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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The relative importance of various source regions on East Asian surface ozone" *by* T. Nagashima et al.

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Response to the comment of referee #2

The authors greatly appreciate your critical reading of our manuscript. First of all, we are really sorry to inform you that we have made an error in the selection of the time of model 1-hourly data to draw Figure 6. We picked up the model data from 10:00 to 16:00 of UTC, but we should select those of JST (Japan Standard Time). The difference between UTC and JST is 9 hours, so we re-draw Figure 6 using 10:00 to 16:00 of JST data. The corrected Figure 6 (Fig.1, below) shows similar characteristics to the previous one, but there are several differences between two figures. The most notable changes from the previous one are (1) The upper boundary of frequency distribution for most months extends to larger O3 range by 10 ppbv, and (2) The drop in ridge line

in summer becomes very weak. Therefore, we changed the description about Figure 6 in the revised manuscript.

Our response to each comment is in the following.

This paper presents the relative contribution of various source regions on surface ozone over East Asia using a tagged tracer method with a global-scale chemical transport model. Ozone tracers are tagged from a number of source regions, and the results are presented for East Asian receptor regions by separating the contribution of PBL, FT and ST ozone. My major concern is that the relationships between PBL/FT ozone and individual ozone precursor emissions regions should be further clarified. For example, FT ozone abundance can result from either inter-continental transport or lofting of local pollution. Simply FT contribution can not tell whether this is long-range transport or local pollution. My second suggestion is that the authors should include some discussion about the comparison of the S-R relationships with those in the HTAP papers, at least for EU->EA, and NA->EA results, since a different method is applied in this study. Some recent HTAP papers are published in the ACP/EMEP special issue, http://www.atmos-chem-phys.net/specialissue152.html. I would recommend publishing this paper in ACP after major revisions and some further clarifications described below:

→ We agree with your concern that FT contribution is not well clarified in the present manuscript. We added some sentences in revised manuscript to clarify the FT contribution (please also refer to our responses to your comments No.4 and 9). We also added some comparison with HTAP results in the revised manuscript.

Specific comments: 1. P9079, L15-20, Some references are needed here

→ There is annual report of the status of air pollution in Japan issued by the Ministry of Environment (MOE) of Japan, but it is written only in Japanese. As long as I have researched, there are no suitable references written in English. Since there seems to be no other choice but to refer to the MOE Japan report, we cited the MOE of Japan report.

2. P9080, L4-8, There is a major discrepancy in the increasing rate of NOx emissions between bottom-up emission inventories and satellite data. So the word "verified" is misleading here.

→ We agree. We changed “verified” to “inferred” in order to avoid misleading.

3. P9085, L15-18, Why there are more regions designated in the stratosphere than in the PBL and FT?

→ The total number of source regions is 45, which includes 22 regions both in PBL and FT and one whole stratosphere (22+22+1=45). We changed the sentence to make it clear.

4. P9085, L18-25, Simply defining the PBL as the six lowermost layers in the model, is somewhat arbitrary. Is this definition consistent with model simulated PBL height? How about the diurnal variation of PBL height?

→ The PBL height diagnosed in the model can greatly vary depending on location and season, and it also has large diurnal variation. The modeled PBL height is ranging from 500-600 m over the ocean or nocturnal continent to some 1000 m over the daytime continent. Because the height of the 6 lowermost layers of the model does not show such great variability but stays at about 2-2.5 km, the large variation in PBL thickness in the model can not be exactly represented by the 6 lowermost layers of the model. However, we do not intend to separate the exact PBL from the rest of the troposphere (FT). As shown in the Figure 1 of Sudo and Akimoto (2007), in many cases, O3 production rate calculated by the model largely increase below about 2 km, which is close to the average height of the 6 lowermost layers, not necessarily corresponding to the PBL height. Therefore, we separate the troposphere vertically into two regions by using the 6th lowermost layer as the boundary, and use the terms “PBL” and “FT” to name each region. The procedure to define the PBL like this (by fixed number of layers in the model) can be found in many previous literatures (e.g, Wang et al., 1998; Fiore et al., 2002; Yamaji et al., 2008).

- Wang, Y., Jacob, D. J., and Logan, J. A.: Global simulation of tropospheric O3-NOx-hydrocarbon chemistry: 3. Origin of tropospheric ozone and effects of nonmethane hydrocarbons, J. Geophys. Res., 103(D9), 10757-10767, 1998.

- Fiore, A. M., Jacob, D. J., Bey, I., Yantosca, Y. M., Field, B. D., Fusco, A. C., and Wilkinson, J. G.: Background ozone over the United States in summer: Origin, trend, and contribution to pollution episodes, J. Geophys. Res., 107(D15), 4275, doi:10.1029/2001JD000982, 2002.

- Yamaji, K., T. Ohara, I. Uno, J. Kurokawa, P. Pochanart, and H. Akimoto: Future Prediction of Surface Ozone over East Asia using Models-3 Community Multiscale Air Quality Modeling System and Regional Emission Inventory in Asia, Journal of Geophysical Research, 113, D8306, doi:10.1029/2007JD008663, 2008

5. P9109, L1-15, the spring/early-summer maximum and summer minimum behavior of surface ozone along the East Asian coast has been well discussed in the literature, including the effects of regional pollution, biomass burning, monsoonal circulations and clouds. Findings from these papers should be acknowledged and described here: Ding,A.J. et al: Tropospheric ozone climatology over Beijing: analysis of aircraft data from the MOZAIC program, Atmos.Chem.Phys., 8(1),1-13,2008. He,Y.J. et al: Significant impact of the east Asia monsoon on ozone seasonal behavior in the boundary layer of eastern china and the west Pacific region, Atmos.ãĂĂChem.ãĂĂPhys.,ãĂĂ8(4),ãĂĂ14927ïÂą'lC14955,2008 Lin, M. et al: Multi-scale model analysis of boundary layer ozone over East Asia, Atmos. Chem. Phys., 9, 3277-3301, doi:10.5194/acp-9-3277-2009

→ Thank you for letting us know useful papers. We referred these studies in Section 3.2.

6. P9091, L1-5, Discussion on the Japanese mountainous site (Happo): In addition to the topographical circulation, modeled excessive removal of ozone by dry deposition during the night may also contribute to the bias at the mountain site located above

the nocturnal boundary layer (Lin M. et al., acp-9-3277-2009). Did you use modeled surface ozone for comparison? Since the model resolution is relatively coarse, consider interpolating the model results according to the pressure altitude of the observation site. The vertical interpolation should partly reduce the bias.

→ We have already interpolated model O3 to the altitude of the observation site for Figure 2. This vertical interpolation could reduce some part of the bias, but there still have bias as depicted in Figure 2.

7. P9091, L7-15, Will the inadequate treatment of emission seasonality and/or the stratosphere-to-troposphere transport also contribute to the underestimate of surface ozone over northern Japan during winter and spring? Do your emissions have a seasonal variation? If not, certainly the underestimate of winter heating emissions from Northeast Asia should play a role in the model bias in the cold season. I am not fully convinced if decreasing dry deposition in wintertime is reasonable. First, the model reproduces wintertime ozone at the Mondy site located in the eastern Siberia reasonably well, suggesting that dry deposition may not be a major issue. Second, by decreasing dry deposition, the lifetime of ozone in winter will increase substantially, and thus affecting the budget of long-range transport contribution during winter. Your results show that "More than half of surface ozone is attributable to the ozone transported from distant sources outside of East Asia in the cold season (October to March)". Is this consistent with the estimates reported in the literature (e.g., HTAP papers)? Will the treatment of temperature-dependent dry deposition have an influence on the budget?

→ The anthropogenic emissions employed in this study (REAS, EDGAR3.2FT2000) have no seasonal variation, so winter heating emission that you mentioned would be underestimated in our simulation, but it is difficult to show how much of model bias is attributable to the underestimation in heating emission. We mentioned the heating emission as a possible cause for the underestimation of surface O3 in the revised manuscript. → At the Mondy site, the model significantly underestimated the winter/spring surface O3 before introducing the decreased dry deposition at lower tem-

perature. As you mentioned, the introduction of decreased dry deposition affects the budget of O3, but the effect on the budget is very small; the global annual mean deposition decrease only 1∼2 % (∼15Tg/yr). Compared with the HTAP model intercomparison results (Fiore et al., 2009), our estimated contributions of Europe to East Asia and North America to East Asia are well inside the uncertainty among models. Considering these things, we think it is reasonable to introduce the reduced dry deposition at lower temperature.

8. P9097, L9-12, Discussion on inter-annual variations of source-receptor relationships: Please clarify if your emissions have an inter-annual variation and how it may affect the variations of S/R relationships.

→ For anthropogenic emissions in Asia, we use REAS data which vary by year. But for those in the rest of the world, we use the emission data of the year 2000 (EDGAR3.2 FT2000) for all years. For biomass burning emission, we use the year 2000 data of RETRO-fires for all-years. We added a sentence to describe this in Section 2.3.

→ It is difficult to solely show the impact of interannual change in emissions on that in S-R relationship, because we inter-annually changed the emission (REAS) and meteorology simultaneously. However, we guess that the main contributor to inter-annual variation of S-R relationship for O3 in our model is not that of emission but meteorology. First, Kurokawa et al. (2009) show that the inter-annual variations of springtime O3 over Japan are largely controlled by the variation in meteorology during the last couple of decades by using the same anthropogenic emission data (REAS) with CMAQ model. Second, we performed additional experiment with fixed chemical production (P) and loss frequencies (L) at the year 2000 but with inter-annually varied meteorology, and the resulted inter-annual variation in S-R relationship is similar to the current results. This suggests that the inter-annual variation in meteorology is the main cause of that in S-R relationship. We mentioned these in the revised manuscript.

- Kurokawa, J., Ohara, T., Uno, I., Hayasaki, M., and Tanimoto, H.: Influence of mete-

orological variability on interannual variations of springtime boundary layer ozone over Japan during 1981–2005, Atmos. Chem. Phys., 9, 6287–6304, 2009

9. P9093, L10-15 (Figure 4), Please clarify if the contributions of FT are identical to contributions of O3 transported from outside of the East Asian PBL? How do you separate out the East Asian sources that are transported to the FT and brought back to PBL again, from the far distance sources like NA and EU? In other words, the FT contribution includes the EA O3 which are lofted into the FT during cold surges and then mixed down to PBL again during subsidence events?

→ In the current form of tagged tracer method, the contribution of FT, say, of CHN represents the O3 chemically created inside the free troposphere (from the 7th vertical layer from the surface to the tropopause) over China with no regard to the origin of its precursors. Therefore, the contribution of FT of CHN can include the O3 created from the precursors emitted in every source regions in the world. Once the O3 is created inside the FT of CHN, it is regarded (tagged) as the contribution of FT of CHN wherever it will be transported; even it will be transported down to PBL of any regions. As to the last question in your comment, therefore, if "EA O3" means the O3 created in the PBL of EA, FT contribution does not include it. But, if "EA O3" means O3 precursors emitted in EA, FT contribution can include the O3 which is created in FT from those precursors.

10. P9098, L28 - P9099, L5. Discussion on nighttime ozone behavior is confusing since you indicated in P9098, L8-12 that only the day time data (10:00-16:00) data are used.

→ We agree that the discussion on nighttime chemistry is confusing. The failure at lower O3 concentrations in non-summer season exists in spite of limiting the comparison to daytime O3. In observation, very low O3 (less than 20 ppbv) in daytime typically occurred in the morning and the evening as a transitional stage between the nighttime depleted O3 and the afternoon maximum. The daytime setting of 10:00-16:00 catches this very low O3 in the morning in the observation. Since the model can not adequately

simulate the nighttime O3 depletion in urban area, the very low O3 in the morning in the transitional stage also can not be simulated in the model. We added the above sentence in the revised manuscript.

11. P9099, L7-8: This sentence is confusing. Why are the hourly data not suitable to discuss the high-ozone events? Did you mean coarse-resolution model results?

→ What we want to mean by this sentence is exactly what you guess. We want to say that the model with coarse resolution can not represent extra-high hourly O3, like over 110 ppbv, therefore, the model results are not suitable for the discussion of extra events. We changed the sentence.

12. Figure 6 and Figure 7: The global model missed the peak of local pollution events during summer (Figure 6). Then the statistics presented in Figure 7 are not reliable at all, especially for the high (60-90ppbv) and extra-high (>90ppbv) classes of ozone.

→ The former Figure 6 is incorrect and the correct figure shows that our model reasonably reproduce the observed behavior of O3 at least in high O3 class (60-90ppbv) in the spring on which the Figure 7 focused. In extra-high O3 class (>90ppbv), our model still miss super-high O3 events (>100ppbv in April, and >110 ppbv in May) but can simulate several events over 90 ppbv with similar frequency of occurrence to observation. Therefore, we think looking at the statistics shown in Figure 7 for the spring is worthwhile.

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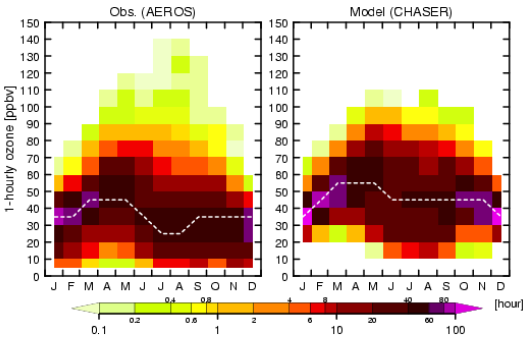

Fig. 1. Revised Figure 6