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Interactive comment on "Comprehensive analyses of source sensitivities to and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques" *by* Satoru Chatani et al.

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Dear Referee #1:

[Referee #1] This manuscript presents an in depth discussion of the differences between sensitivity analysis and source apportionment methods in terms of non-linear effects concerning transport and local emissions. The presented data is new and relevant to future ozone and PM2.5 control in Japan. However, I cannot recommend its publication in its current form. I suggest the following revisions before reconsideration.

[Reply] Thank you for valuable comments on our manuscript. I will revise the

manuscript based on your comments.

[Referee #1] Major Comments: 1) An English mistake that makes the manuscript very difficult to read must be corrected. The authors refer to "sensitivity of emissions to the pollutant (ozone or PM)". This mistake starts in the title and continues throughout the main text and the supplemental material. It should be corrected as "sensitivity of a pollutant to emissions" meaning that the pollutant is the dependent variable which respond to changes in emissions as independent variables. Similarly, there is another mistake in the use of the term "source apportionment". The existing literature refers to "source apportionment of a pollutant". The title is correct in this sense but in the text the authors refer to "source apportionment to the pollutant". This should be corrected as "source apportionment of a pollutant (ozone or PM) to the emissions (e.g., on-road vehicle emissions or NOx emissions from on-road vehicles).

[Reply] I am so sorry for this English mistake for the important words of this study. Another referee raised the same issue. I fully agree that "sensitivity of pollutant concentrations to emissions" and "apportionment of pollutant concentrations to emissions" are correct expressions.

The title will be changed as follows.

"Comprehensive analyses of source sensitivities and apportionments of PM2.5 and ozone over Japan via multiple numerical techniques"

In addition, I will check the main text, tables, figures, and supplemental material, to correct all the relevant parts. A grammar check will be also done again. Please check in the revised manuscript.

[Referee #1] 2) It is not clear how HDDM-100 differs from HDDM-20. They both seem to be using the same sensitivity coefficients, i.e., slopes and curvatures at unperturbed level of emissions. If these coefficients were being calculated in different simulations with different levels of emissions then it might have been interesting to compare them.

But the description in lines 330-335 suggests that they are the same thing. Similarly, it might be interesting to compare the results of BFM with 100% reduction for some of the most nonlinear pollutant-emission relations. At that level of reduction BFM results might be more similar to source apportionment.

[Reply] It is true that HDDM-100 and HDDM-20 are calculated using the same sensitivity coefficients, but their values could be different. The explanations in the lines 330-335 will be revised as follows. I hope they make discussions clearer.

"The HDDM-20 corresponds to the value calculated by applying $\Delta\varepsilon$=-0.2 and multiplication by 5. If a sensitivity is represented by a second-order polynomial function, HDDM-20 is equivalent to the value obtained by BFM. However, influences of the second-order term for a perturbation beyond 20% are not reflected in HDDM-20 because the value at a 20% perturbation is just linearly extrapolated. They are reflected in the HDDM-100, which corresponds to the value calculated by applying $\Delta\varepsilon$=-1.0. Differences between BFM and HDDM-20 correspond to the deviations of sensitivities from second-order functions, and differences between HDDM-20 and HDDM-100 correspond to the influences of the second-order term for a perturbation beyond 20%"

The sentence in the lines 411-412 will be revised as follows to make consistent with the explanations above.

"Differences should be recognized as difficulties in representing sensitivities only with first- and second-order sensitivity coefficients derived by HDDM."

I fully agree that it might be interesting to compare the results of BFM with 100% reduction for some of the most nonlinear pollutant-emission relations. Therefore, additional simulations were conducted with 100% reduction for s04 and s08 for the discussions in the Section 3.6.2, and for s01-NOX and s01-VOC for the discussions in the Section 3.6.3. Interesting results were obtained. I confirmed that sensitivities become closer to apportionments in some conditions.

The following descriptions will be added to the end of the Section 3.6.2 based on the results for s04 and s08 with 100% reduction.

"In addition to BFM with 20% perturbation (denoted as BFM-20), additional simulations were conducted to derive sensitivities by BFM with 100% perturbation (denoted as BFM-100) for s04, which emits NOX and no NH3, and s08, which emit NH3 and no NOX. Figure S7 in the Supplementary Material shows the sensitivities derived by BFM-20, BFM-100, HDDM-20, and HDDM-100, and apportionments derived by ISAM of the daily NO3- and NH4+ concentrations to s04 and s08 for the two target weeks in winter in ST. The sensitivities derived by BFM-100 are higher than those derived by BFM-20 due to nonlinear responses. Similar features are evident in the sensitivities derived by HDDM-100 and HDDM-20, implying that HDDM is capable to represent directions of nonlinear responses beyond 20% perturbation. It is notable that the sensitivities derived by BFM with a larger perturbation become closer to the apportionments for NO3- to s04, and NH4+ to s08. However, there are still deviations among them caused by indirect influences of factors including other sectors, complex photochemical reactions, and gas-aerosol partitioning. Moreover, the apportionments of NO3- to s08 and of NH4+ to s04 never appear while NO3- and NH4+ concentrations are nonlinearly sensitive to s08 and s04, respectively."

The third paragraph of the Section 3.6.3 will be divided into two and the latter one will be revised as follows based on the results for s01-VOC and s01-NOX with 100% reduction. Figure 8 (attached as Fig. 1 in this reply) will be replaced accordingly.

"We note that the sensitivities to VOC emissions derived by BFM-20 and BFM-100 are almost identical. That means ozone formation from VOCs is linear to emissions. The sensitivities of NOX emissions derived by BFM-20 and BFM-100 are also almost identical when they are negative. That means titration of ozone by NOX is also linear to emissions. In contrast, the sensitivities to NOX emissions derived by BFM-100 are higher than those derived by BFM-20 when they are positive. That means ozone formation from NOX is nonlinear to emissions. Cohan et al. (2005) also reported that the sensitivities of ozone concentrations are lower when perturbations of precursor emissions are smaller because other remaining precursors are more likely to contribute to ozone formation instead. This may also be the reason why the sums of the sensitivities to all the sources are lower than the simulated ozone concentrations in spring and summer, as shown in Figs. 2, 3, and 5. While the sensitivities derived by BFM-100 become closer to the apportionments, the apportionments are still higher than the sensitivities as discussed for NO3- and NH4+ in the Section 3.6.2. That implies effects on concentrations of ozone, NO3-, and NH4+ may be less than those inferred by BFM-100 and ISAM when reductions of emissions of NOX and NH3 are small."

[Referee #1] 3) The issue of how model performance might affect sensitivities and source apportionments in this study is an important one. An elaborate discussion would be very helpful instead of just a generic statement that it is important. For example, given the poor performance in nitrate, which source apportionments and sensitivities are more uncertain? How does the poor performance in nitrate affect the nonlinear sensitivities to NOx and NH3 emissions?

[Reply] While it is a difficult question to answer because reasons of poor model performance have not been clarified, I will add an analysis to answer to this important question. Please see the reply to the minor comment below.

[Referee #1] 4) The conclusions are somewhat generic; they could be written in a way that praise the findings of this study. See the minor comments below for places in the abstract and conclusions where more specific information might give this study the credit that it deserves.

[Reply] I will revise the conclusions to praise the findings of this study more clearly and to add findings of additional simulations. Please check them in the revised manuscript.

[Referee #1] Minor Comments: 1) The last statement of the abstract (lines 24-26) is very generic; it should be replaced with a statement of specifically what was found in this study.

[Reply] They will be replaced by the following sentences including relationships between sensitivities and apportionments described in the reply above.

"While the sensitivities become closer to the apportionments when perturbations in emissions are larger in highly nonlinear relationships including those between NH3 emissions and NH4+ concentrations, NOX emissions and NO3- concentrations, and NOX emissions and ozone concentrations, the sensitivities did not reach the apportionments due to various indirect influences including other sectors, complex photochemical reactions and gas-aerosol partitioning. It is essential to consider nonlinear influences to derive strategies for effectively supressing concentrations of secondary pollutants."

[Referee #1] 2) We don't find out about the horizontal grid resolutions until Section 3.5. This information could be given in Section 2.1.

[Reply] A description on the horizontal grid resolutions will be inserted to the line 90 as follows.

"Horizontal resolutions of d01, d02, d03, and d04 are 45 x 45 km, 15 x 15 km, 5 x 5 km, and 5 x 5 km, respectively."

[Referee #1] 3) Did you report the HDDM convergence problems to the CMAQ modeling community? Others who experienced similar issues may be able to recommend solutions.

[Reply] While we did not report to the CMAQ modeling community (e.g. CMAS forum), one of members in the CMAQ developing team agreed that there are still convergence problems in HDDM embedded in CMAQ. The problem could be avoided by altering some model configurations, but that could not be done because consistencies among BFM, HDDM, and ISAM are important in this study.

[Referee #1] 4) Line 177: Actually, I believe your model performance meets some of the goals in Emery et al (2017). You may want to distinguish between criteria and goals.

[Reply] Thank you for your suggestion. I confirmed that our model performance meets the goals for limited species and in limited regions. However, I would avoid making discussions complicated by distinguishing goals and criteria.

[Referee #1] 5) Line 195: Add “following sulfate” after “OC is the second major component of PM2.5”

[Reply] It will be added as suggested as follows to the line 195.

“As OC is the second major component of PM2.5 following SO42-”

[Referee #1] 6) Lines 196-197: “Less overestimates dots: : :” Consider deleting this sentence.

[Reply] This sentence will be deleted as suggested.

[Referee #1] 7) Lines 221-224: Please explain how the “chlorine loss” works in more detail and consider moving this discussion to the previous paragraph since the negative sensitivities to sea salt are first seen in Figure 2.

[Reply] The following sentence will be added to the previous paragraph to just show the fact of the negative sensitivities to sea salts.

“The sensitivity of PM2.5 to s12 (sea salt) is negative.”

Explanations in the lines 221-224 will be revised as follows. I hope they make discussions clearer.

“The sensitivities of NO3- and NH4+ to s12 (sea salt) are negative. Cl– originated from sea salts and mostly involved in coarse particles tend to be replaced by NO3– due to so-called chlorine loss caused by gas-aerosol partitioning (Pio and Lopes, 1998; Chen et al., 2016). Therefore, if sea salts are present, more HNO3 gases are partitioned to coarse particles. That provides capacities for NO3- and associated NH4+ involved in PM2.5 to evaporate to the gas phase, resulting in negative sensitivities of PM2.5 including NO3- and NH4+ to sea salts.”

[Referee #1] 8) Lines 226-249: This discussion is difficult to follow. Perhaps you should use past tense for the previous studies and present tense for the current study. Also, state the two possible reasons upfront: 1) Japanese emissions are underestimated and 2) Foreign countries other than China are included (if I understand them correctly). I also recommend an explanation of the normalization mentioned in Table S3.

[Reply] I am sorry for these difficult sentences. I will correct all the tense so that past tense is used for the previous studies and present tense is used for the current study.

There are two reasons: (1) reduction of emissions in Japan, and (2) other factors than emissions in China. The paragraph in the lines 227-250 will be divided into three to make easier to follow. The latter two paragraph, originally in the lines 237 and 248, will start with the following sentences, respectively.

“One of possible reasons for these elevated contributions is reduction of emissions in Japan” “Another reason of the higher contributions of foreign sources owes to other factors than emissions in China.”

The explanation of the normalization will be added to the beginning of this paragraph as follows.

“Table S3 in Supplementary Material lists the ratios of the source sensitivities of the annual mean ozone and PM2.5 concentrations simulated in the regions, which were compared with previous studies. While sums of the ratios of the sensitivities to all the source groups are not 100% due to nonlinearities, they were often normalized to 100% in previous studies. Therefore, the ratios normalized to make their sums equal to 100% are also shown in Table S3.”

[Referee #1] 9) Line 283: Do we know what the background concentration levels are?

[Reply] It may be inappropriate to mention background concentrations because they are unknown in this study. Therefore, this and the preceding sentences will be deleted.

[Referee #1] 10) Figure 4: What is the rationale of selecting s01 EC for normalization?

[Reply] There is no rationale. Anything can be used for normalization because Figure 4 just shows relative relationships among sensitivities. s01 EC was selected just because they are inert and emitted only in the bottom layer. Such an explanation will be inserted in the line 295 as follows.

"All the values shown in Figure 4 were normalized by the EC value for s01, which is inert and emitted only in the bottom layer."

[Referee #1] 11) Line 328: Is there a caveat of assuming that "OTHR" in ISAM is SOA.

[Reply] It would be ideal that ISAM can calculate apportionments of SOA. However, it is impossible for ISAM embedded in CMAQ version 5.0.2. Descriptions around the line 328 will be revised as follows.

"The simulated SOA concentrations were characterised as apportionments of "OTHR" in ISAM in this study because apportionments of SOA concentrations were not calculated by ISAM embedded in CMAQ version 5.0.2"

[Referee #1] 12) Line 381: Replace "an oxidative capacity" with "the oxidative capacity"

[Reply] It will be replaced as suggested.

[Referee #1] 13) Lines 454-455: Replace "can provide the" with "provides"

[Reply] It will be replaced as suggested.

[Referee #1] 14) Line 462: "similar" or "more"?

[Reply] The sentence in the lines 461-464 will be revised as follows.

"While PM2.5 concentrations were lower than those simulated by previous studies for past years due to emission reductions, the relative contributions of transport from outside Japan to the total sensitivities were even larger, suggesting that emissions in Japan have been similarly reduced to surrounding countries including China."

[Referee #1] 15) Lines 479-480: Give an example for each of the direct and indirect

influences that could not be distinguished only by the sensitivities.

[Reply] The following example will be inserted in the line 481. This paragraph will be merged to the precedent one because both contain similar descriptions.

"For example, the sensitivities of SO42- and NO3- to the transport from outside Japan encompassed at least two undistinguished influencing factors, including the direct transport of SO42- and NO3-, which were evaluated by their corresponding apportionments, and oxidation of SO2 and NOX emitted from domestic sources by OH originating in ozone transported from outside."

[Referee #1] 16) Lines: 482-483: Give examples of how model performance may skew specific source sensitivities and apportionments in this study.

[Reply] Figure S10 will be added to show sensitivities of PM2.5 uniformly scaled by the ratios of observed and simulated concentrations of PM2.5 components. Discussions on this figure will be inserted in the line 484 as follows.

"Figure s10 in the Supplementary Material shows source sensitivities of the annual mean PM2.5 concentrations derived by BFM in the regions. The values shown in (b) were uniformly scaled by the ratios of observed and simulated concentrations of PM2.5 components shown in Table S2. The scaled sensitivities of PM2.5 to the transport from outside Japan are higher by 1.0-2.2 μg/m3 (15-40%) due to their high contributions to underestimated POA and SOA. The scaled sensitivities of PM2.5 to other sources are different by 0-0.5 μg/m3. This is the case that deviations between observed and simulated PM2.5 concentrations can be proportionally explained by the source sensitivities. Uncertainties could be higher if specific sources cause poor model performance. In particular, this study revealed NH4+ and NO3- concentrations are nonlinearly sensitive to NH3 and NOX emissions. Uncertainties in NH3 and NOX emission sources could largely influence source sensitivities as well as model performance of NH4+ and NO3- concentrations. More studies are necessary to obtain more confidence in source sensitivities and apportionments as well as model performance."

[Referee #1] 17) Lines 489-490: Consider deleting the sentence that begins with “In reality”

[Reply] This sentence will be replaced by the following one.

“However, model configurations and inputs may not be necessarily consistent.”

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2020-236/acp-2020-236-AC1-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2020-236, 2020.

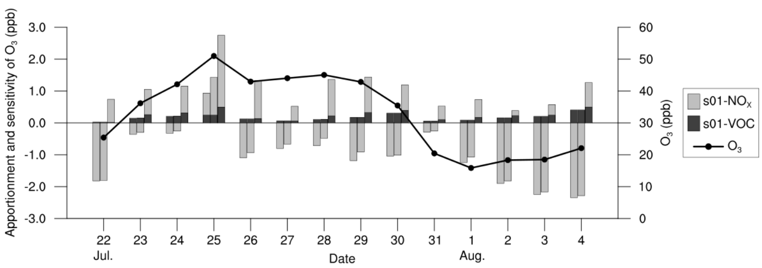

Fig. 1.