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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.

Satoru Chatani et al.

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Dear Referee #3:

[Referee #3] General comment: technically sound but conclusions unclear and disappointing, possibly overstated The authors have performed air quality simulations using the CMAQ model over various nested domains including Japan or parts of that country. They show in a convincing way that their simulations are realistic and have resonable (even good) performance. They study various methods to study the impacts of different types of sources in terms of concentrations of PM2.5 and ozone, including "Brute force method" (i.e. sensitivity simulations), ISAM and HDDM. As far as I can tell, all

the methods implemented by the authors are technically sound and, at least in terms of modelled concentrations, they are easily on par with the State of the art. However, even though this study has obviously involved a big amount of work, its point is not clear to me. In the abstract, the authors state that "This study demonstrated that a combination of sensitivities and apportionments derived by the BFM, HDDM, and ISAM can provide critical information to identify key emission sources and processes in the atmosphere, which are vital for the development of effective strategies for improved air quality". A similar statement appears in the conclusion: "This study demonstrated that a combination of sensitivities and apportionments derived by the BFM, HDDM, and ISAM can provide critical information to identify key emission sources and processes in the atmosphere, which are vital for the development of effective strategies for improved air quality, using consistent model configurations and inputs.". However, in-between I (and only "I" because that feeling is very possibly due to the fact that I am not so familiar with the issues the authors discuss) felt overwhelmed by a mass of plots and figures quite often lacking physico-chemical interpretation. In summary, I have failed to understand which of the actual information unveiled by the authors was "critical" or even "vital" for policy design. On the contrary, I have the feeling that the methods they deploy are advanced but the actual results that they show are often disappointing when compared to the weaponry that they have used. For example, in the conclusion, the authors state that "Domestic sources had certain sensitivities to PM 2.5 , but significantly smaller or even negative sensitivities to ozone due to titration and nonlinear responses against precursor emissions.", which is hardly a surprise, it is discussed in all the good atmospheric composition textbooks that ozone concentrations are having a twofold sensitivity to emissions depending on the chemical regime. Here the authors' methodology seems to lead the reader to conclusions that are already very well-known. I think the authors have realized good simulations of air quality over their areas of interest, convincingly shown that point, they have deployed methods they claim to be extremely useful in terms of understanding the rôle of different source areas and activity sectors in air pollution in Japan, but in my opinion they fail to make that second point, leading

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to disappointing conclusions.

[Reply] Thank you so much for critical comments. I also think that the works like this study have not be completed in any other previous studies. The results provided various interesting information. Indeed, nonlinear relationships between ambient concentrations of secondary pollutants including ozone and PM2.5 and precursor emissions are well-known and written in textbooks. However, I believe that it is still worthwhile to investigate them further. As mentioned in the introduction, we are facing problems involving ozone and PM2.5 in Japan in spite of stringent emission controls. That means our understandings on nonlinear relationships between concentrations and precursor emissions are not enough. Currently, we do not have any clear understandings for effectively suppressing concentrations of ozone and PM2.5. We hope to contribute to solving the problems by providing useful scientific and quantitative information through this study. In addition, nonlinear relationships are not phenomena limited to Japan. We suppose our findings would be valuable everywhere on the globe.

The paragraph will be added at the end of Section 1 as follows to explain our idea.

"There are well-known nonlinear relationships between ambient concentrations of secondary pollutants including ozone and secondary components involved in PM2.5 (Seinfeld and Pandis, 1998). They are likely to cause deviations between source sensitivities and apportionments due to complex photochemical reactions and gas-aerosol partitioning. Nevertheless, it is important to investigate magnitudes of deviations and major causes of nonlinear relationships for considering effective strategies to suppress concentrations of secondary pollutants. Processes causing nonlinear relationships are universal phenomena and not limited to Japan. Findings of this study would contribute not only to solving remaining issues involving ozone and PM2.5 in Japan, but also to add more understandings on possible influences of nonlinear relationships everywhere on the globe."

I will try to explain the importance of this study throughout the manuscript. In addi-

tion, results and discussions of additional simulations, which were conducted based on the comments of another reviewer, will be added. Please check them in the revised manuscript. I hope these revisions are interesting for you and readers.

I will also revise the manuscript based on your comments below.

[Referee #3] Title: I have a hard time understanding the title, "Comprehensive analyses of source sensitivities to and apportionments of PM 2.5 and ozone over Japan via multiple numerical techniques". Even though it might be due to my partial knowledge of the jargon in this particular field, I have the feeling that, in the title and the rest of the text (e.g. l. 55, l. 74 and following, etc.). It seems that in the author's vocabulary they adress the sensitivity of the NOx emissions to ozone concentrations (this is just an example) while the ordinary way of thinking is more to assess the sensitivity of ozone concentrations to NOx emissions.

[Reply] I am so sorry for this English mistake for the important words of this study. Another referee raised the same issue. "Sensitivity of pollutant concentrations to emissions" and "apportionment of pollutant concentrations to emissions" should be 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 #3] Major comments: l. 461-464: "While PM 2.5 concentrations and their absolute sensitivities of all the sources were lower than those calculated 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

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in Japan have been reduced similar to surrounding countries, including China." I think the sensitivities and apportionment calculated by the authors do not depend on the actual emissions by Japan and China but on the emission hypotheses and inventories that have been chosen by the authors. I do not think the authors can draw any conclusion from their study regarding the emission reduction paths followed by Japan or China. I think the logical path leading to this result is circular: the authors make certain choices regarding emissions in Japan and China, they observe that the results they obtain are consistent with the hypothesis they made, but in my opinion this is no proof that their initial hypothesis is correct.

[Reply] I admit that this is very important issue. However, emissions compiled in the emission inventory have been estimated based on various information including changes in energy consumption, emission factors, and implementation of emission controls. Every simulation study must rely on one of emission inventories as a first assumption. Agreement of observed and simulated concentrations could be considered as one of proofs for accuracy of the emission inventory. However, it is indeed impossible to conclude only from this fact that the emission inventory is definitely accurate. Circular exercises including validation of simulations and improvement of emission inventories are necessary. Regarding this study, while simulations implied that emissions in Japan have been reduced as estimated in emission inventories, they also implied reductions may be too much and caused underestimation of PM2.5 concentrations. Not only the former but also the latter aspects are discussed in the lines 237-248. Discussions for the latter aspect will be revised to make clearer as follows.

"However, we can also state that the underestimations of the PM2.5 concentrations are larger in eastern than western Japan as described in section 3.1. Influences of domestic sources should be accumulated more in eastern than western Japan because a prevalent air flow over Japan is westerly. Therefore, worse model performance in eastern Japan imply underestimation of domestic emissions. Reductions of domestic emissions from fiscal years 2005 to 2015 may be excessively estimated."

[Referee #3] Minor comments, typos : p. 1, l. 16-17: "While domestic sources had certain source apportionments to ozone concentrations, transport from outside Japan dominated the source sensitivities." If possible, many sentences of this kind should be formulated in a more intuitive way, e.g., while domestic sources can contribute to a certain extent to simulated ozone concentrations, transport from outside Japan can be considered as the main overall driver of ozone concentrations in Japan (this is only my interpretation of course, just as an example on how the authors should make their conclusions more accessible to readers in the field but not specialized). At all places where this is possible, the authors should formulate their statements and partial conclusions in more physical terms.

[Reply] While it is a bit difficult to revise as suggested because sensitivities and apportionments should be clearly distinguished in this study, I will try to make descriptions in a more intuitive way. Please check them in the revised manuscript.

[Referee #3] p. 1, l. 22: "that that"

[Reply] I am sorry for this mistake. It will be corrected.

[Referee #3] l. 96: "Following" seems useless.

[Reply] It will be removed.

Reference Seinfeld, J. H., and Pandis, S. N.: Atmospheric chemistry and physics: From air pollution to climate change, John Wiley & Sons, Inc., 1998.

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