

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 reviewer for their constructive comments on our manuscript. We provide their comments and our responses below.

Referee 1:

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

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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 hire 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 km²) 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 back-

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ground 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,

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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-chemistry-and-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=SDZ2>

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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

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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 Schneidmesser, 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 Schneidmesser, 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,

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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 applies 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.

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Does the ozone correspond to Figure 2d?

Response: Indeed, O₃ and NO₂ showed in Figure 8 are the results of long-term trend calculated by KZ365,3, just as Figure 2d.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 31951, 2015.

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