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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Worsening urban ozone pollution in China from 2013 to 2017 – Part 1: The complex and varying roles of meteorology" *by* Yiming Liu and Tao Wang

Anonymous Referee #1

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This paper provides a thorough analysis of the impact of meteorological variability on observed ozone changes across China from 2013 to 2017. The analysis is sound, for the most part, but there are a few inaccuracies that need to be addressed, as described below. Once these items are addressed I think the paper would be acceptable for publication in ACP.

Major comments: 1) The panels in Figure 1 are entirely too small and need to be increased by at least a factor of two, and rearranged on the page so that they fit. I had to enlarge the images on my computer to 400% and even then they were difficult to understand as the resolution was poor. Each panel has an inset in the lower right

corner, which doesn't seem to provide any information. These insets are distracting and should be removed. Likewise, the panels in Figures 3, 5 and 7 are also too small. For these figures you can expand the size of each panel by about 15% if you place the color bars underneath the panel, and move the labels on the left of the panels to positions above the panels. You can also delete the latitude and longitude labels, which aren't necessary. Then if you allow the panels to fill the full width of the page you should be able to make them significantly larger.

2) It would be helpful to place these 2013-2017 surface ozone changes in China within the context of broader trends across Asia, as well as long-term trends in the region of China. For example, Gaudel et al. use IAGOS observations to show that ozone in the lower and mid-troposphere has increased above China, India and Southeast Asia since 1994. Xu et al. show the long-term positive trend at Mt Waliguan, and Sun et al. show the positive trend at Mt. Tai. Wang et al. show the increase of ozone at Hok Tsui when transport is from the South China Sea. And Ziemke et al. show satellite retrievals that demonstrate a board increase of tropospheric column ozone across Asia and the tropics.

Gaudel, A, et al. 2018. Tropospheric Ozone Assessment Report: Present-day distribution and trends of tropospheric ozone relevant to climate and global atmospheric chemistry model evaluation. Elem Sci Anth, 6: 39. DOI: https://doi.org/10.1525/elementa.291

Sun, L, Xue, L, et al. 2016. Significant increase of summertime ozone at Mount Tai in Central Eastern China. Atmos. Chem. Phys. 16: 10637–10650. DOI: https://doi.org/10.5194/acp-16-10637-2016

Wang, T., Dai, J., Lam, K. S., Poon, C. N., and Brasseur, G. P. (2019), TwentyâĂŘfive years of lower tropospheric ozone observations in subtropical East Asia: The influence of emissions and weather patterns, Geophysical Research Letters, 46, https://doi.org/10.1029/2019GL084459

Xu, W, Lin, W, Xu, X, Tang, J, Huang, J, Wu, H and Zhang, X. 2016. Long-term trends of surface ozone and its influencing factors at the Mt Waliguan GAW station, China–Part 1: Overall trends and characteristics. Atmos. Chem. Phys. 16: 6191–6205. DOI: https://doi.org/10.5194/acp-16-6191-2016

Ziemke, J. R., Oman, L. D., Strode, S. A., Douglass, A. R., Olsen, M. A., McPeters, R. D., Bhartia, P. K., Froidevaux, L., Labow, G. J., Witte, J. C., Thompson, A. M., Haffner, D. P., Kramarova, N. A., Frith, S. M., Huang, L.-K., Jaross, G. R., Seftor, C. J., Deland, M. T., and Taylor, S. L.: Trends in global tropospheric ozone inferred from a composite record of TOMS/OMI/MLS/OMPS satellite measurements and the MERRA-2 GMI simulation , Atmos. Chem. Phys., 19, 3257-3269, https://doi.org/10.5194/acp-19-3257-2019, 2019.

3) Section 3.5 What is meant by “potential velocity”? Do you mean potential vorticity? Potential vorticity has long been used as in indicator of stratospheric intrusions into the upper and mid-troposphere, where it works very well, but it just doesn’t work for the lower troposphere or the surface because the signal decays by the time the intrusion reaches the lower troposphere (if it ever reaches the lower troposphere). Linking an increase of ozone at the surface to an increase of PV in the upper troposphere is just speculation. How do you know the ozone reaching the surface is from the stratosphere? Couldn’t it just be ozone from the mid-troposphere? (as shown by the IAGOS profiles in Gaudel et al. 2018, there is plenty of ozone in the mid-troposphere above China during the summer months) To provide a convincing argument that there was an increase of stratospheric ozone reaching the surface you will have to implement a conserved stratospheric ozone tracer in both MOZART and in CMAQ to see if there really is an increase of this tracer at the surface (see the papers by Meiyun Lin at NOAA GFDL, or papers by Andreas Stohl using the FLEXPART model). If you can’t run a tracer all you can say is that there is likely an increase of ozone transport from the free troposphere to the surface, but you don’t have any way of knowing if the ozone is from the mid-troposphere or if it’s from the stratosphere.

4) Line 304 It would be helpful to treat humidity in a consistent manner throughout the paper. In Table 1 you report values of relative humidity, while in Figure 5 your show specific humidity. Why show both types of humidity? From an ozone chemistry perspective specific humidity is most important because it scales with water vapor concentration. Relative humidity isn’t useful for understanding ozone photochemistry due to its non-linear relationship to water vapor concentration.

5) Line 322 This claim that precipitation can remove ozone is incorrect. The modeling study by Meleux et al. vaguely implies that precipitation removes ozone, but they don’t give any mechanism or explanation, and this claim goes against the long established fact that ozone has very low solubility in water (Wesely et al., 1981). I can’t think of any experimental studies that have shown that rain removes ozone from the air, although some studies have shown that chemicals in water (such as the ocean) can react with ozone if air bubbles are mixed into the ocean, or lakes (see the review by Monks et al., 2015, Atmos. Chem. Phys., 15, 8889–8973, 2015, www.atmos-chem-phys.net/15/8889/2015/ doi:10.5194/acp-15-8889-2015)

Wesely, M. L., Cook, D. R., and Williams, R. M.: Field measurement of small ozone fluxes to snow, wet bare soil, and lake water, Bound.-Lay. Meteorol., 20, 459–471, doi:10.1007/bf00122295, 1981.

Minor comments:

Line 45 Well, it’s not the relative humidity value that is important, but rather the number of water vapor molecules that are available. It would be best to replace relative humidity with water vapor.

Line 48 Change “Cloud has” to “Clouds have”

Line 49 I’m not sure what you mean by “cleaning efficiency”. Please use another term.

Line 50 How does the wet removal process increase ozone? Ozone is not water soluble. Is something else being removed by precipitation, which would otherwise destroy

ozone?

Line 65 If you are going to report ozone values in units of ppb, rather than in micrograms per cubic meter, you cannot use the term “concentration”. Instead, please use mixing ratio.

Line 81 implications (plural) observational data

Line 82 . . .based on the observations.

Line 104 Would sound better as: The equations for these statistical parameters can be found in Fan et al. (2013).

Line 114 . . .which is a few grids cells smaller. . .

Line 157-158 Please see how I modified the following sentence to improve the English. The authors can make similar changes throughout the document. Original: “Like the temperature, the simulated relative humidity was also slightly under-predicted and had a high correlation coefficient with the observation.” Corrected: “Like temperature, the simulated relative humidity values were also slightly under-predicted and had a high correlation coefficient with the observations.”

Line 164 . . . conditions on ozone levels.

Line 205 . . .emissions on ozone changes. . .

Line 217 . . .could be comparable to or. . .

Line 312 I don’t understand what is being said here: “Possible reasons for the ozone increase with the increase in the PBL height include low primary pollutant concentrations with the development of PBL” Are you saying that fresh emissions of NO can destroy ozone close to the surface in an urban environment, but if you have deep vertical mixing you can spread the NO vertically, which then limits ozone destruction at the surface?

Line 317 What is cleaning efficiency?

Line 365 Change “decreased” to “decrease”

Lines 383-386 These last two sentences should be revised. The first sentence is rather long and cumbersome and can be shortened as shown below. The second sentence is making a recommendation to policy-makers (by using the word “should”) and does not belong in a scientific paper. However, it’s perfectly fine to state how your results might be useful to policy-makers, by replacing “should” with “could” as shown below. “It is therefore necessary to consider meteorological variability when assessing the effectiveness of emission control policies on changes in the levels of ozone (and other air pollutants) in different cities and/or regions of China. Such an approach could be useful for the development of future air pollution mitigation policies.”

Figure 2, caption The second sentence is difficult to understand. The following change would help: In panel (b) only environmental monitoring sites (493) with data available in all 5 years are presented.

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