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Atmospheric Chemistry and Physics Discussions

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Interactive Comment

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

T. Nagashima et al.

nagashima.tatsuya@nies.go.jp

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Response to the comment of referee #1

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

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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.

General comments

The manuscript presents a detailed source attribution for surface ozone over several regions within East Asia. While the methods are not new, their application yields new information regarding seasonal cycles of surface ozone over this region, and the emphasis on intra-continental transport is complementary to a recent international coordinated effort focused on intercontinental transport, mentioned in the introduction. It would be useful to discuss how the findings presented here compare with, extend, or contradict the conclusions coming out of that effort. Throughout the text, the English could use some improvement.

→We added the comparison between the current results and the results coming from the international coordinated effort (HTAP) in the revised manuscript.

Specific comments

Abstract: "Tagged tracer method" should be explained.

→ We added a brief explanation of "tagged tracer method" in Abstract.

Abstract: Rather than "recent years", give the specific years examined.

→ We change the manuscript as you pointed out.

L14: What is increasing greatly in summer?

→ Domestically-created O3 is increasing in summer. We changed the sentence to avoid ambiguity.

The discussion in Section 2 overlaps with that in 2.2 and could be combined.

→ In the first part of Section 2, a brief and concise explanation of tracer tagging method

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is given in order to easily seize the difference between two methods, and then we give more intensive explanation in Section 2.2. We believe this two-step explanation can offer a close and comprehensible explanation of tracer tagging method to readers. We know this would introduce some redundancy, but we do not think it is fatal one. Therefore, we want to keep these sentences unchanged.

Section 2.1. awkward phrasing final sentence of first paragraph – is the stratospheric model output assimilated into CHASER? Does this vary interannually?

→ We are really sorry, but we could not catch the point of this comment. Here, we mean that O3 and NOy species above the tropopause in CHASER are approximated to the stratospheric model output. We used almost the same phrase at P9085 L3-4.

→ Yes, the output of stratospheric model varies interannually; we added this in the sentence.

Section 2.2 The treatment of the stratospheric tracer needs to be better explained. As shown by Hess and Lamarque (Hess , P. G. and J. F. Lamarque (2007), Ozone source attribution and its modulation by the Arctic oscillation during the spring months, J. Geophys. Res., 112, D11303, doi:10.1029/2006JD007557), the conclusions will depend on how this tracer is defined, with a large sensitivity in the northern hemisphere. They argue that an approach similar to the one employed here will overestimate the stratospheric contribution. Some discussion is appropriate.

→ Actually, there is large difference in estimated stratospheric contribution between the methods to deduce it. The method in Hess and Lamarque (2007) produce much smaller stratospheric contribution than the method used in our study (also in many previous studies). They argued that their method has some conceptual advantages over the current method, but there has not been a consensus on which method is better for estimating the stratospheric contribution. Therefore, we think it is plausible to employ the current method for estimating the stratospheric portion of O3 in our study. We added some sentences to describe this situation.

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Section 2.2 Why does the concentration of the tagged tracer need to be scaled at all? Shouldn't the sum of the tracers equal the total ozone?

→ In our simulation, what we exactly calculated are tagged "Ox" tracers, since the temporal evolution of these tracers are calculated using P(Ox) and L(Ox). The sum of the tracers (= total Ox) generally shows good agreement with the total O3 calculated in the standard full-chemistry run; the annual global mean difference between the two is well within 3 % in the troposphere. However the total Ox tends to exceed the total O3 in the polluted boundary layers, because the amounts of Ox species other than O3 (e.g., NO2, HNO3, PAN, etc.) can not be negligible in such region.ãĂĂIn the polluted region, O3 can only account for about 60 % of total Ox in some cases, but O3 is still the dominant component of Ox even in those cases. The difference between the total Ox and total O3 is unavoidable in the current form of tagged tracer method. Therefore we decided to scale the tagged tracers so that the sum of the tracers becomes equal to the total O3.

Section 2.3 Do the emissions vary by year?

→ 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

Section 2.3 Do the biogenic and lightning sources respond to the model meteorology?

→ Biogenic emissions of VOCs do not respond to the model meteorology, but NOx emission from lightning can respond to the model meteorology since the lightning NOx emission parameterization is linked to the convection scheme in the AGCM.

Section 3.1 The discussion jumps around here and could be more clearly organized. Same for Section 3.2.

→ Here, we compared the observations and the model results by picking up the points

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of difference or those well simulated. Consequently, the description tends to have a flavor of jumping around those points. However, we tried to organize this section in order as the comparison in inland and maritime remote regions is firstly described in Section 3.1 and the seasonal cycles in East Asian region is secondly focused on in Section 3.2. We want to keep this organization of the section, but in order to make our intension clearer, we merged Section 3.1 and 3.2 into one section and reconstruct it into two subsections, each of which is titled appropriately. Of course, your suggestion for the title of Section 3.2 is adopted.

P9089 L10 Plus also a lack of summertime photochemistry at these sites?

→ As to the sites mentioned in this sentence, a "lack" of photochemistry in summer is overstatement. There is non-negligible contribution of O_3 created in PBL even in summer at these sites, although the peak of the PBL contribution occurs in the spring.

P9089 L14. Why are the model estimates for stratospheric contribution here lower?

→ STE flux of O_3 estimated in our simulation is about 120 Tg/yr; this is smaller than those estimated in previous literatures (∼500 Tg/yr) and would be the cause of lower estimated stratospheric contribution in our model. The main cause of small STE flux in our model would be the O_3 data assimilated in the stratosphere. The data is taken from the output of stratospheric chemistry model simulation of the last couple of decades (Akiyoshi et al., 2009), which include the large decline in the lower stratospheric O_3 in the high latitude of both hemispheres. This modeled stratospheric O_3 might be smaller than the stratospheric O_3 data employed in the other models. We added a sentence mentioning the above in the manuscript.

P9089 L20. Why should the minimum in the stratospheric contribution change over interior continental sites?

→ The autumn to early winter minimum in the stratospheric contribution can be seen only in the interior of the Eurasian continent. This is about two months later than that

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in the other regions in the same latitude band. We checked the seasonal variations of chemical loss rate and potential vorticity, which have large impact on the seasonal behavior of stratospheric contribution to the surface O3, but we could not find any significant differences of them between the interior of the Eurasian continent and other regions in the northern mid latitude. Therefore, it is quite difficult to answer this comment at the present moment.

Section 3.2 This section might be better titled “Seasonal cycles of East Asian surface O3”.

→ We adopted your suggestion for the title of newly defined subsection 3.1.2.

P9091 L11-15. This seems like it belongs in Section 2.

→ We removed the sentence from here to Section 2.1.

Section 3.3 Are these results expected to depend on the model resolution (e.g., see Lin et al., Atmos. Chem. Phys., 10, 4221-4239, 2010)?

→ We haven’t done the simulation with different resolution, so it is difficult to say conclusive comments on this. However, the difference in both of the export of East Asian pollutants and the influence of European emission on East Asia between global model and regional fine-scale models that Lin et al. (2010) shown can occur if we employ the different resolutions, because the causes of differences which Lin et al. (2010) pointed out (including inadequate vertical venting and excessive dilution of pollutants in coarse resolution) are common to every model.

Section 3.3 How do the seasonal cycles of stratospheric contribution compare to those shown in earlier publications for North America or Europe?

→ We have been already made some comments about the difference in seasonal cycle of stratospheric contribution between the current study and previous literatures (Roelofs et al., 1997; von Kuhlmann et al., 2003) at some maritime rural observation sites in Europe and North America in Section 3.1. The comparison shows that

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our model exhibits the similar seasonal cycle to the previous studies but calculates somewhat smaller stratospheric contribution. We described the possible cause of this difference in the revised manuscript.

P9093 L18-20. Is this true even in October? This sentence should clarify that it is referring to the "cold" season.

→ We changed the sentence to make it clear.

P9095 L14-19 seems to repeat points above – combine, and include the numbers directly from figure 5.

→ We agree. We combined the two parts, and included the numbers directly from Figure 5.

Somewhere "domestic" should be defined. → We defined "domestic pollution" in the third paragraph of Section 3.2.

P9097 L9-20. Are these variations those associated only with meteorology or do model emissions also change each year?

→ We changed the emission inter-annually (please refer to our response to your comment above for details), so it could play a role on the inter-annual variation of S-R relationship. 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.

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- Kurokawa, J., Ohara, T., Uno, I., Hayasaki, M., and Tanimoto, H.: Influence of meteorological variability on interannual variations of springtime boundary layer ozone over Japan during 1981–2005, Atmos. Chem. Phys., 9, 6287–6304, 2009

Section 3.4 Is the model sampled at the site locations for this comparison?

→ No. As explained in the second paragraph of Section 3.4, observed data obtained at the sites located inside of a model grid are averaged to calculate "gridded" observation. The frequency distribution of this "gridded" observation and model are compared in Figure 6.

P9098 L16-19. The model does not show this feature so L25-26 should be rephrased.

→ We rephrased these sentences.

P9099 L2-3. Why does the nighttime matter since daytime values are used here? Can the urban observations be excluded for model evaluation?

→ 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 can not be simulated in the model. We added the above sentence in the revised manuscript.

→ Urban observations have already been removed for the model evaluation.

P9099 L6-9. Might this be shown more directly by summing the hours above 60 ppb and comparing the percentage above this value in the observations and in the model?

→ We don't think the percentages of hours over 60 ppbv are good indicator of the ability of our model to properly simulate the intermediate range of 1-hourly surface O3.

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Section 3.5 Is it meaningful to look at the model values above 90 ppb when it doesn't simulate the frequency of these events well?

→ 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 meaningful.

P9100 L15. Why is the stratospheric contribution so much higher at CHN-NCP (P9100 L15) than in JPN/KOR? Is the elevation higher?

→ Yes, the average elevation in CHN-NCP is higher than that in JPN/KOR. Besides, CHN-NCP locates more northward than JPN/KOR. As depicted in Figure 5, stratospheric contribution is larger in northern region.

P9103 L14-17. This statement is problematic since Figure 6 shows that the model fails to simulate the increase in high O3 in summer.

→ We changed the sentence not to mislead that the model can simulate the extreme O3 in summer.

P9104 L1. It would be useful to provide a quantitative estimate for this baseline o3.

→ Since the baseline O3 varies by season and source region, presenting the baseline O3 should be lengthy. So, we think it is not appropriate for "Summary" section. The estimate of baseline O3 has already been shown in Section 3.5 only for spring though.

Table 1 can move into supporting information since the key points are evident from Figure 2. Alternatively, the statistics could be added to each individual panel in Figure 2 if the Figure is substantially enlarged.

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→ Figure 2 thoroughly shows good performance of our model to represent the observed features of surface O3, as you pointed out. But, we think it is also important to confirm that good performance with suitable statistics. Since Table 1 is not so large table and insertion of these statistics into each individual panel would make Figure 2 clutter, we want to keep Table 1 as it stands.

Table 2. Are the source and receptor region labels swapped?

→ No.

Table2. This seems repetitive with Figure 4. I suggest moving the contribution values to supplemental information, and keeping only the coefficient of variation which is the key point discussed in the text from this table. IDC+ needs to be explained.

→ We agree to keep only the coefficients of variation in Table 2. The definition of IDC+ has already been described in the manuscript, but we added it also in the caption.

Figure 4 can be condensed to focus on the most important information. I suggest just showing the top panels and decreasing the total number of panels by combining regions as done in Figure 5. In the top panels, do I interpret correctly that the contributions are only the shaded regions? That means we have to read the difference of the top and bottom of the white area for the free troposphere contribution? This plotting method makes it a little complicated to easily see seasonal cycles; might it be clearer to just plot the amount from the different regions rather than shading this way? It's hard to extract information from the barplots (bottom panels figure 4). While this might be improved by showing only the top 3-4 source regions, it's probably best to move these to supplemental information and enlarge them so that the interested reader can extract quantitative information.

→ We agree concentrating on the most important points. We totally changed the Figure 4 according to your comments, which includes the change in the plotting method of upper panels and the removal of illegible bar plots from bottom panels. Moreover,

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new bottom panels which show the relative contributions of transported O3 for each receptor area were introduced. What the new bottom panel show is the same as what the red lines in original upper panels of Figure 4 show. As to combining receptor areas as done in Figure 5, because it is necessary to show the similarity between JPN-W and JPN-E (and also KOR-N and KOR-S) prior to introducing Figure 5 in which those receptor areas are combined, we want to keep showing ten receptor areas. We accordingly changed the manuscript describing Figure 4. The original Figure 4 is moved to supplemental information (as Figure S1).

The information is much better presented in Figure 5; the total O3 could be given at the top of each bar in Figure 5 to allow the reader to estimate the absolute contributions.

→ We added the value of total O3 in Figure 5.

Technical comments Abstract L24: “expect” should be “except”

→ We revised the manuscript as you pointed out.

P9079 L17 reinforcement -> enforcement ?

→ No. Here we mean that the regulation against emissions of O3 precursors in Japan has been intensified in last several decades. Therefore, reinforcement is suitable here.

P9084 L21. Clarity -> brevity?

→ We revised the manuscript as you pointed out.

P9084L24: refer to the map of the regions.

→ We added a reference to Figure 1 in the manuscript as you pointed out.

P9089 L2 what is the “observed lower portion of daily mean. . .”?

→ For the comparison at Hohenpeissenberg, the range of daily means for observation (red boxes) is much wider than that for the model (black error bars), especially the minimum of the range for observations is much smaller than that for the model almost

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all months. This means the model can not represent the lower range of daily means. We change the sentence in order to clarify the meaning.

P9089 L24. “condensed O3” is awkward.

→ We changed “condensed” to “dense”.

P9094 L6 is awkward; the foreign source regions are not in East Asia.

→ The “foreign” source regions in East Asia for e.g. Japan means the regions located in East Asia other than Japan; here Japan source region is referred as “domestic” source region. But, this sentence is surely unclear, so we removed the term “foreign” from the sentence.

P9095 L1-5 seems to repeat earlier points – cut or condense.

→ Yes, we agree what you pointed out. We condensed the sentence.

P9096 L15-18 where and when?

→ The contribution of ASeas to central Japan and Korean Peninsular in both spring and summer is large. We changed the sentence to make it clear.

Section 3.4 The region labels should be consistent in the text and figures.

→ We made an error in the name of observation system in the text. We corrected it.

Section 4. Define “S-R relationship”

→ We inserted the definition of S-R relationship in the first sentence of Section 4 and also changed the following sentence accordingly.

Figure 1. red letters are hard to see.

→ We changed the color of letters on Figure 1

Figure 2 is too small. It’d be better to organize the panels in the order in which they’re discussed in the text (or at least explain the current logic). The red vs. green is not

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discussed separately in the text so I don't see the need for the distinction in record length. How are the model calculations interpolated to the longitude, latitude, and altitude of each site?

→ We agreed this comment, so we separated the Figure 2 into two figures and enlarged each plot. We also changed the way to plot the modeled stratosphere, PBL, and FT contributions in the same way as Figure 4 is modified (please refer to the change in Figure 4). Besides, we dropped the distinction of plot color in record length from the figure.

Figure 3. The season labels are hard to see on the plots.

→ We changed the season labels in Figure 3

Figure 4. What is the black line? The dashed red line needs more spaces as its hard to distinguish from the solid line. The caption should explain the map of the regions.

→ We totally changed Figure 4. Please refer to our reply to your comment on Figure 4

Figure 6 caption. Define what is meant by "ridge line". Are the frequency distributions constructed by using 10 ppb bins and then the average is taken across the 6 years within each bin?

→ Ridge line means the line that connects the mode of frequency distribution (FD) of each month. Here, the value of the mode is defined as the center of the most frequent 10-ppbv bin (e.g., 25 ppbv for 20-30 ppbv bin). The monthly FDs are firstly calculated for each year and then they are averaged to get the 6-year averaged monthly FD, which is used to draw Figure 6. We changed the caption of Figure 6.

Figure 7 is too small.

→ We vertically enlarged the figure and changed the font to larger one.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 9077, 2010.

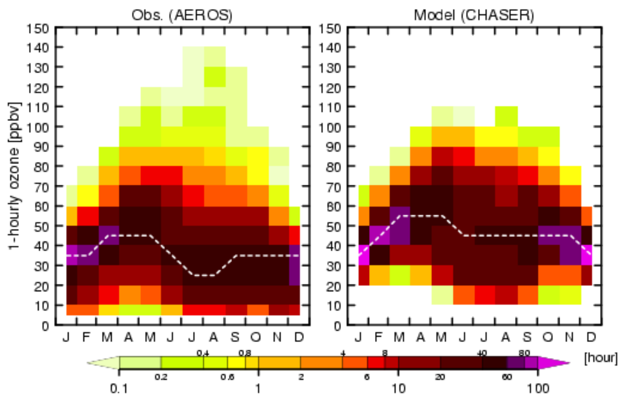

Fig. 1. Revised Figure 6

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