

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.

Anonymous Referee #1

Received and published: 20 May 2020

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. 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

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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). 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. 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? 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. 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. 2) We don't find out about the horizontal grid resolutions until Section 3.5. This information could be given in Section 2.1. 3) Did you report the HDDM convergence problems to the CMAQ modeling community? Others who experienced similar issues may be able to recommend solutions. 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. 5) Line 195: Add "following sulfate" after "OC is the second major component of PM2.5" 6) Lines 196-197: "Less overestimates dots..." Consider deleting this sentence. 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. 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. 9) Line 283: Do we know what the background concentration levels are? 10) Figure 4: What is the rationale of selecting s01 EC for normalization? 11) Line 328: Is there a caveat of assuming that "OTHR" in ISAM is SOA. 12) Line 381: Replace "an oxidative capacity" with "the oxidative capacity" 13) Lines 454-455: Replace "can provide the" with "provides" 14) Line 462: "similar" or "more"? 15) Lines 479-480: Give an example for each of the direct and indirect influences that could not be distinguished only by the sensitivities. 16) Lines: 482-483: Give examples of how model performance may skew specific source sensitivities and apportionments in this study. 17) Lines 489-490: Consider deleting the sentence that begins with "In reality"

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