Manuscript # ACP-2013-925

Responses to Reviewer #1

This study examines the impacts of the East Asian summer monsoon (EASM) on interannual variations of summertime surface-layer O_3 concentrations over China through a suite of sensitivity model simulations using the GEOS-Chem model. The impacts associated with EASM are also compared with the impacts from changes in anthropogenic emissions of ozone precursors. I feel this is an interesting topic which fits the scope of ACP well. The numerical experiments appear carefully designed. I would recommend publication of this paper after the authors address some relatively minor issues.

Major Comments:

1. P3270, L25-27 (and multiple places in the text) on the discussion of the impacts of EASM compared to that from changes in anthropogenic emissions: "We also find that the changes in the EASM strength are as important as the changes in anthropogenic emissions over 1986–2006 in influencing JJA surface layer O3 concentrations in China"

I think some clarification maybe needed to avoid/reduce the potential confusion for readers: The impacts on ozone associated with the interannual variations in EASM maybe particularly strong/important for certain regions and certain years, but that doesn't mean the EASM has been affecting the ozone over China the same or similar way anthropogenic emissions have.

Response:

Thanks for the suggestion. We have clarified this in the abstract as "We also find that the impacts the EASM strength on JJA surface-layer O_3 can be particularly strong (comparable in magnitude to the impacts on O_3 by changes in anthropogenic emissions over 1986–2006) for certain years. The largest increases in O_3 by anthropogenic emissions are simulated over southeastern China, whereas the largest impacts of the EASM on O_3 are found over central and western China."

2. P3280, L17: On the discussion related to high surface ozone over western China – I think that literature studies often show higher surface ozone over the eastern China than that over the western China, but that does not seem to be the case for this study? Any particular factors driving this? Some discussion on this would be helpful.

Response:

Summertime surface-layer O_3 concentrations were found to be high in both western and eastern China in previous observational and modeling studies. Yan et al. (1997) compared surface-layer O_3 concentrations measured at three background stations, including Lin'an station (30.4°N, 119.7°E, 132 m above sea level) in eastern China, Longfengshan station (44.7°N, 127.6°E, 325 m above sea level) in northeastern China, and Waliguan station (36.3°N, 100.9°E, 3816 m above sea level) in western China, and showed that summertime surface-layer O_3 concentrations were 31±8, 33±7, and 61±7 at Lin'an, Longfengshan, and Waliguan, respectively. Wang et al. (2006) also reported an average summertime O_3 concentration of 54 (±11) ppbv in 2003 at Mount Waliguan in western China.

Our simulated surface-layer concentrations of O_3 are high in both western and eastern China, which agree with the modeling studies of Wang et al. (2011), Jeong and Park (2013), and Lou et al. (2014). The high O_3 concentrations in western China are associated with the downward transport of O_3 from the stratosphere to troposphere (Wild and Akimoto, 2001; Wang et al. 2006; Ding and Wang, 2006), and the high O_3 concentrations in eastern China result from anthropogenic emissions (Wang et al., 2011).

We have added the above comparisons with previous studies and the mechanisms for high O_3 concentrations in western and eastern China in the first paragraph of Section 3.

Minor Comments:

1. P3270, L11: over the 21 yr → over the 21 yr period.

Response:

Changed.

2. P3270, L17: nation mean → national mean

Response:

Changed.

3. P3271, L2: incorrect statement; O3 is the third most important "anthropogenic" greenhouse gas (as reported by IPCC), but we can't say it's the third most important contributor to greenhouse effect (H2O should be the #1, but it's not an "anthropogenic" GHG).

Response:

We have revised the sentence as suggested: "It is also (after CO_2 and CH_4) the third most important anthropogenic greenhouse gas (Intergovernmental Panel on Climate Change (IPCC), 2007)."

4. P3274, L2: are simulated driven by → are simulated using

Response:

Changed.

5. P3278, L20-24: This section does not read well and easy to lead to some confusion; suggest to rewrite this section.

Response:

We have rewritten this section as "The typical features in winds during the EASM can be seen in Figs. 5a and 5b. The southerlies prevail in southeastern China in the lower and middle troposphere in JJA. Figs. 5c and 5d also present the differences in winds between the weakest and strongest EASM years (weakest minus strongest). In JJA, anomalous southerlies are found in southern China and anomalous westerlies are found in southeastern China, as the winds in the five weakest monsoon years are compared with those in the five strongest monsoon years. Such differences in winds between weak and strong EASM years were also reported in Li and Zeng (2002) and Huang (2004)."

6. P3283, L26: "with the largest values of exceeding" → "with the largest values exceeding"

Response:

Changed.

7. P3284, L11-12: "with a nation mean lower O3 concentration by" → "with the national mean O3 concentration lower by"

Response:

Changed.

8. P3284, L18-19: Tg should be Tg/season here, right?

Response:

Right. Changed.

References:

Ding, A., and Wang, T.: Influence of stratosphere-to-troposphere exchange on the seasonal cycle of surface ozone at Mount Waliguan in western China, Geophys. Res. Lett., 33, L03803, doi:10.1029/2005GL024760, 2006.

Huang, G.: An index measuring the interannual variation of the East Asian summer monsoon - The EAP index, Adv. Atmos. Sci., 21, 41–52, doi:10.1007/Bf02915679, 2004.

Jeong, J. I. and Park, R. J.: Effects of the meteorological variability on regional air quality in East Asia, Atmos. Environ., 69, 46–55, doi:10.1016/J.Atmosenv.2012.11.061, 2013.

Li, J. and Zeng, Q.: A unified monsoon index, Geophys. Res. Lett., 29, 115-1–115-4, doi:10.1029/2001GL013874, 2002.

Lou, S. J., Liao, H., and Zhu, B.: Impacts of aerosols on surface-layer ozone concentrations in China through heterogeneous reactions and changes in photolysis rates, Atmos. Environ., 85, 123–138, doi:10.1016/j.atmosenv.2013.12.004, 2014.

Wang, T., Wong, H. L. A., Tang, J., Ding, A., Wu, W. S., and Zhang, X. C.: On the origin of surface ozone and reactive nitrogen observed at a remote mountain site in the northeastern Qinghai-Tibetan Plateau, western China, J. Geophys. Res., 111, D08303, doi:10.1029/2005JD006527, 2006.

Wang, Y., Zhang, Y., Hao, J., and Luo, M.: Seasonal and spatial variability of surface ozone over China: contributions from background and domestic pollution, Atmos. Chem. Phys., 11, 3511–3525, doi:10.5194/acp-11-3511-2011, 2011.

Yan, P., Li, X. S., Luo, C., Xu. X. B., Xiang, R. B., Ding, G. A., Tang, J., Wang, M. L. and Yu, X. L.: Observational analysis of surface O_3, NO_x and SO_2 in China, Quarterly Journal of Applied Meteorology, 1997 (in Chinese).

Wild, O., and Akimoto, H.: Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, J. Geophys. Res., 106, 27729–27744, doi:10.1029/2000JD000123, 2001.

Responses to Reviewer #2

This manuscript presents several modeling simulations to understand the impact of Asian monsoon on the interannual variations of summertime ozone in China with a focus on the interannual variation. So far, very limited modeling works were conducted to understand ozone variation for such a long period. This topic of this paper is interesting, the modeling simulations were well-performed, and the main analysis and discussions were also well-organized. This referee would like to recommend the publication of this paper if the following points were addressed.

Major Comments:

1. One of the main weaknesses of this paper is modeling evaluation. It's acceptable that the authors only evaluated the modeling results using measurements at two sites, i.e. Hok Tsui and Ryori, which are the only available sites for a long-term measurement in Asia. However, there are so many works of ozone measurements, especially summertime ozone, have been done in differences places in China, including western China, the northern China and eastern China. The main mechanisms controlling summertime ozone, including monsoon, stratosphere-troposphere exchange, photochemistry etc., have already be widely discussed for different areas. Although these previous works were not on an interannual scale, the authors

should make a more complete literature review on these works and have these main findings appropriately discussed and compared.

Response:

Following the reviewer's suggestion, we have added a number of studies with measurements of O_3 in the first paragraph of the introduction section. The newly added studies reported ozone measurements in western China (Tang et al., 1995; Zhu et al., 2004; Ding and Wang, 2006; T. Wang et al., 2006a) and eastern China (Cheung and Wang, 2001; Gao et al., 2005; Chou et al., 2006; Shao et al., 2006; T. Wang et al., 2006b; Z. Wang et al, 2006; Ding et al., 2008; Xu et al., 2008; Wang et al., 2009; Wang et al., 2011).

The GEOS-Chem simulations of present-day surface-layer O_3 concentrations in China have been evaluated in Wang et al. (2011) and Lou et al. (2014). Wang et al. (2011) demonstrated that the model captured well the magnitude and seasonal variation of surface-layer concentrations and column burdens of O_3 in China. Lou et al. (2014) reported that the simulated O_3 in China agreed fairly well with measurements collected from the literature with an average high bias of 9%.

For comparisons with previous studies and for clarifying the mechanisms of high O_3 concentrations in China, we have added the following sentences in the first paragraph of Section 3: "Our simulated surface-layer concentrations of O_3 are high in both western and eastern China, which agree with the modeling studies of Wang et al. (2011), Jeong and Park (2013), and Lou et al. (2014). The high O_3 concentrations in western China are associated with the downward transport of O_3 from the stratosphere to troposphere (Wild and Akimoto, 2001; T. Wang et al. 2006b; Ding and Wang, 2006), and the high O_3 concentrations in eastern China result from anthropogenic emissions (Wang et al., 2011)."

2. There is a clear definition of monsoon and non-monsoon regions in the East Asia. The simulation results show that there is strong interannual variation of ozone in the western China (Fig. 3b) and over the Siberia, which are typically considered as non-monsoon regions. The authors should give some explains on this. Also, it will be better to explore the relationship between EASMI and ozone for monsoon and non-monsoon regions separately but not for the whole China.

Response:

We had the same thoughts as the reviewer at the beginning of our study; we tried to examine interannual variations of O_3 in eastern China and western China. Model results showed that the maximum O_3 changes by the EASM were located in northeastern China, southwestern China, and over or near the Tibetan Plateau (Figure 4 of our manuscript), which surprised us for some time. However, monsoon and non-monsoon regions defined by previous studies (Gao, 1962; An et al., 2000) actually showed that monsoon region covers almost the whole China except for the northwestern region (see Figure A below). Our simulated monsoon-induced changes in O_3 in China (Figure 4 of our manuscript) are mostly within the monsoon region.

The changes in O_3 over the Siberia are large (Figure 4 of our manuscript), which can be explained by the anomalous northerlies over the north border of China between the five weakest and strongest EASM years (weakest–strongest) (Figure 5 of our manuscript) that transport O_3 to China.

We have added the above discussions in the second paragraph of Section 4 in the revised manuscript.

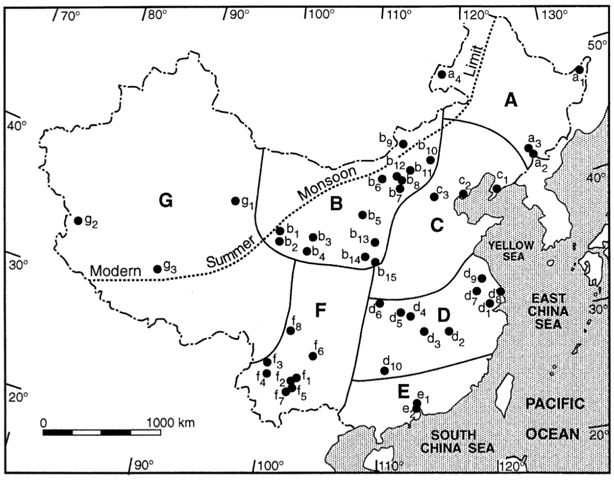

Fig. A. Monsoon and non-monsoon regions in China from An et al. (2000). North of Modern Summer Monsoon Limit is non-monsoon region, and south of Modern Summer Monsoon Limit is monsoon region.

Minor Comments:

1. The "whole China": is it for a definition of box in latitude and longitude or exactly according to the territory of China?

Response:

"Whole China" is defined by the national borders of China. We have clarified this in the manuscript.

2. For the Hok Tsui and Ryori site, it will be better to have the site location marked on a figure, for example on the Figure 2.

Response:

Site location is added in Figure 2.

3. Page 3282, first paragraph of Sect 5.4: Figure 8 or Figure 8f should be change into Figure 7 and 7f.

Response:

Changed.

4. For the data at Ryori, it looks that the result in 1992 was unusual. Please check the original data; also please pay attention to the data coverage for that year. If it's due to measurement problem, that data point can be removed and the correlation can be higher.

Response:

Thanks for pointing this out. We had a bug in the code for 1992. See below for the revised Figure 2.

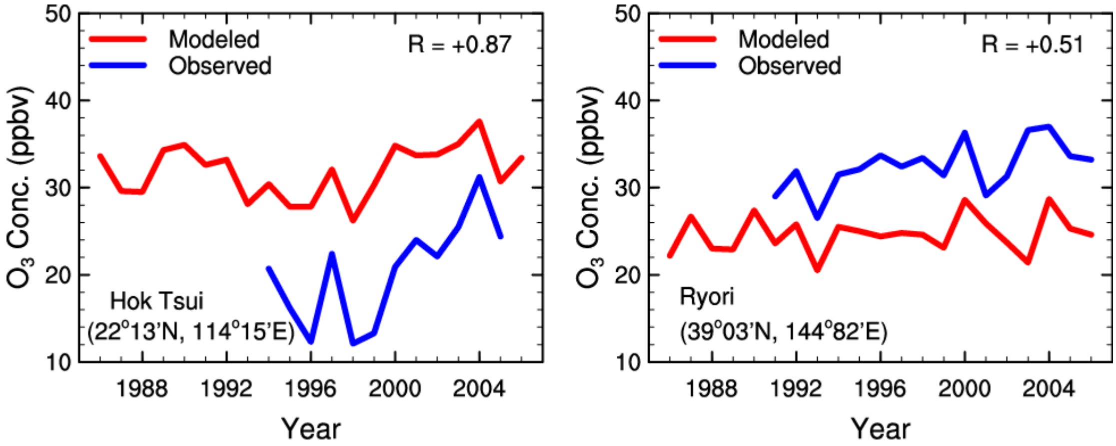

Fig. 2. Comparisons of observed and simulated JJA mean surface-layer O_3 concentrations at Hok Tsui (22°13' N, 114°15' E) in Hong Kong (left) and Ryori (39°03'N, 144°82'E) in Japan (right). Correlation coefficient between simulations and observations is shown at top right corner of each panel, which is calculated over the time period with observations available.

References:

An, Z., Porter, S. C., Kutzbach, J. E., Wu, X., Wang, S., Liu, X., Li, X., Zhou, W.: Asynchronous Holocene optimum of the East Asian monsoon, Quaternary Science Reviews, 19, 743–762, doi:10.1016/S0277-3791(99)00031-1, 2000.

Cheung, V. T. F. and Wang, T.: Observational study of ozone pollution at a rural site in the Yangtze Delta of China, Atmos. Environ., 35, 4947–4958, doi:10.1016/S1352-2310(01)00351-X, 2001.

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.

Ding, A., and Wang, T.: Influence of stratosphere-to-troposphere exchange on the seasonal cycle of surface ozone at Mount Waliguan in western

China, Geophys. Res. Lett., 33, L03803, doi:10.1029/2005GL024760, 2006.

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.

Gao, J., Wang, T., Ding, A. J., and Liu, C. B.: Observational study of ozone and carbon monoxide at the summit of mount Tai (1534 m a.s.l.) in central-eastern China, Atmos. Environ., 39, 4779–4791, doi:10.1016/j.atmosenv.2005.04.030, 2005.

Gao, Y. X.: Some Problems on East-Asia Monsoon, Science Press, Beijing, 49–63, 1962 (in Chinese).

Jeong, J. I. and Park, R. J.: Effects of the meteorological variability on regional air quality in East Asia, Atmos. Environ., 69, 46–55, doi:10.1016/J.Atmosenv.2012.11.061, 2013.

Shao, M., Tang, X. Y., Zhang, Y. H., and Li, W. J.: City clusters in China: air and surface water pollution, Front. Ecol. Environ., 4, 353–361, doi:10.1890/1540-9295(2006)004[0353:CCICAA]2.0.CO;2, 2006.

Lin, J. T., Patten, K. O., Hayhoe, K., Liang, X. Z. and Wuebbles, D. J.: Effects of future climate and biogenic emissions changes on surface ozone over the United States and China, J. Appl. Meteorol. Clim., 47, 1888–1909, doi:10.1175/2007JAMC1681.1, 2008.

Lou, S. J., Liao, H., and Zhu, B.: Impacts of aerosols on surface-layer ozone concentrations in China through heterogeneous reactions and changes in photolysis rates, Atmos. Environ., 85, 123–138, doi:10.1016/j.atmosenv.2013.12.004, 2014.

Tang, J., Wen, Y. P., Xu, X. B., Zheng, X. D., Guo, S., and Zhao Y. C.: China Global Atmosphere Watch Baseline Observatory and its measurement program, in CAMS Annual Report 1994–95, pp. 56–65, China Meteorol. Press, Beijing, 1995.

Tu, J., Xia, Z., Wang, H. and Li, W.: Temporal variations in surface ozone and its precursors and meteorological effects at an urban site in China, Atmos. Res., 85, 310–337, doi:10.1016/j.atmosres.2007.02.003, 2007.

Wang, T., Ding, A. J., Gao, J., and Wu, W. S.: Strong ozone production in urban plumes from Beijing, China, Geophys. Res. Lett., 33, 1–5, doi:10.1029/2006GL027689, 2006b.

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.

Wang, T., Wong, H. L. A., Tang, J., Ding, A., Wu, W. S., and Zhang, X. C.: On the origin of surface ozone and reactive nitrogen observed at a remote mountain site in the northeastern Qinghai-Tibetan Plateau, western China, J. Geophys. Res., 111, D08303, doi:10.1029/2005JD006527, 2006a.

Wang, Z. F., Li, J., Wang, X. Q., Pochanart, P., and Akimoto, H.: Modeling of regional high ozone episode observed at two mountain sites (Mt. Tai and Huang) in East China, J. Atmos. Chem., 55, 253–272, doi: 10.1007/s10874-006-9038-6, 2006.

Wang, Y., Zhang, Y., Hao, J., and Luo, M.: Seasonal and spatial variability of surface ozone over China: contributions from background and domestic pollution, Atmos. Chem. Phys., 11, 3511–3525, doi:10.5194/acp-11-3511-2011, 2011.

Wild, O., and Akimoto, H.: Intercontinental transport of ozone and its precursors in a three-dimensional global CTM, J. Geophys. Res., 106, 27729–27744, doi:10.1029/2000JD000123, 2001.

Xu, X., Lin, W., Wang, T., Yan, P., Tang, J., Meng, Z., and Wang, Y.: Long-term trend of surface ozone at a regional background station in eastern China 1991–2006: enhanced variability, Atmos. Chem. Phys., 8, 2595-2607, doi:10.5194/acp-8-2595-2008, 2008.

Yan, P., Li, X. S., Luo, C., Xu. X. B., Xiang, R. B., Ding, G. A., Tang, J., Wang, M. L. and Yu, X. L.: Observational analysis of surface O3, NOx and SO2 in China, Quarterly Journal of Applied Meteorology, 1997 (in Chinese).

Yan, P., Wang, M., Cheng, H., Zhou, X.: Distributions and variations of surface ozone in Changshu, Yangtze Delta region, Acta Metallurgica Sinica, 17, 205–217, 2003.

Zhu, B., Akimoto, H., Wang, Z., Sudo, K., Tang, J., and Uno, I.: Why does surface ozone peak in summertime at Waliguan?, Geophys Res Lett, 31, doi:10.1029/2004gl020609, 2004.