

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

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We thank the referees and editor for their very constructive comments and suggestions. We have revised our manuscript according to their comments and suggestions.

Referee 2: 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

C12718

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

C12719

year) for long term. We include the uncertainty of the rate of the daily maximum O₃ 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 $+1.13 \pm 0.01$ 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.bjgtgl.gov.cn/jgj/ywsj/index.html>, figure 1). The changes of the maximum value of O₃ 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.bjgtgl.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 O₃ production, such as solar radiation, relative

C12720

humidity. However, the correlation was only improved by no more than 0.5%.” “In summer, the temperature is not the dominant restricting factor for O₃ production compared to other factors, such as rainfall and precursor concentrations.” “Accordingly, we believe that the changes of VOCs emission and the ratio VOCs/NO_x might have caused the increase of surface O₃ 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 described 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 O₃ concentrations ($Okz(t)$) and the filtered Temperature lagged by 17 days ($Tkz(t+17)$) in this study was performed: $Okz(t) = aTkz(t+17) + b + \bar{\epsilon}(t)$ where a and b are fitted parameters, $\bar{\epsilon}(t)$ are the residuals of the relationship. Here, $\bar{\epsilon}(t)$ 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.

C12721

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.

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C12722



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

C12723