Response to Referee Comment 2 by Anonymous Referee #2

Itahashi et al. (2019) investigated the impacts of emissions from East Asia and US on surface O3 over Northern Hemisphere using HDDM in H-CMAQ. They found comparable impacts by the emissions from East Asia and US on surface ozone over western US with US domestic emissions having larger impacts on surface ozone over eastern US while emissions from East Asia have much larger impacts on free troposphere through trans-pacific transport. But they also found the impacts of recent emission changes in East Asia on US O3 levels were small. The manuscript is in general well written but there are several concerns about the methodology used in the manuscript.

Reply:

We thank the reviewer for providing helpful and constructive comments. We have revised our manuscript according to the reviewer's comments and suggestions. We believe that these revisions address all points raised by the reviewer. Our point-by-point responses are provided below, and revisions are indicated in blue in the revised manuscript.

ZOC of emissions

We have divided this comment into three smaller parts and replied to each portion individually.

The estimation on the emission impacts is based on only one H-CMAQ simulation, is that correct? If so, the ZOC estimates are not really accurate as if we reduce all the targeted emissions, meteo/dynamics will also change, which could have feedbacks on the estimations of the emission impacts. The atmospheric conditions will be different under normal and ZOC scenarios. Any explanations on this?

Reply:

Yes, the estimation is based on one-time simulation of HDDM embedded in CMAQ model. The emission variations can cause the changes in meteorology and dynamics; however, the version of the CMAQ modeling system used in this study is based on the so called "off-line" approach which does not consider the feedback between meteorological variables and chemistry. Therefore, in this study, our estimation just focuses on the emission impacts on the concentration of air pollutants.

Also, ZOC is not a really realistic scenario and the impacts of ZOC are probably overestimated in this work. So your conclusion about the impacts of the emissions from East Asia may not be that robust. As in your last part of the work, the impacts of recent changes in the emissions from East Asia were found to be small to insignificant. Unless all the emissions from East Asia were removed, significant impacts would exist over western US at surface and entire US in free troposphere? In other

words, To what degree of the changes in the emissions of East Asia should be achieved to show noticeable impacts on US?

Reply:

In this study, HDDM embedded in CMAQ is used to obtain sensitivity coefficients regarding NO_x and NMVOC emissions as shown in Fig. 1. As an example to illustrate the emission impacts, we showed the result of ZOC based on Eq. (5) in Fig. 3. Because the impacts of emissions can be estimated in detail based on Eq. (4), the conclusion can provide the robustness of emission impacts.

In the latter parts of the analysis presented in this work, the impacts of recent emission changes are also estimated based on Eq. (4), and these results showed that the impact of emission changes from 2010 to 2015 in East Asia were insignificant on simulated O_3 levels over U.S.A. In contrast, as illustrated in Figure 6, a complete removal of emissions from East Asia does result in significant impacts on surface and free-troposphere O_3 over U.S.A. can be found if emissions from East Asia is removed. As we already state in Section 4.4, Chinese emissions after 2010 showed complex variations. To assess the possible impacts of these changes on our sensitivity analysis results as well as likely changes in trans-Pacific transported pollution, we summarize the impacts of these emission changes via the illustrations in Figs. 9 and 10.

For ZOC scenario, does it simply remove all the NOx and NMVOCs emissions? How to distinguish between anthropogenic and natural emissions for NMVOCs? Are the impacts of ZOC the total impacts from the removal of both NOx and NMVOCs? Can we have individual impacts from the removal of NOx only and removal of NMVOCs only? Or maybe we can infer from the sensitivity to these emission? This information could provide more guidance on future emission mitigations.

Reply:

These estimations are based on HDDM embedded in CMAQ. Because of the unified input dataset for H-CMAQ, we first estimated the sensitivities for all emission sources (i.e. anthropogenic and biogenic), and then we further estimated sensitivities to isoprene (a proxy for biogenic emissions) as shown in Fig. S1. HDDM allows both the separate and combined estimation of the impacts of NOx and NMVOC emissions as we have shown in Fig. 1 (a and b for NO_x emissions, c and d for VOCs emissions, and e for the combined effects of NO_x and VOCs).

Specific comments:

Page 1, Abstract, line 24, “with a magnitude of about 3 ppbv impacts on a monthly mean …” 3 ppbv O3?

Reply:

Yes, we have added 'O_3' to explicitly mention it.

Page 4, line 4, "O3 mixing rations and an those of inert tracer …", grammatical error

Reply:

We have corrected this error.

Page 5, line 5-6, equation (3), should Si,j(2) have the unit of square of the concentration?

Reply:

No, as we have explicitly stated after Eq. (3), the unit of $S_{i,j}^{(2)}$ is same as the concentration, which is ppbv in this case.

Page 6, and Figure 1, any physical meaning on second-order sensitivity? line 9-10, can you explain more on "concave response"?

Reply:

The second-order sensitivity reflects the nonlinear response. The large value of second-order sensitivity corresponds to the strong nonlinearity, and if the response is linear, the value of second-order sensitivity is negligible. Under the typical O_3 concave response to NO_x emissions, the first-order sensitivity is positive and the second-order sensitivity is negative; and the absolute value of this negative second-order sensitivity represents the magnitude of nonlinearity.

Page 6, line 27-28, what do you mean by "Svocs and Snox"? it is not equal to Svocs +Snox, right? Maybe you mean Svocs-nox ? Please clarify.

Reply:

These equations include two formulae. To avoid the misread, we have revised these equations to be separated by ','.

Page 7, line 17-18, regions in VOC-sensitive regime are not clearly shown in Fig 2. You may want to change the color scale to improve the quality.

Reply:

The VOC-sensitive regime is not clear as indicated by the weak responses shown in Fig. 1. This is not due to the color scale. We have revised this sentence as follows:

"In some areas over the U.S.A. that are characterized by a weak VOC-sensitive regime in Fig. 2…"

Page 21, Figure 3, are these impacts based on zero-out of both NOx and NMVOCs at the same time? See my general comments above.

Reply:

As we have replied in general comments, these impacts are based on Eq. (5) using the sensitivity coefficients obtained by HDDM. The impacts shown in Figure 3 are estimated by considering both NO_x and VOCs emission changes. HDDM estimates both the individual and combined ozone sensitivities towards these pollutants.