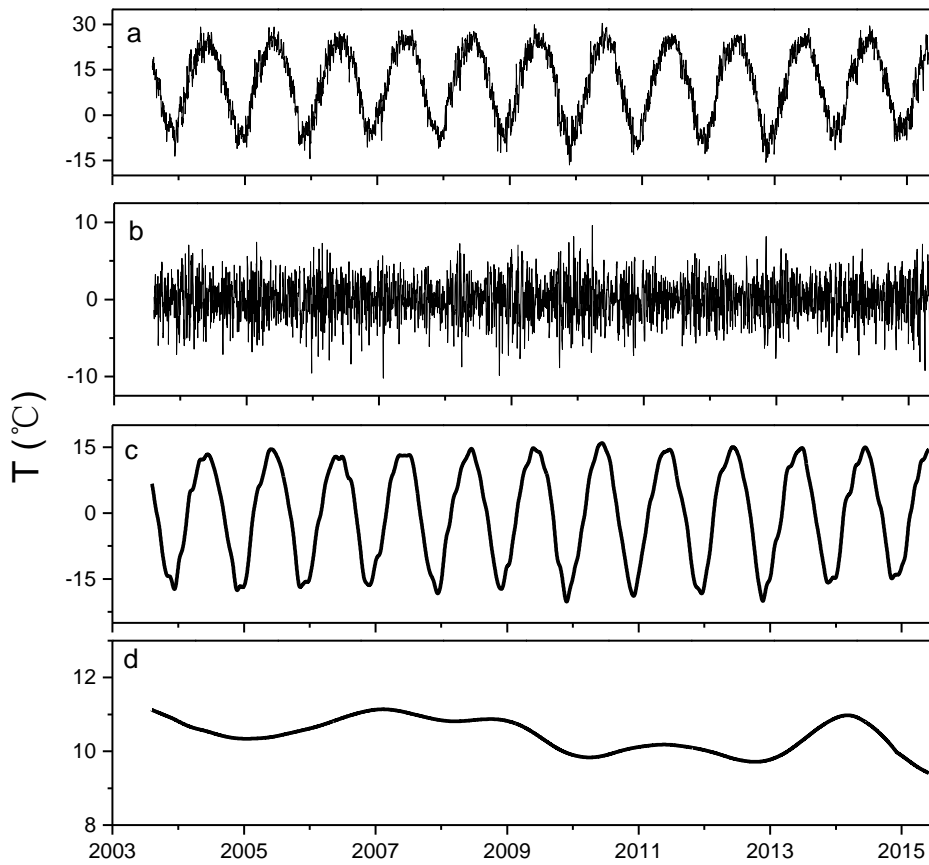


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 6 Fig 2. Separated time series of MDA8 values of ozone at SDZ: (a) the original data;
 7 (b) the short-term component, $W(t)$; (c) the seasonal component, $S(t)$; (d) the
 8 long-term component, $e(t)$.



1

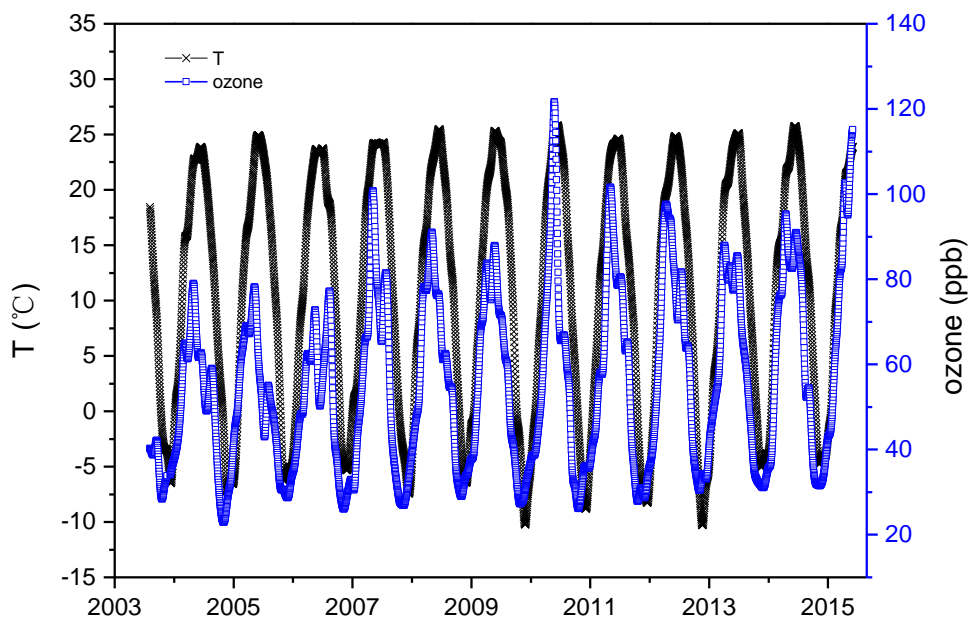
2 Fig 3. QQ plot of the short-term component $W(t)$ for ozone.



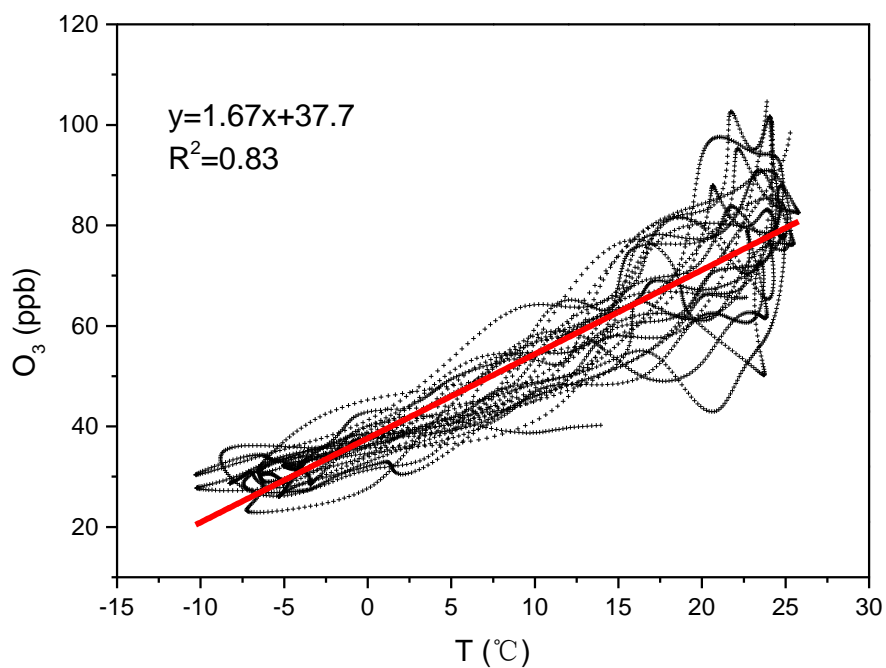
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4 Fig 4. Separated time series of daily mean values of temperature at SDZ: (a) the

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

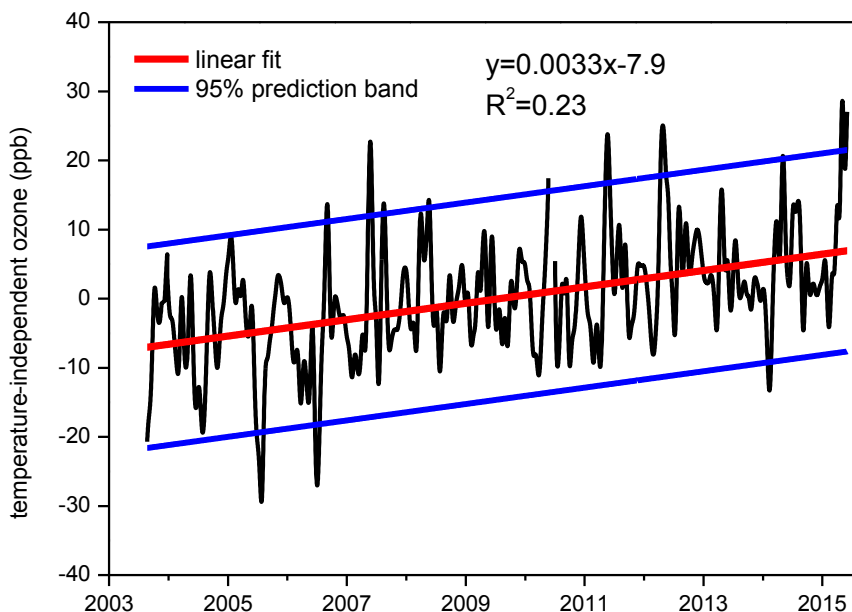


- 3
- 4 Fig 5. Results of the daily mean temperature and the MDA8 value of ozone after the
- 5 application of $KZ_{15,5}$ filter to the original time series. The results indicate the sum of
- 6 the seasonal and long-term components.



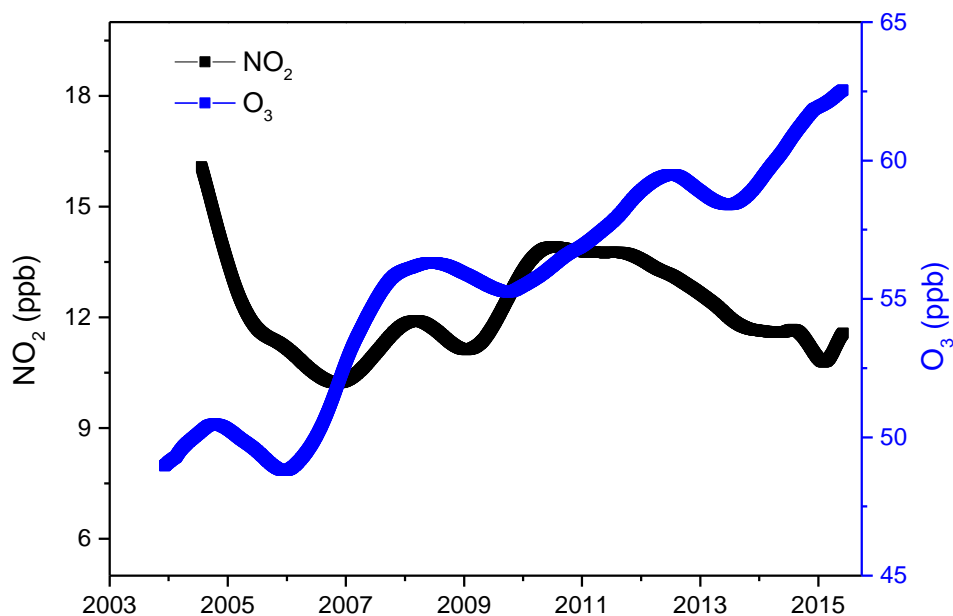
- 7
- 8 Fig 6. Linear regression fit on the filtered daily maxima of temperature and ozone.

1 Temperature data are lagged by 17 days to ozone data.



2

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 NO₂ and the MDA8 ozone value calculated by KZ_{365,3}.

7