

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 #3

Received and published: 18 May 2020

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

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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 conclusionÂă: "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 to disappointing conclusions.

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. I. 55, I. 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.

Major comments:

I. 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 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 circularÂă: 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.

Minor comments, typos :

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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, eÂă.g., ÂńÂăwhile 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 JapanÂăÂż (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.

p. 1, l. 22: "that that"

I. 96: "Following" seems useless.

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