Atmos. Chem. Phys. Discuss., 15, 31951–31972, 2015
www.atmos-chem-phys-discuss.net/15/31951/2015/
doi:10.5194/acpd-15-31951-2015

Atmospheric Chemistry and Physics
Discussions
Open Access

Significant increase of surface ozone at a regional background station in the eastern China

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

[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 Center, China Meteorological Administration, Beijing, China

Received: 6 September 2015 – Accepted: 27 October 2015 – Published: 13 November 2015

Correspondence to: W. L. Lin (linwl@cams.cma.gov.cn)

Published by Copernicus Publications on behalf of the European Geosciences Union.

Abstract

Ozone pollution has become one of the top environmental concerns in eastern China. Quantifying temporal trend of surface ozone concentrations is very meaningful to assess the impacts of the anthropogenic precursor reductions and the effects of emission control strategies. The level of surface ozone is impacted by both emissions of precursors and meteorological conditions. In order to examine the variation trend of ozone from 2003 to 2015 in Shangdianzi regional atmosphere background station, the modified KZ filter method was performed in this study to remove the influence of meteorological fluctuations on ozone concentrations. Results reveal that the short-term component, seasonal component and long-term component of ozone account for 36.4, 57.6 and 2.2 % of the total variance, respectively. The long-term trend shows that the surface daily maximum 8-h O_3 has undergone a significant increase during 2003–2015, with a rate of 1.1 ppb yr^{-1}. We find that the increase was completely resulted from the change of the emissions when the influence of the meteorological factors was eliminated. Furthermore, the variation of NO_2 indicated that VOCs seemed to play more important role in the increase trend of the surface ozone.

1 Introduction

Ground-level 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, 2007). Moreover, it is a precursor of OH radical, hence can exert indirect radiative forcing to the atmosphere by changing the lifetimes of some other reactive gases. Tropospheric O_3 primarily comes from in situ photochemical production, supplemented by stratospheric exchange. Ground-level O_3 produced by in situ production is directly affected by precursors emission, temperature, solar radiation and other meteorological factors.

Both observations (Oltmans et al., 2006) and model simulations (Hauglustaine and Brasseur, 2003) indicate that ground-level ozone 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 ozone precursor emission are declining in Europe and North America while increasing in East Asia (Granier et al., 2011). In East Asia, NO_x emissions grew by 4–6 % $year^{-1}$ since 1980 (Streets et al., 2001). The increase in NO_x emissions is largest in China and appears to have continued into the 21st century based on emission inventories (Streets et al., 2001), and the satellite data (Richter et al., 2005). From 1990 until 2010, surface ozone trends have varied by region. In the eastern US surface ozone has decreased strongly in summer, is largely unchanged in spring, and has increased in winter, while ozone increases in the western US are strongest in spring. Surface ozone in East Asia is generally increasing (Cooper et al., 2014). The increasement of ground-level ozone mixing ratios from 1990s to 2000s in the Northeast Asian area has been well known in Japan (Lee et al., 1998) and Hong Kong (Chan et al., 2004). Factors probably contributing to the increasing background ozone are variability in stratospheric flux (Ordonez et al., 2007; Hess and Zbinden, 2013) and changes in transport patterns (Pausata et al., 2012). Nevertheless, some researchers are likely to attribute ozone increase to the background ozone outflowing 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). The favorable photochemical conditions, together with the increasing emission of O_3 precursors due to the sustained economic growth in China (Zhang et al., 2007), may contribute to the long-term change of surface O_3 level. Ding et al. (2008) analyzed the long-term change of tropospheric O_3 over Beijing and its surrounding areas using tropospheric O_3 data collected by commercial aircrafts and obtained an increase rate of 2 % of the daytime O_3 concentration per year in the lower troposphere. Xu et al. (2011) analyzed the TOR data during 1979–2005 and significant increasing trends of tropospheric O_3 were found 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 increasing ozone levels at an average rate of 0.58 ppbv yr^{-1} at Hong Kong from 1994 to 2007 and the trend is associated with the increase in tropospheric NO_2. In addition, the changes of NO titration played a key role in the trend of surface ozone (Chou et al., 2006; Itano et al., 2007; Wang et al., 2009), which couldn't be neglected when investigating ozone trend, especially for the rapid developing countries.

In this paper, we present the trends of surface O_3 in the eastern China, based on monitored data at a regional atmospheric background station for the time period 2003–2015. Furthermore, the relative contributions of meteorological factors and the change of anthropogenic emissions are investigated, which provides further insight into the temporal trends of ozone concentration.

2 Data and methods

2.1 Site and measurements

Surface O_3 and other data from the station of Shangdianzi (SDZ) are investigated. SDZ (40.65° N, 117.1° E, 293.3 m a.s.l.) is one of the regional Global Atmosphere Watch (GAW) stations, located about 100 km northeasterly to the urban area of Beijing. The observations of pollutants at SDZ could reflect large scale air quality of eastern China. Hourly concentrations of O_3 and NO_2 were extracted for the period from October 2003 to June 2015. Detailed information about the observation and the QA/QC procedure were described by Lin et al. (2008). Also, surface temperature measured at SDZ was assembled for the same time period. A subset of data consisting of daily average of temperature and daily maximum 8-h ozone was derived from the hourly time series in the following analysis.

2.2 Analysis methods

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

$$Y_i = \frac{1}{m} \sum_{j=-k}^{k} X_{i+j} \tag{1}$$

Where $m = 2k + 1$, the Y_i become the input for the second pass, and so on.

Data filtered by KZ filter reflect physical phenomena unlike data treated by techniques that 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. The results indicate that the KZ filter method 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 much higher (about 10 times) confidence than the other methods. However, the long-term moving average of KZ filter 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 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 (1997) had 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

$$O(t) = W(t) + S(t) + e(t) \tag{2}$$

Where $O(t)$ is the original daily maximum 8-h ozone 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 (1997), when $\mathrm{KZ}_{15,5}$ and $\mathrm{KZ}_{365,3}$ filters are applied to the raw data, several influences could be removed and the actual variation of ozone at different scales would be obtained.

$$W(t) = O(t) - \mathrm{KZ}_{15,5} \tag{3}$$

$$S(t) = \mathrm{KZ}_{15,5} - \mathrm{KZ}_{356,3} \tag{4}$$

$$e(t) = \mathrm{KZ}_{356,3} \tag{5}$$

3 Result and discussion

3.1 General characteristics of yearly ozone distribution

The daily maximum 8-h O_3 distributions are presented in Fig. 1. Since ozone data started in October 2003, Fig. 1 didn't show the data of 2003. Although, there are only 6 months records in 2015, the maximum of 8-h ozone exceed 160 ppb, ranking only second to 2012. The yearly average of ozone varies from 33.1 to 40.5 ppb, excluding 2015. We can find a slight fluctuation of the median concentration from 42 to 48 ppb, but with no evident trend. Ozone distribution shows steady situation during 2004–2006, with the maximum concentration around 120 ppb. However, the maximum values exhibit dramatic increase, from 123 ppb in 2006 to 165 ppb in 2015, which seems to coincide with increase of vehicles in eastern China areas. Actually, whether median value or maximum value couldn't demonstrate long-term trend of ozone. The box diagram couldn't provide more information about ozone variation because of complicated effect factors.

3.2 Separation of ozone by KZ filter

Figure 2 shows the Daily Maximum 8-h Ozone at SDZ, as a raw time series, the separated short term, seasonal components and long-term component. We can find that the raw data exhibit a distinct seasonal variation, with peaks in summers. Additionally, the annual cycle is irregular and the year to year variation in seasonality is so large that it has to be accurately removed in order to perform statistical tests, in which the data are required to be statistically independent and identically distributed for detecting long-term trends. The short-term component (Fig. 2b) showed high frequency variations, ranged between 60 and 70 ppb, which composed of meso-scale and synoptic-scale motions acting as noise in time series. 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, the Quantile–quantile (Q–Q) plot of $W(t)$ is presented in Fig. 3 The Q–Q plot demonstrated $W(t)$ basically obey normal distribution, with a mean value of 0.002 ppb. Therefore, $\mathrm{KZ}_{15,5}$ filter could remove the $W(t)$ effectively.

Figure 2c illustrates the seasonal variation of ozone after removing white noise. There are evident double peaks of ozone during the summers of each year, while it is not obvious in the raw data plot. Generally, the double peaks occur in June and September approximately, and the relatively low value occurred in July and August is due to abundant rainfalls which can dampen ozone formation and accumulation. Through the previous steps, we get the long-term trend in Fig. 2d, which reveals a rapid increase of ground level ozone in the last decade. Linear regression (not shown here) indicates that the increasing rate is 1.1 ppb yr^{-1} ($r^2 = 0.92$). Previous study (Ding at al., 2008) demonstrates a yearly increase of 2 % in the lower tropospheric O_3 around Beijing during 1995–2005, which are in good agreement with our result. Therefore, the eastern China area has been suffering rapid ozone increase for almost two decades. The government has realized the severe situation and made revision for O_3 criterion in 2012, by adding the daily maximum 8-h average O_3 limit with a primary level of $100\,\mu g\,m^{-3}$ and secondary level of $160\,\mu g\,m^{-3}$.

We also examined the contribution of various scales of motion to the total variance, which is calculated from the unfiltered temperature data. The contributions of short-term component and seasonal component to the total variance are about 36.4 and 57.6 %, respectively. The long-term component accounts for 2.2 % of the total variance. The sum of covariance terms is less than 4 % of the total variance, indicating good separation of components. The long-term component is only a small part relative to the other two components, which confirms the necessity to clearly separate the short-term scales and seasonal variations from the data to obtain long-term trends correctly.

3.3 Trend analysis

The long-term trend of ozone concentrations could be attributable to the changes of both pollutant emissions and meteorological variables. If we want to assess the influence of precursor emissions on the ozone trend, the meteorological and chemical signals have to be separated primarily.

Although there are many meteorological variables which can influence ozone formation, temperature is the prevailing one, the increasing of which can increase reaction rates, natural VOC emissions, and reduce wind speeds, etc. (Lin et al., 2001; NRC, 1991). Thus, we can get the appropriate influence of emission change on the long-term trend of O_3 by only excluding the influence of temperature. At first, we divide temperature into three parts according to Eq. (2). Figure 4 shows the result of the different components of temperature, which is similar as ozone. Whereas, there is not any significant long-term trend for temperature in SDZ and the long-term component only accounts for 0.16 % of the total variance.

When using the raw data, the scatter plot of temperature versus ozone (not shown) indicates a poor relationship because of the strong influence of the white noise. Figure 5 shows the time series of the daily mean temperature and the maximum of 8-h ozone which have removed the white noise. There is a distinct phase lag of the annual cycle between ozone and temperature, due to the influence of other processes on ozone production. Rao (1995) found the phase lag was about 3 weeks observed in the northeastern United States. The linear relationship between ozone and temperature becomes stronger when the temperature data are lagged by 17 days in our result (Fig. 6).

Rao and Zurbenko (1994) have accounted for 93 % of the variance in the seasonal- and long-term component of ozone, considering only temperature for the Cliffside Park, New Jersey. While our result just accounted for 83 %. We tried to add more meteorological factors that could affect ozone production, such as solar radiation, relative humidity, but the correlation improved no more than 0.5 %. Thus, we can deduce that the influence of emissions had more complex influence at SDZ than at Cliffside Park, which is reasonable because of the rapid increase of anthropogenic emission around eastern China in the last decade.

The long-term trend of ozone concentrations, attributable to pollutant emissions, is determined by performing a linear least-squares regression between the temperature-independent ozone time series and time. Figure 7 demonstrates the time series of noise-free temperature-independent ozone, basically equal to the influence of emission changes on long-term trend of ozone. Most of the data are in the range of 95 % confidence prediction band except for some special cases happened in summertime. In summer, the temperature is not the dominant restricting factor for ozone production compared to other factors, such as rainfall and precursor concentrations. The substantial negative influence occurred at 2005 and 2006 can be explained by stronger impact of Asian summer monsoon on surface ozone (Lin et al., 2008). The result indicated that the influence of emission had been increasing continually, with an averaged influence of 1.2 ppb yr^{-1} (0.0033 · 365). Compared to the long-term trend of ozone (1.1 ppb yr^{-1}) in Fig. 2d, we can conclude that the increase of O_3 during 2003–2015 mainly resulted from the emission changes and the meteorological factors just played 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 ozone 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 NO titration on long-term trend of ozone, we examined the NO_2 concentration during 2004–2015 at SDZ. Comparison of long-term trend of ozone and NO_2, which was extracted as previous steps, was displayed in Fig. 8. The trend of NO_2 can be divided into three parts. The substantial decrease of NO_2 occurred during the first 3 years was mainly due to the control of coal consumption around Beijing, for the Olympic Games in 2008 (Zhang et al., 2010; Gao et al., 2011). NO_2 concentration increases from 10.2 ppb to 13.5 ppb between 2007 and 2010, as the vehicles increase from 3.1 million to 4.8 million in Beijing. From 2011 to 2015, NO_2 decreases gradually because the new emission standard for vehicle was implemented. Ozone shows an "N-type" variation while NO_2 decrease in the first part, which seems no relationship of these two variables. Ozone increases steadily no matter whether NO_2 increases or decreases from 2008 to 2015. Thus, the titration of NO has little effect on the ozone increase at SDZ. In addition, we can find the fluctuation of NO_2 only with a range about 4 ppb, which does not consist with amplitude of ozone. Accordingly, we are inclined to believe that the changes of VOCs emission would possibly lead to the increase of ozone at SDZ. Unfortunately, no long-term VOCs observations are carried out at SDZ.

4 Summary

We applied modified KZ filter to separate the various spectral components of surface ozone at SDZ during 2003–2015. Separation leads to a clearer understanding of ozone and its relationships with the meteorological and precursor variables, enabling us to track the ozone trend due to the changes of the precursor emissions. Our analysis reveals that the short-term component, seasonal component and long-term component of ozone at SDZ account for 36.4, 57.6, and 2.2 % of the total variance, respectively.

Long-term trend shows that the daily maximum 8-h O_3 in the eastern China has undergone a significant increase during 2003–2015, with a rate of 1.1 ppb yr^{-1}. Ding at al. (2008) showed a yearly increase of 2 % in the lower tropospheric O_3 around Beijing during 1995–2005. Thus, eastern China areas have been suffering rapid ozone increase for almost two decades. Through eliminating the influence of temperature, we find that the increase of surface ozone during 2003–2015 was mainly induced by the emission changes and the meteorological factors just played a tiny negative influence. Our result also indicates that VOCs seem to play more important role in the ozone increase than the effect of NO titration.

Fine particles pollution is severe in eastern China and government has implement many strategies to control $PM_{2.5}$, including reduce both NO_x and VOCs, which has the potential risk to additional ozone increase because of suitable ratio of VOCs and NO_x for ozone production. Thus, further studies are needed to track ozone 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), Beijing Natural Science Foundation (8132025, 8152018), CCSF201505.

References

Chan, C. Y., Chan, L. Y., and Harris, J. M.: Urban and background trend in 1984–1999 subtropical Hong Kong, South China, Ozone-Sci. Eng., 25, 513–522, doi:10.1080/01919510390481829, 2003.

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.

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.

Ding, A. J., Wang, T., Thouret, V., Cammas, J.-P., and 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.

Eskridge, R. E., Ku, J. Y., Rao, S. T., Porter, P. S., and Zurbenko, I. G.: Separating Different Scales of Motion in Time Series of Meteorological Variables, B. Am. Meteorol. Soc., 78, 1473–1483, doi:10.1175/1520-0477(1997)078<1473:SDSOMI>2.0.CO;2, 1997.

Gao, Y., Liu, X., Zhao, C., and Zhang, M.: Emission controls versus meteorological conditions in determining aerosol concentrations in Beijing during the 2008 Olympic Games, Atmos. Chem. Phys., 11, 12437–12451, doi:10.5194/acp-11-12437-2011, 2011.

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.

Hauglustaine, D. A. and Brasseur, G.: Evolution of tropospheric ozone under anthropogenic activities and associated radiative forcing of climate, J. Geophys. Res., 106, 32337–32360, 2003.

Hess, P. G. and Zbinden, R.: Stratospheric impact on tropospheric ozone variability and trends: 1990–2009, Atmos. Chem. Phys., 13, 649–674, doi:10.5194/acp-13-649-2013, 2013.

IPCC: Climate change 2007: the physical science basis, in: Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, edited by: Solomon, S., Qin, D., Manning, M., Chen, Z., Marquis, M., Averyt, K. B., Tignor, M., and Miller, H. L., Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, p. 996, 2007.

Itano, Y., Bandow, H., Takenaka, N., Saitoh, Y., Asayama, A., and Fukuyama, J.: Impact of NO_x 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.

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.

Kurokawa, J., Ohara, T., Morikawa, T., Hanayama, S., Janssens-Maenhout, G., Fukui, T., Kawashima, K., and 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, doi:10.5194/acp-13-11019-2013, 2013.

Lee, S.-H., Akimoto, H., Nakane, H., and Kurnosenko, S.: Lower tropospheric ozone trend observed in 1989–1997 in Okinawa, Japan, Geophys. Res. Lett., 25, 1637–1640, 1998.

Lin, C., Jacob, D. J., and Fiore, A. M.: Trends in exceedances of the ozone air quality standard in the continental United States, 1980–1998, Atmos. Environ., 35, 3217–3228, 2001.

Lin, W., Xu, X., Zhang, X., and 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.

NRC (National Research Council): Rethinking the Ozone Problem in Urban and Regional Air Pollution, National Academy Press, Washington, DC, 1991.

Ohara, T., Akimoto, H., Kurokawa, J., Horii, N., Yamaji, K., Yan, X., and Hayasaka, T.: An Asian emission inventory of anthropogenic emission sources for the period 1980–2020, Atmos. Chem. Phys., 7, 4419–4444, doi:10.5194/acp-7-4419-2007, 2007.

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., and Cuevaso, E.: Long-term changes in tropospheric ozone, Atmos. Environ., 40, 3156–3173, 2006.

Ordonez, C., Brunner, D., Staehelin, J., Hadjinicolaou, P., Pyle, J. A., Jonas, M., Wernli, H., and Prevot, A. S. H.: Strong influence of lowermost stratospheric ozone on lower tropospheric background ozone changes over Europe, Geophys. Res. Lett., 34, L07805, doi:10.1029/2006GL029113, 2007.

Pausata, F. S. R., Pozzoli, L., Vignati, E., and Dentener, F. J.: North Atlantic Oscillation and tropospheric ozone variability in Europe: model analysis and measurements intercomparison, Atmos. Chem. Phys., 12, 6357–6376, doi:10.5194/acp-12-6357-2012, 2012.

Penkett, S. A.: Indications and causes of ozone increase in the troposphere, in: TheChangingAtmosphere, edited by: Rowland, F. S. and Isaksen, I. S. A., J. Wiley & Sons, p. 91, 1988.

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.

Rao, S. T., Zalewsky, E., and Zurbenko, I. G.: Determining Temporal and Spatial Variations in Ozone Air Quality, J. Air Waste Manage., 45, 57–61, doi:10.1080/10473289.1995.10467342, 1995.

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, B. Am. Meteorol. Soc., 78, 2153–2166, doi:10.1175/1520-0477(1997)078<2153:SATSIA>2.0.CO;2, 1997.

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.

Streets, D. G., Tsai, N. Y., Akimoto, H., and Oka, K.: Trends in emissions of acidifying species in Asia, 1985–1997, Water Air Soil Poll., 130, 187–192, doi:10.1023/A:1013883628877, 2001.

Wang, T., Wei, X. L., Ding, A. J., Poon, C. N., Lam, K. S., Li, Y. S., Chan, L. Y., and Anson, M.: Increasing surface ozone concentrations in the background atmosphere of Southern China, 1994–2007, Atmos. Chem. Phys., 9, 6217–6227, doi:10.5194/acp-9-6217-2009, 2009.

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.

Zhang, Q. H., Zhang, J. P., and Xue, H. W.: The challenge of improving visibility in Beijing, Atmos. Chem. Phys., 10, 7821–7827, doi:10.5194/acp-10-7821-2010, 2010.

Zhang, X. Y., Zhang, P., Zhang, Y., Li, X. J., and Qiu, H.: The trend, seasonal cycle, and sources of tropospheric NO_2 over China during 1996–2006 based on satellite measurement, Sci. China Ser. D, 50, 1877–1884, 2007.

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, J. Climate, 9, 3548–3560, 1996.

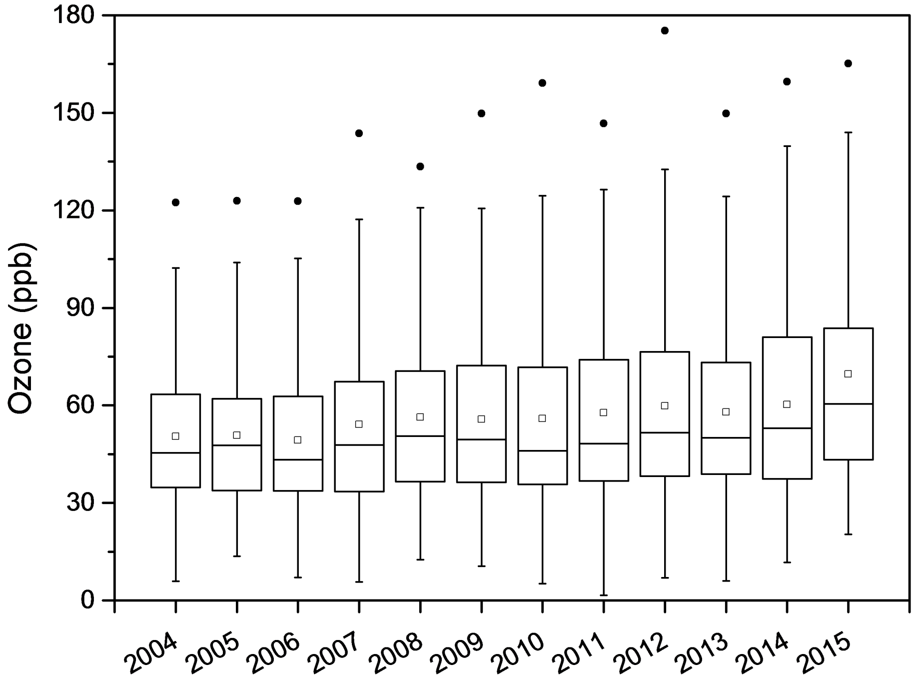

Figure 1. Daily Maximum 8-h Ozone Values at SDZ, 2004–2015. Box Depicts interquartile range and median; the square depicts the mean; whiskers depict 10th and 90th percentile; dot depicts maximum values.

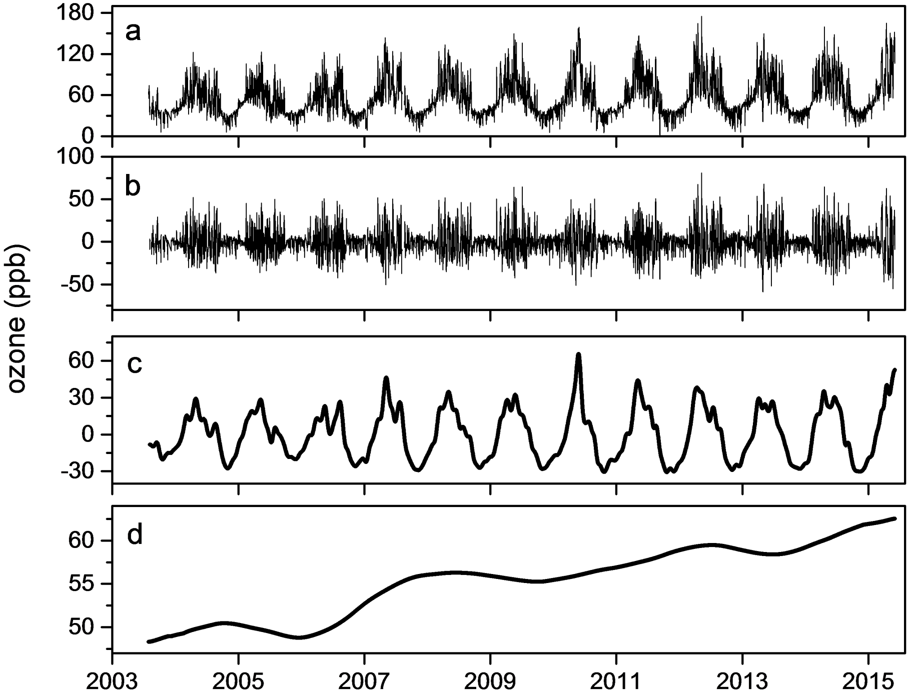

Figure 2. Daily Maximum 8-h Ozone from SDZ: **(a)** the raw data, **(b)** the short-term component, $W(t)$, **(c)** the seasonal component, $S(t)$, **(d)** the long-term component, $e(t)$.

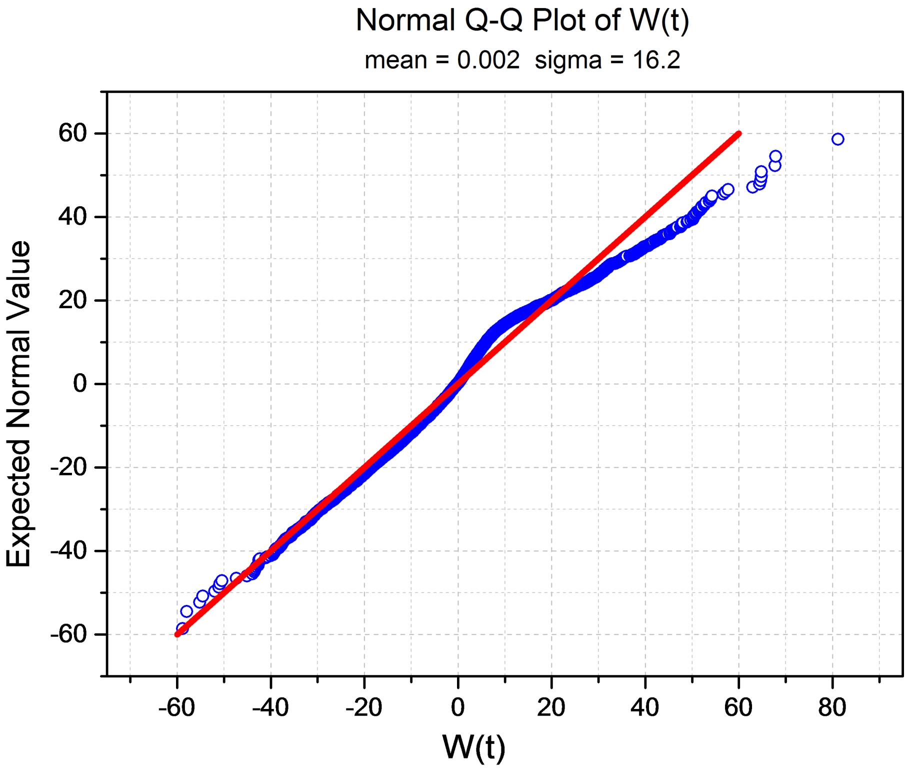

Figure 3. Q–Q plot of $W(t)$ for ozone.

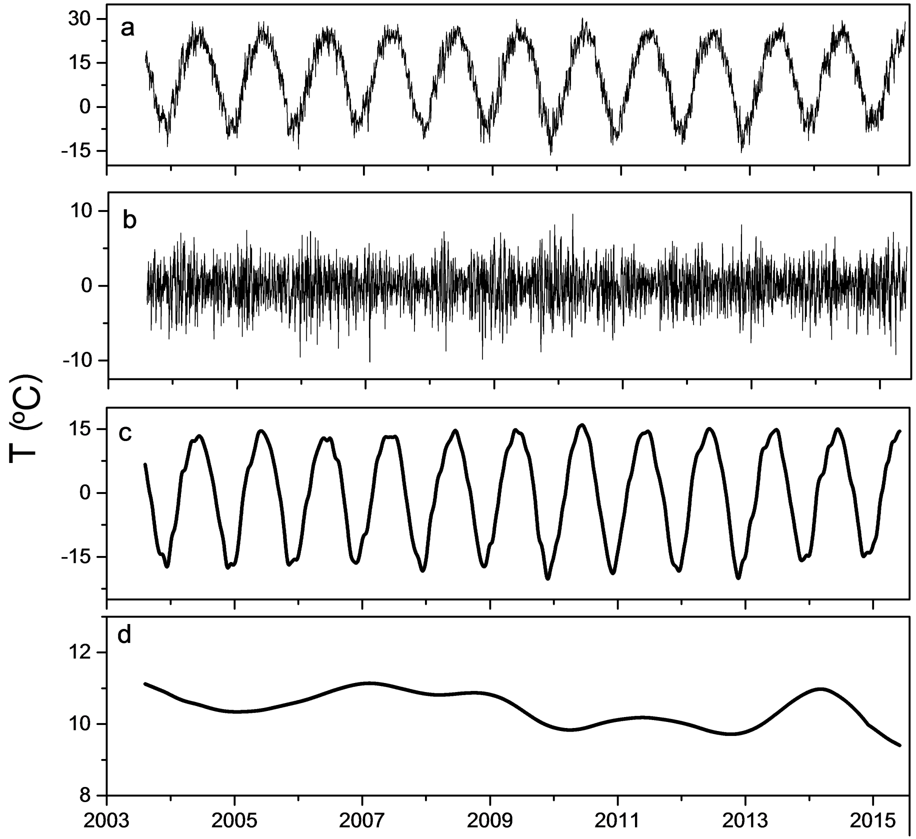

Figure 4. Same as Fig. 2 except for daily mean temperature.

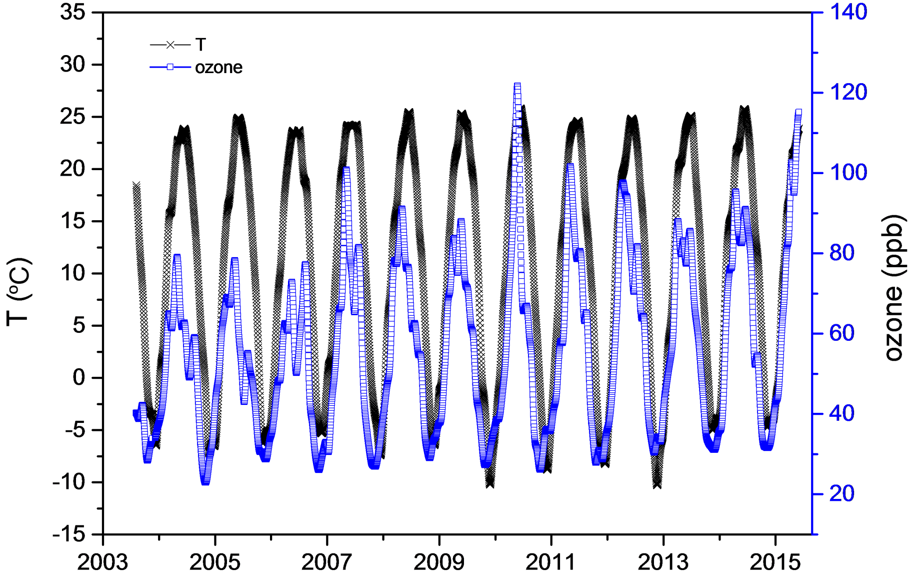

Figure 5. Daily mean temperature and maximum of 8-h ozone derived from the application of $KZ_{15,5}$ to the original time series, which indicates the sum of seasonal and long-term components.

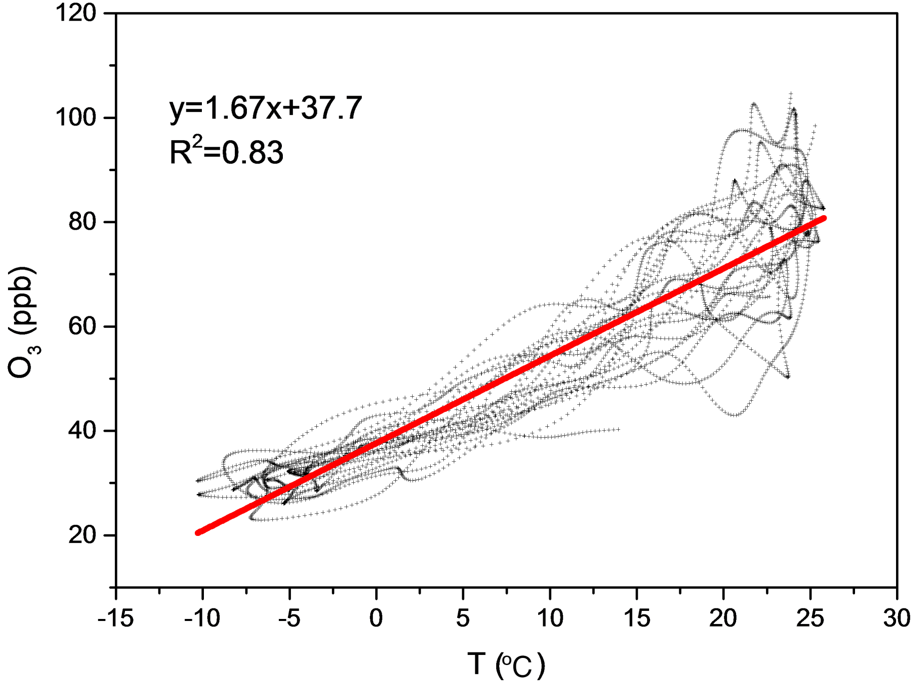

Figure 6. Linear least-squares regression fit for filtered temperature and ozone daily maxima. Temperature data are lagged by 17 days.

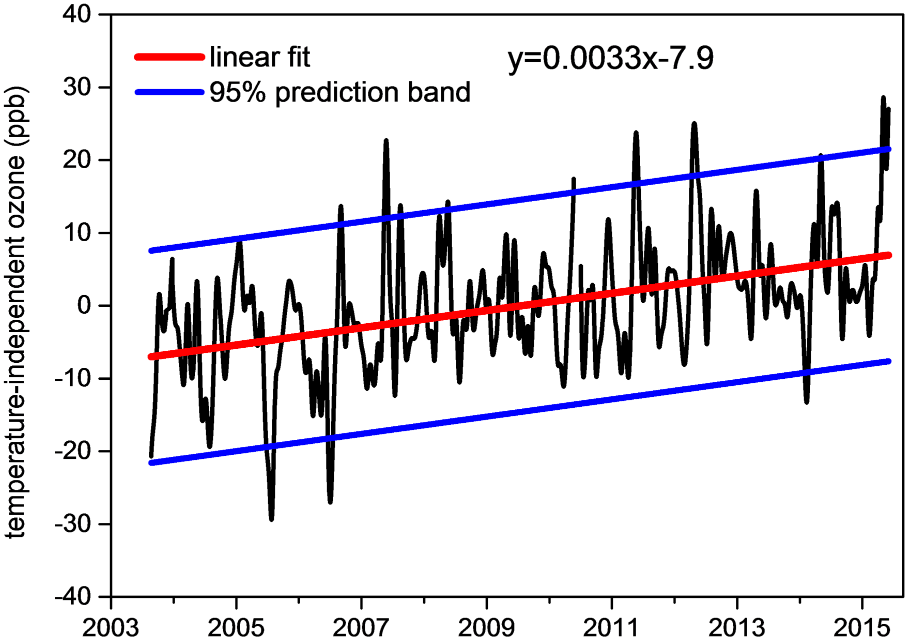

Figure 7. Noise-free temperature-independent ozone time series. The red line is the linear fit trend and blue lines are the 95 % confidence prediction band.

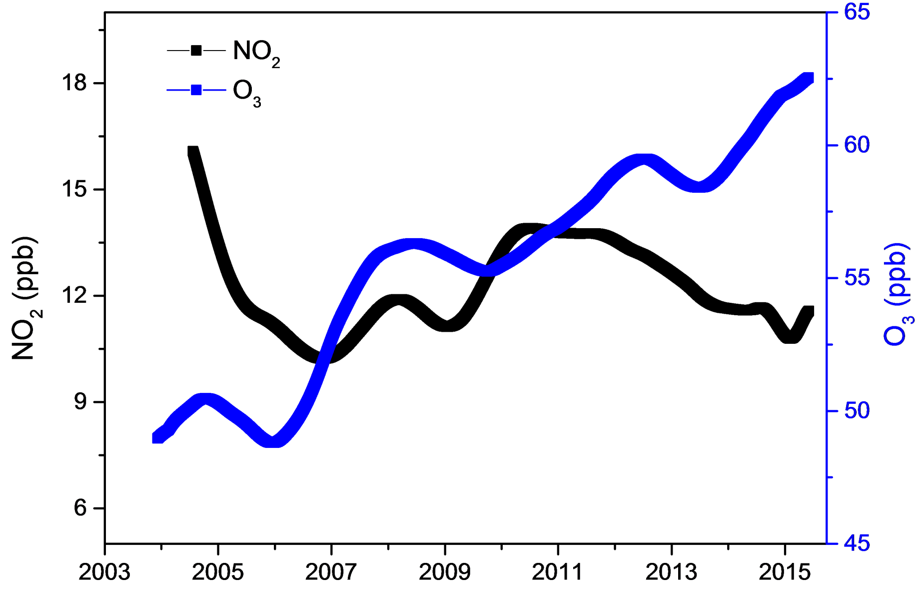

Figure 8. Long-term trend of NO$_2$ and 8-h maximum ozone.