We thank the referees and editor for their very constructive comments and suggestions. We also appreciate the comments by Dr. Meiyun Lin. We have revised our manuscript according to their comments and suggestions.

This comment is posted by Meiyun Lin (Princeton University, http://www.gfdl.noaa.gov/meiyun-lin-homepage).

This study presents a valuable 13-year record on surface ozone over Eastern China. Since the record is quite short, attempts to attribute observed ozone changes to regional emission trends must consider internal climate variability. In your literature review (Page 31953), I suggest the authors discuss the new findings from a few recent papers on the role of climate variability and circulation shifts on observed ozone changes:

Meiyun Lin, L.W. Horowitz, S. J. Oltmans, A. M. Fiore, Songmiao Fan (2014): Tropospheric ozone trends at Manna Loa Observatory tied to decadal climate variability, Nature Geoscience, 7, 136-143, doi:10.1038/NGEO2066.

Meiyun Lin, A.M. Fiore, L.W. Horowitz, A.O. Langford, S. J. Oltmans, D. Tarasick, H.E. Reider (2015): Climate variability modulates western US ozone air quality in spring via deep stratospheric intrusions, Nature Communications, 6, 7105, doi:10.1038/ncomms8105

Meiyun Lin, L.W. Horowitz, O.R. Cooper, D. Tarasick, S. Conley, L.T. Iraci, B. Johnson, T.Leblanc, I.vPetropavlovskikh, E.L. Yates(2015): 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.

Response: Thanks for your suggestion. We have added related contents in the revision.

Response to referee 1:

Interactive comment on "Significant increase of surface ozone at a regional background station in the eastern China" by Z. Q. Ma et al.

Anonymous Referee #1 Received and published: 9 December 2015

The authors have presented a concise study of the observed ozone trend at the Shangdianzi station in northern China and have used statistical methods to separate the trend due to emissions from the trend due to meteorology. If such an analysis were presented for a single station in the US or Europe where ozone is well understood the paper would not warrant publication in ACP. But because this study gives the research community a rare look at rapidly changing ozone levels in China, a region that is basically driving atmospheric chemistry in the northern hemisphere, the paper is appropriate for ACP. I recommend that the paper be published after a major revision as described below.

MAJOR COMMENTS

1) The standard of English in the manuscript needs to be greatly improved. The paper has too many grammatical and word-choice errors for me to correct and I recommend that the authors either find a colleague with excellent English skills to edit the grammar line-by-line, or employ the assistance of an ACP journal copy-editor.

Response：Accepted. The manuscript has been edited line-by-line by a colleague with excellent English skills. Changes can be seen in the whole text. We hope the revision can meet the requirement for the publication but are open for further language editing if necessary.

2) Please be clear regarding what you mean when you use the term "background ozone", which is a model construct. In contrast, "baseline ozone" is the observed ozone that flows into a region before it is impacted by local emissions. See the description and implications of these two quantities in:

Cooper, O. R., A. O. Langford, D. D. Parrish and D.W.Fahey (2015), Challenges of a lowered U.S. ozone standard, Science, 348, 1096-1097.

Response：

The background concentrations of certain atmospheric compositions generally refer to the atmospheric concentrations over a defined area under the ideal status of being "well mixed" (i.e., there is no significant direct effect of local sources). This can follow the definitions by Bronnimann et al. (Bronnimann, S., Schuepbach, E., Zanis, P., Buchmann, B., Wanner, H.: A climatology of regional background ozone at different elevations in Switzerland (1992–1998). Atmos. Environ. 34, 5191–5198, 2000.): regional background composition is the composition level over a large area (1000×1000 km^2) produced by the mixing of air masses of different origin outside and inside the defined area. The definition of regional background is made under the ideal status of "well mixing". The background levels and variations for certain species depend highly on its sources and sinks and on its atmospheric residence time. Although the background concentrations of important atmospheric compositions are greatly needed, so far there is no unified and widely accepted method of determining the background concentrations of different species. In practice, sites far from pollution sources are selected for long-term monitoring of atmospheric compositions according to a number of criteria and with careful and rigorous assessment. These sites are referred to as atmospheric background stations, like WMO GAW global or regional stations. Observational data recorded at these background sites are statistically

processed to extract the background concentrations by assuming that air masses were well mixed before arriving at the sites.
The Shangdianzi station was selected according to WMO/BAPMoN (one of the predecessors of GAW, see http://www.wmo.int/pages/prog/arep/gaw/history.html) criteria for regional background stations and named "Shangdianzi Regional Atmosphere Background Station" by the China Meteorological Administration.

To avoid misunderstanding, we have used the terms more carefully in the revision.

On page 31953, line16, you mention increasing background ozone. Do you mean to say baseline ozone? If so, where is this increase in baseline ozone observed (and please provide references)? Similarly, on line 18 you mention researchers attributing the increase in background (baseline?) ozone to China. Who is making these claims? Please provide references.
Response: The word of background ozone was used in the papers we cited. According to the definition of Cooper et al., we think it might also refer the baseline ozone.

We have added the following references attributing the increase ozone to China.
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.
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.
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.
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
Lin, M. et al. Transport of Asian ozone pollution into surface air over the western United States in spring. J. Geophys. Res. 117, D00V07 (2012).

3) According to the ACP data policy, the underlying chemical observations used in the analysis should be publicly available, as described here: Statement on the availability of underlying data: http://www.atmospheric-chemistryand-physics.net/about/data_policy.html#data_availability
"Authors are required to provide a statement on how their underlying research data can be accessed. This must be placed as the section "Data availability" at the end of the manuscript before the acknowledgements." This paper contains no data availability statement and the authors need to provide one. I am bringing this up because there is great interest within the atmospheric chemistry community regarding the quantity of ozone produced in East Asia as well as the ozone produced by East Asian emissions once the pollutants have been exported from the continent. With ozone pollution decreasing in North America and Europe, East Asia is the main driving

force behind any increase in tropospheric ozone. By having access to the ozone and ozone precursor observations described in this paper the scientific community can further its understanding of the global tropospheric ozone budget. It would be a great service to the community if the authors of this paper can make available the ozone data presented in the paper. Because Shangdianzi is a WMO GAW site the data should be available from the World Data Center for Greenhouse Gases. But when I visit their webpage all I can find for Shangdianzi are CO, CO2 and CH4: http://ds.data.jma.go.jp/gmd/wdcgg/cgibin/wdcgg/accessdata.cgi?cntry=China&index=SDZ240N00CMA_NOAA&para=CO2&select=inventory

Response： Submission of these data and those from other GAW station in China subjects to the regulations of CMA. As staff of the data owner, we are allowed to publish results obtained using the data but not allowed to submit the data without permission. We hope that the submission can be started as soon as possible.

4) Please provide more description of the Shangdianzi station and its surroundings. How far is it from urban areas? Is it elevated? Is it surrounded by forest or farmland? Are mountains nearby? A map showing the location of the station in relation to urban areas and mountains would be very helpful.

Response: Detail site information can be seen Lin et al. 2008: Contributions of pollutants from North China Plain to surface ozone at the Shangdianzi GAWStation, Atmos. Chem. Phys., 8, 5889–5898, 2008, www.atmos-chem-phys.net/8/5889/2008/. We add some information in the revision draft.

MINOR COMMENTS

Change the title to: “Significant increase of surface ozone at a regional background station in eastern China” There is also the problem regarding what you mean by background. Seeing as background refers to modelled ozone when a particular source is switched off, a better title would be: “Significant increase of surface ozone at a rural regional monitoring station in eastern China” .

Response: We have changed the title to “Significant increase of surface ozone at a rural site, north of eastern China”.

Page 31952 line 19 Radiative forcing due to ozone is not just observed at the surface, it impacts the entire troposphere. The Introduction needs references that are up to date:

1) when referencing IPCC, use the 2013 report, not the outdated 2007 report.

Response: We use the 2013 IPCC report in the revision.

IPCC, 2013: Climate Change 2013: The Physical Science Basis. Contribution of Working Group I to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change [Stocker, T.F., D. Qin, G.-K. Plattner, M. Tignor, S.K. Allen, J. Boschung, A. Nauels, Y. Xia, V. Bex and P.M. Midgley (eds.)]. Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 1535 pp.

2) when describing basic chemical and dynamic processes that impact ozone use references like Cooper et al., 2014, and Monks, P.S., A.T. Archibald, A.Colette, O.Cooper, M.Coyle, R.Derwent, D.Fowler, C. Granier, K.S. Law, G.E. Mills, D.S. Stevenson, O. Tarasova, V. Thouret, E. von Schneidemesser, R. Sommariva, O. Wild, and M.L. Williams (2015), 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, doi:10.5194/acp-15-8889-2015.

Response: The following paper is cited in the revision.

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, doi:10.5194/acp-15-8889-2015.

3) when describing ozone trends use reference like: Cooper et al., 2014 and Oltmans SJ, Lefohn AS, Shadwick D, Harris JM, Scheel HE, et al. 2013. Recent tropospheric ozone changes- A pattern dominated by slow or no growth. Atmos. Environ 67: 331–351. and Parrish DD, Law KS, Staehelin J, Derwent R, Cooper OR, et al. 2012. Long-term changes in lower tropospheric baseline ozone concentrations at northern mid-latitudes. Atmos. Chem. Phys 12: 11485–11504. doi:10.5194/acp-12-114852012

Response: We add these two references in the revision.

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.

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

4) Why use the outdated study of Streets et al 2001 when much more recent studies are available?

Response: We use more recent studies in the revision.

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.

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.

Page 31958 line 19, A better reference for the impact of temperature on ozone is: Pusede et al., (2015), Temperature and recent trends in the chemistry of continental surface ozone, Chem. Rev.,

115, 3898-3918.

Response: Accepted.

Pusede Sally E., Allison L. Steiner, and Ronald C. Cohen. Temperature and Recent Trends in the Chemistry of Continental Surface Ozone, Chem. Rev., 115, 3898-3918.

Page 31953 line 27 Please define TOR

Response: TOR, Tropospheric Ozone Residue. TOR data can be seen at http://science.larc.nasa.gov/TOR/data.html.

Page 31958 line 28 White noise is a very vague term and not one that specifically apples to atmospheric chemistry. Please find a different, more descriptive term.

Response: We replace the white noise with short-term component.

Figure 8 Be specific in the figure caption as to how these data were smoothed. Does the ozone correspond to Figure 2d?

Response: Indeed, O_3 and NO_2 showed in Figure 8 are the results of long-term trend calculated by $KZ_{365,3}$, just as Figure 2d.

Response to referee 2:

Atmos. Chem. Phys. Discuss., 15, C11936–C11938, 2016
www.atmos-chem-phys-discuss.net/15/C11936/2016/

Interactive comment on "Significant increase of surface ozone at a regional background station in the eastern China" by Z. Q. Ma et al.

J. B. Burkholder (Referee)
james.b.burkholder@noaa.gov

Received and published: 22 January 2016

In the absence of a review from one of the initial referees, I am submitting my comments on the submitted manuscript in order to complete the open discussion on this paper.

As pointed out by Referee #1, this paper presents results from a station in China where little information is currently available. The atmospheric science community would therefore benefit with the release of the ozone data and the interpretation. Therefore, it is imperative that the authors make the data publicly available, either prior or simultaneously with the publication of their paper. Major revision and re-review of the paper will be requiring prior to final publication. I encourage the authors to make this effort. Overall, I agree with the comments of Referee #1 and have some additional comments that the authors should address as listed below.

Thanks very much!

* The communication of the scientific results from this work is severely hampered by the language used. The manuscript needs to be critically edited (line-by-line) by a native English speaker.

Response: We have improved the English language in the revised manuscript.

* The reliability of the analysis results (short-term, seasonal, long-term components) is not clear. Although, some mention of variance is given in the text I do not find that very informative. For example, how sensitive are the results to the parameters of the filtering (smoothing) of the data. Is the data record sufficiently long to establish reliable results? Especially in the abstract, the uncertainty in the components and the rate of the daily maximum needs to be included.

Response: To get the short-term, seasonal, long-term components, the objective $KZ_{m,p}$ filter is used. Determination of the final low-pass filter (specifying "m" and the number of passes "p") is an iterative process in which the data user determines that the white noise has been removed. In this study, a window size of 15 days for m and 5 iterations for p are finally used and the result is optimized so that W(t) basically obeys a normal distribution, with a mean value of 0.002 ppb. A window size of 365 days for m and 3 iterations for p are used to obtain the low-frequency (long term) component. In the study, the sum of covariance terms of separated components is less than 4% of the total variance, indicating a good separation of components. Rao et al. (1997) made a good explanation on the choice of parameters.

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.

A 12-year long data is sufficient to establish reliable results because the choice of the time window size are 15 days for short term and 365 (1 year) for long term.

We include the uncertainty of the rate of the daily maximum O_3 trend in the revised manuscript. The long-term trend shows that the daily maximum 8-h surface ozone has undergone a significant increase during 2003-2015, with a rate of +1.13±0.01 ppb/yr (R^2=0.92). For different components, the variance contributions are used instead of uncertainty.

* Another general criticism is that several of the "conclusions" mentioned in the text are rather speculative and require a more quantitative analysis to support the author claims. Examples of this include:

Page 31956 ". . .which seems to coincide with increase of vehicles in eastern China areas."

Response: We cite related information: For example, in Beijing, the vehicle fleet is about 2.30 million in 2004, 2.88 million in 2006, 4.81 million in 2010, and 5.60 million in 2014 (data from: http://www.bjjtgl.gov.cn/jgj/ywsj/index.html, figure 1). The changes of the maximum value of O_3 and vehicle fleet both have a dramatic increase trend during 2004-2015.

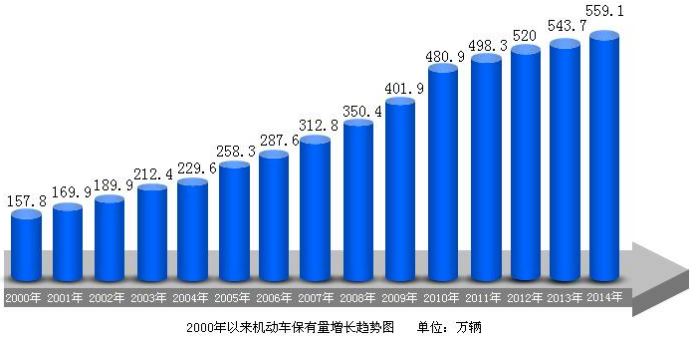

Figure 1. Trend of vehicle fleet in Beijing during 2000-2014.

(Source: http://www.bjjtgl.gov.cn/jgj/ywsj/index.html)

Page 31957 ". . . is due to abundant rainfalls. . ."

Response:

Generally, the double peaks occur in June and September respectively, and the dip in between occurs in July or August when relatively abundant rainfalls damps ozone formation and accumulation. Under the influence of the summer Asian monsoon, rainfalls in July and August at SDZ can amount to more than 40% of the whole year's rainfall.

Page 31959 "We tried to add more meteorological factors" and "In summer, the temperature is not. . ."

Page 31960 "We are inclined to believe. . ."

The text needs to be revised to reflect accordingly.

Response: We have revised the sentences as following.

"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%."

"In summer, the temperature is not the dominant restricting factor for O_3 production compared to other factors, such as rainfall and precursor concentrations."

"Accordingly, we believe that the changes of VOCs emission and the ratio VOCs/NOx might have caused the increase of surface O_3 observed at SDZ."

* Including a map of the site and possibly some indication of the prevailing meteorology would be very helpful to the reader.

Response: In the previous study (Lin et al., 2008) , the map and the prevailing meteorology have been descripted in detail. Here, we do not need to duplicate it.

* Define QA/QC on page 31954

Response: QA/QC is a short word for Quality Assurance and Quality Control. We define it in the revised paper.

* Page 31955: It is not clear how the filtering reflects "physical phenomena".

Response: We have changed "physical phenomena" to "physical processes" that is more accurate. Ozone concentrations are influenced by both emissions and meteorological variables, whereas temperature is dictated primarily by the prevailing meteorological conditions. The linear regression between the filtered O_3 concentrations ($O_{kz}(t)$) and the filtered Temperature lagged by 17 days ($T_{kz}(t+17)$) in this study was performed:

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

where a and b are fitted parameters, $\varepsilon(t)$ are the residuals of the relationship. Here, $\varepsilon(t)$ reveals changes in ozone attributable to changes in emissions.

* Page 31958, why not show the correlation (or lack of) between temperature and ozone, it seems like this is an important issue.

Response: We add the correlation (R^2=0.50, P<0.05) between temperature and ozone raw data, as well as the different components. For the long-term component, there is a slight negative correlation (R^2=0.015) between temperature and ozone. For the seasonal component, there are significant correlation (R^2=0.83, P<0.0001) between 17-day lagged temperature and ozone.

* Figure 3: what is the line on the graph?

Response: It is the line with a slope of 1.

* Figure 4: Provide a complete stand alone figure caption instead of referring to another figure.

Response: New caption is "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)."

* Figure 7: What does "noise-free" mean?

Response: "noise-free" mean the white noise (short-term variations) is filtered using Kolmogorov-Zurbenko (KZmp) filter. It's hard to understand, so we use the short-term variation instead of it.

* Figure 8: Specify where the NO2 data used in this analysis came from. This also needs to be clarified in the text.

Response: NO2 data are also measured at SDZ site. We add the information about it in section 2.1.

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]

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Abstract

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

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

1. Introduction

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

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

Dynamical factors may contribute to the long-term variations of

surface O_3. For example, the long-term increase of surface O_3 was found to be related with the variability in stratosphere-to-troposphere transport of O_3 (Ordonez et al., 2007; Hess and Zbinden, 2013; Lin et al., 2015a) and changes in transport patterns (Pausata et al., 2012). Decadal circulation shifts have played a key role in the autumnal ozone increase and the absence of spring ozone change measured at Mauna Loa Observatory (3.4 km altitude) over the subtropical Pacific Ocean during 1974-2012 (Lin et al., 2014). However, some studies (Brown-Steiner and Hess 2011; Parrish et al., 2012; Lin et al., 2012; Oltmans, et al., 2013; Derwent et al., 2015; Verstraeten et al., 2015) attribute O_3 increase in some areas mainly to the transport of O_3 and related pollutants from the continental China, where the emissions of O_3 precursors (NOx and VOC) had steadily increased (Ohara et al., 2007; Kurokawa et al., 2013). The model results indicate that mean springtime ozone levels over western North America in the most recent decade has increased by 5.9 ± 2.1 ppbv compared to the 1980s, which could be attributed in part to rising Asian ozone precursor emissions and global methane (Lin et al., 2015b). Because of increasing emissions of O_3 precursors due to the sustained economic growth in China (Zhang et al., 2007), fueled by favorable photochemical conditions, China itself may have been suffering severe photochemical pollution. Particularly over polluted regions of China, the long-term change of O_3 is expected. Indeed, limited previous studies

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

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

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

2. Data and methods

2.1 Site and measurements

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

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

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

2.2 Analysis methods

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

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

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

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

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

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

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

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

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

would be obtained.

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

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

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

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

3. Results and discussion

3.1 General characteristics of yearly ozone distribution

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

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

3.2 Ozone time series separated by KZ filter

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

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

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

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

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

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

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

3.3 Cause analysis

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

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

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

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

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

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

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

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

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

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

the data in Fig. 7 are within the range of 95% confidence prediction band except for some special cases happened in summertime. In summer, temperature is not the dominant restricting factor for O_3 production compared to other factors, such as rainfall and precursor concentrations. Substantial negative influences occurring in 2005 and 2006 can be explained by stronger impact of Asian summer monsoon on surface ozone (Lin et al., 2008). The results in Fig. 7 indicate that the influence of emission has been varying substantially but with an average increase rate of 1.19 ± 0.03 ppb/yr. This increase rate is very close to the average long-term trend of MDA8 of O_3 (1.13 ± 0.01 ppb/yr) in Fig. 2d, implying that the increase of O_3 during 2003-2015 was mainly resulted from the emission changes and the meteorological factors had only a tiny negative influence. Jaffe and Ray (2007) also found that the temperature change had little influence on long-term ozone trends in the western US.

Some studies suggested that the trends of surface O_3 at the similar latitude as SDZ could be attributed partly to the reduced titration by NO (Chou et al., 2006; Itano et al., 2007). In order to assess the effect of changing NO titration on the long-term trend of O_3, we examined the long-term measurements of NO_2 at SDZ during 2004-2015. A comparison of the long-term trend of O_3 with that of NO_2, which was also extracted using the previous methods, is displayed in Fig. 8. The evolution of the NO_2 trend can be divided into three stages, i.e., a

substantial decrease of NO_2 occurring during the first 3 years, followed by a small increase during 2007-2010, and finally a gradual decrease during 2011-2015. The large decrease of NO_2 during 2004-2006 corresponded to the control of coal consumption around Beijing, especially for the Olympic Games in 2008 (Zhang et al., 2010; Gao et al., 2011) and to the relocation of the Capital Steel and Iron Company, one of the large industrial source. The NO_2 increase from 10.2 ppb to 13.5 ppb between 2007 and 2010 corresponded the rapid increase in numbers of vehicles in Beijing from 3.1 million to 4.8 million (http://www.bjjtgl.gov.cn/jgj/ywsj/index.html). From 2011 to 2015, the new standard for vehicle emissions and measures for reduction of NOx emission from power plants were implemented, which may have helped to reduce the NO_2 concentration. The long-term trends of O_3 and NO_2 given in Fig. 7 do not show any coincidence. Therefore, it is nearly impossible that the reduced NO titration had led to the increase of surface O_3 at SDZ. Previous studies (Ge et al., 2010; Ge et al., 2012) showed that the ozone production efficiency at SDZ varied in from 0.2 to 21.1, with an average of 4.9, implying that ozone formation at SDZ could be more sensitive to VOCs than to NOx. Accordingly, we believe that the changes of VOCs emission and the ratio VOCs/NOx might have caused the increase of surface O_3 observed at SDZ. Unfortunately, no systematic VOCs observations are available from the SDZ site so that we cannot

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

4. Summary

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

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

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

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

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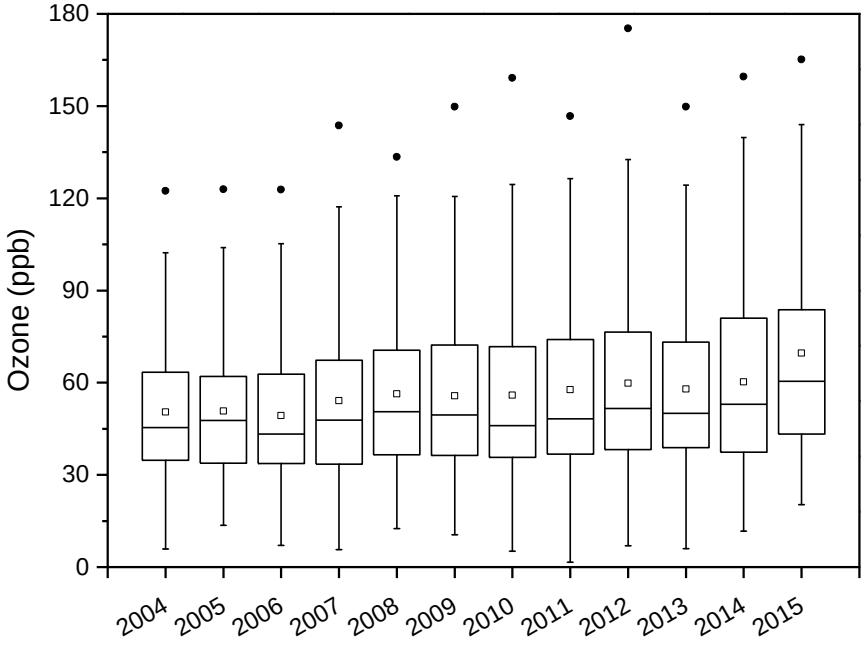

Fig 1. Percentile-box plot of yearly statistics of MDA8 values of ozone at SDZ, 2004-2015. Box depicts interquartile range and median; the square depicts the mean; whiskers depict 10th and 90th percentile; dot depicts maximum values.

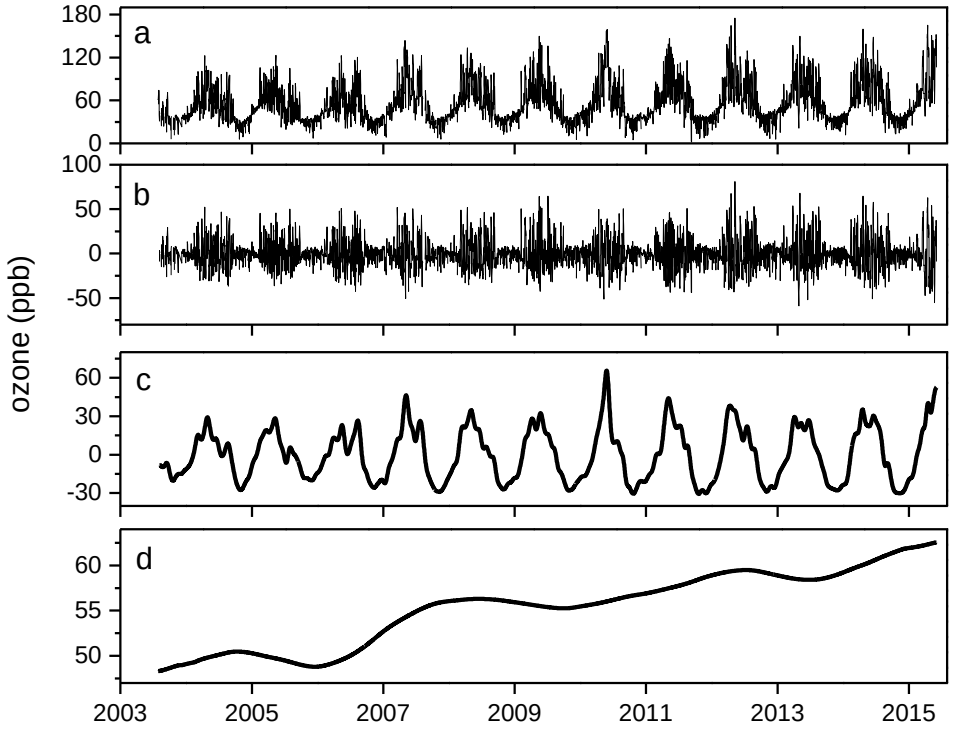

Fig 2. Separated time series of MDA8 values of ozone at SDZ: (a) the original data; (b) the short-term component, W(t); (c) the seasonal component, S(t); (d) the long-term component, e(t).

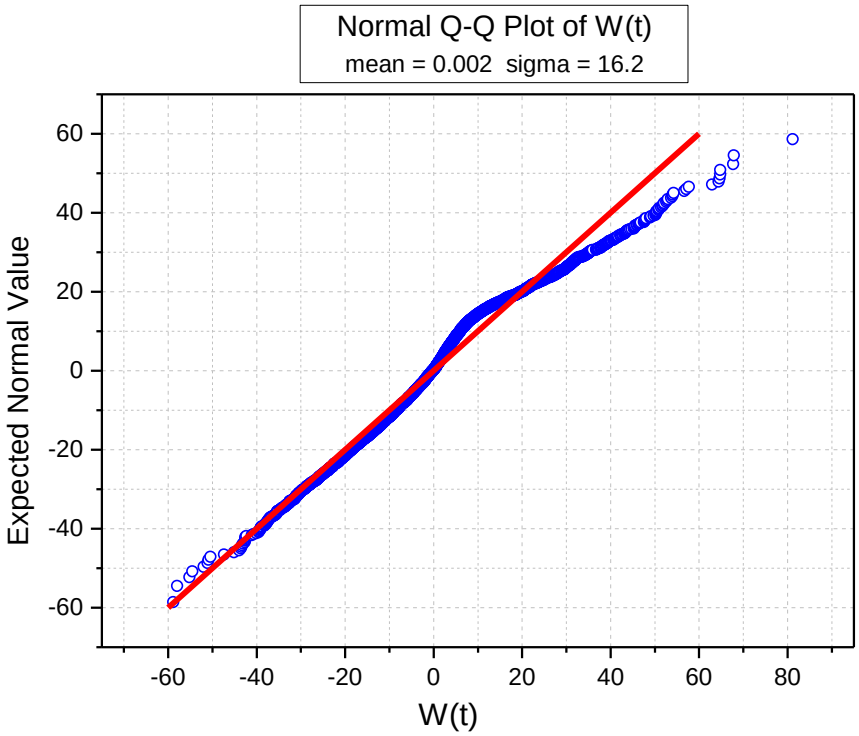

Fig 3. QQ plot of the short-term component W(t) for ozone.

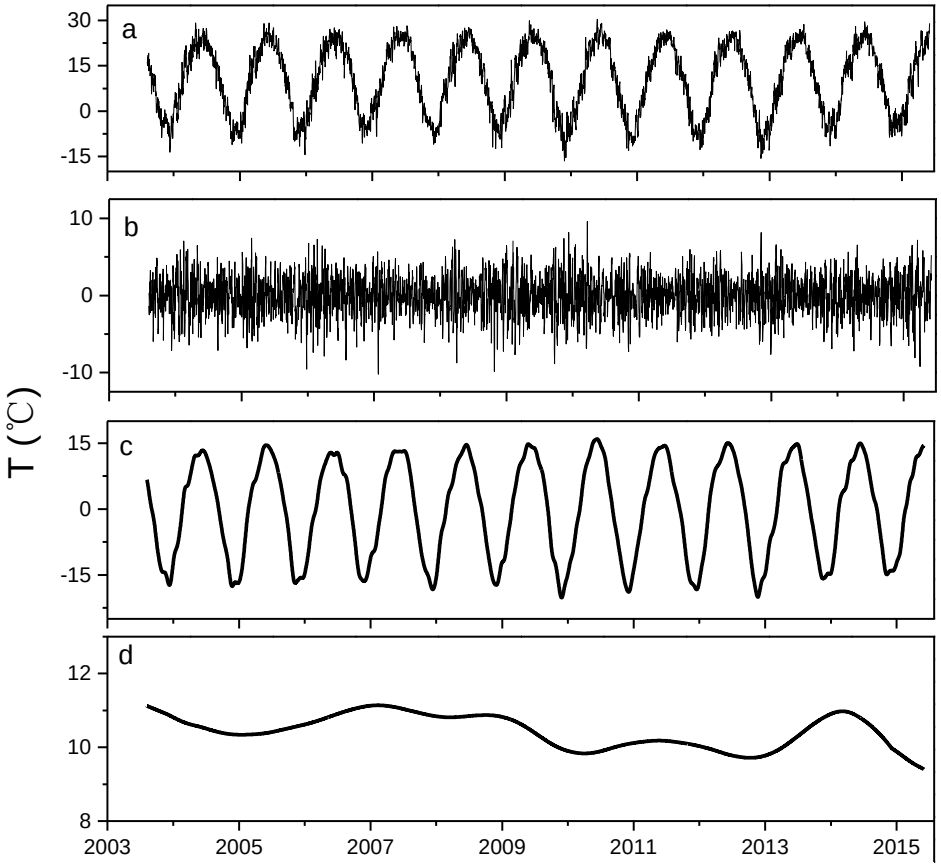

Fig 4. Separated time series of daily mean values of temperature at SDZ: (a) the

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

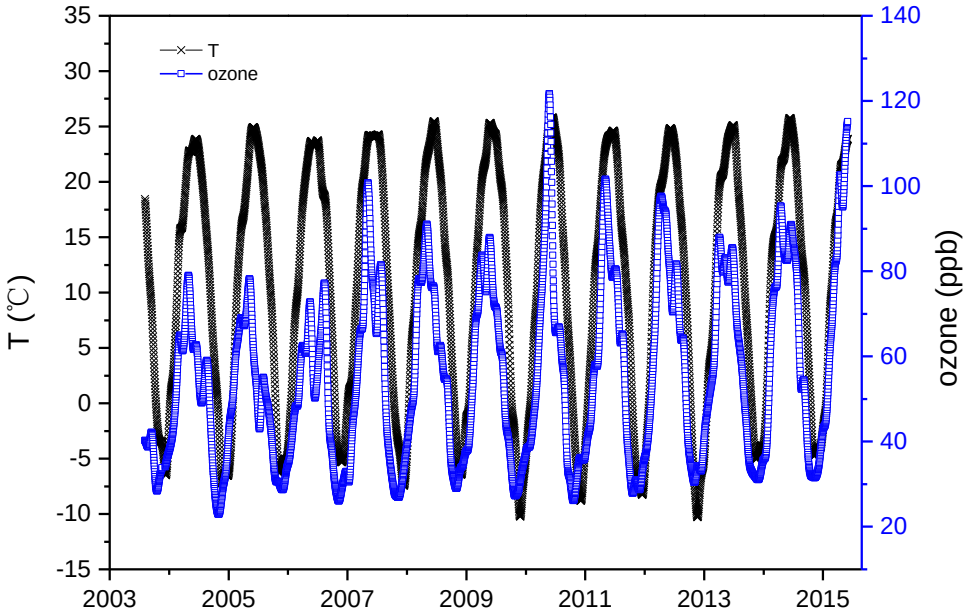

Fig 5. Results of the daily mean temperature and the MDA8 value of ozone after the application of $KZ_{15,5}$ filter to the original time series. The results indicate the sum of the seasonal and long-term components.

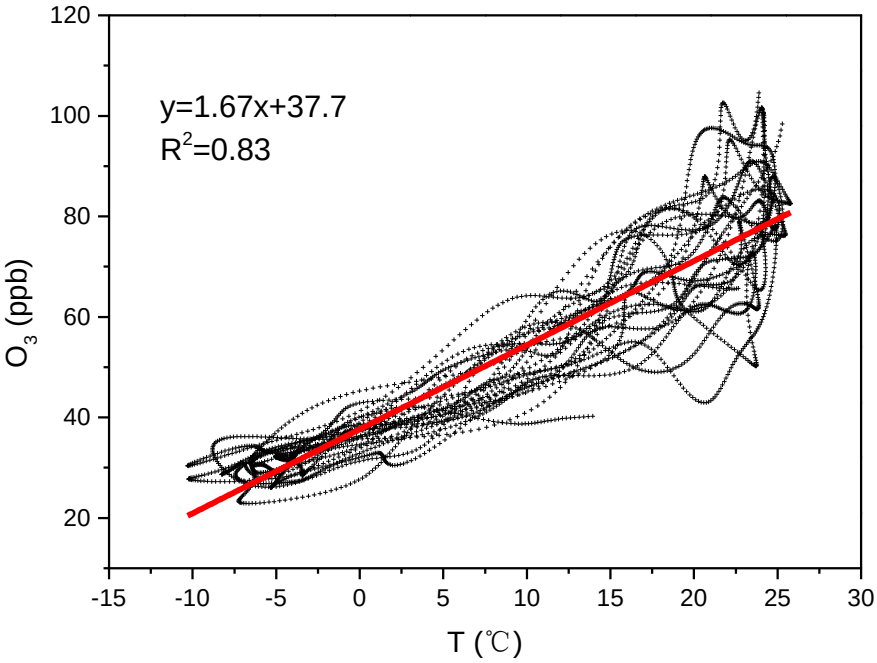

Fig 6. Linear regression fit on the filtered daily maxima of temperature and ozone.

Temperature data are lagged by 17 days to ozone data.

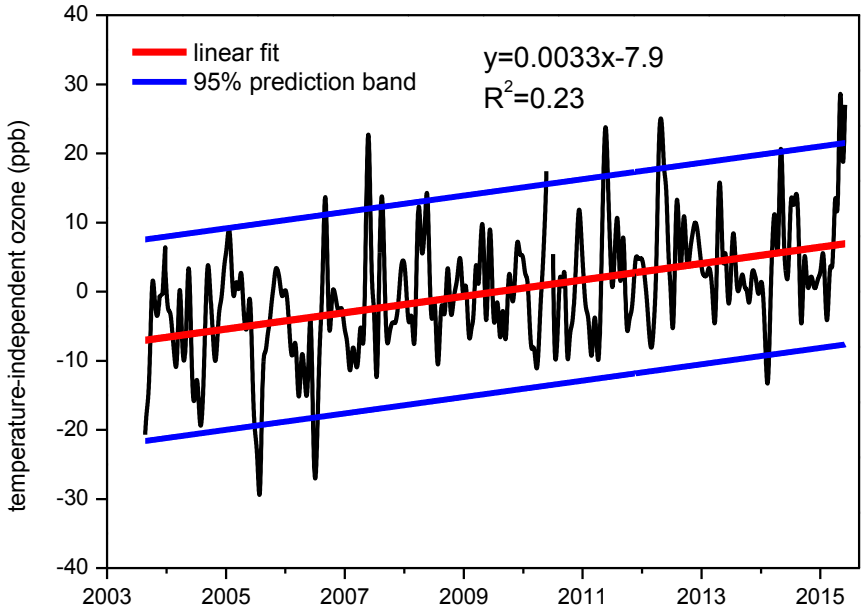

Fig 7. Time series of the noise-free and temperature-independent ozone. The red line is a linear fit and the blue lines are the 95% confidence prediction band.

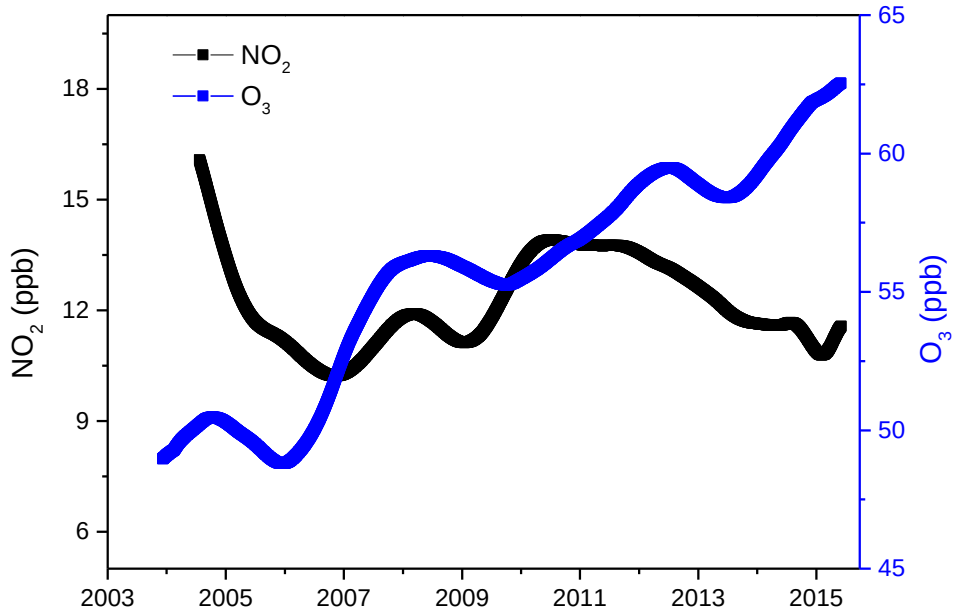

Fig 8. Long-term trends of NO_2 and the MDA8 ozone value calculated by $KZ_{365,3}$.