

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 #2:

[Referee #2] The paper by Chatani et al. is based on source sensitivities and apportionments of O₃ and PM_{2.5} over Japan by comparing 3 numerical techniques, 4 grids, 12 source groups. The paper is well organised and written, and the overall discussion is properly articulated. Figures are clear and they are all them necessary.

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

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[Referee #2] I have only two minor comments for the authors. Line 178-179. According to the simulations and statement, “The PM_{2.5} concentrations were underestimated in all regions. The statistics tended to be worse in eastern Japan as opposed to western Japan.” If the problem with the simulation has a clear geographical gradient (W-E), and after reading the discussion is mainly due to OC and nitrate, there is a probability of missing sources/atmospheric processes from local origin. Western Japanese sites are affected by long-range transport aerosols from other Asian countries, but Eastern sites are also affected by Japanese sources (considering a prevalent western to eastern air flow).

[Reply] I fully agree your comment. Actually, this issue has been already discussed in the lines 245-248, but I will revise it to make this issue clearer.

The following sentence will be inserted in the line 181.

“A possible reason will be discussed in the Section 3.2.”

The description in the lines 245-248 will be revised as follows.

“However, we can also state that the underestimations of the PM_{2.5} 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 #2 Line 381-384. The authors say “If ozone transported from outside Japan is not as reduced in future, efforts to reduce precursor emissions in Japan will not effectively contribute to the reduction in the concentrations of secondary PM_{2.5} components because OH that originated in ozone transported from outside Japan affects their formation”, which is an interesting statement. But it is hard to figure out which sources are releasing PM_{2.5} precursors (for example NO_x, SO_x or VOCs) but not releasing O₃ pre-

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cursors. All combustion sources are strong VOC emitters, and efforts are made/have been made to abate NO_x and SO_x. Of course that the efforts in reducing emissions in Japan will not counteract the arrival of steady emissions from outside, but the reduction in precursor emissions in Japan will lead to a lesser formation of secondary aerosols (although not in the same proportion as the applied reduction) and will contribute to the reduction of the continental O₃ background.

[Reply] I agree your comments. Source releasing PM_{2.5} precursors emit O₃ precursors. Discussions were too generalized. They should focus on SO₄²⁻ and NO₃⁻ as target species and SO₂ and NO_x as precursors. Corresponding expressions in this paragraph in the lines 377-385 will be revised as follows.

“Section 3.2 discussed higher relative contributions than previous studies and less contrasts between western and eastern Japan for the sensitivities of PM_{2.5} to s₁₁ obtained in this study. Oxidation of SO₂ and NO_x emitted from domestic sources by OH that originated in ozone transported from outside Japan is another factor that causes higher sensitivities of s₁₁. The entirety of Japan is equally affected by ozone transported from outside Japan, as shown in Fig. 2(a), due to its long lifetime in the atmosphere, resulting in less contrasts in the sensitivities of s₁₁ between western and eastern Japan, while the sensitivities of domestic emissions are small. Ozone governs the oxidative capacity of the atmosphere (Prinn, 2003). If ozone transported from outside Japan is not as reduced in future, efforts to reduce SO₂ and NO_x emissions in Japan will not effectively contribute to the reduction in the concentrations of SO₄²⁻ and NO₃⁻ because OH that originated in ozone transported from outside Japan affects their formation.”

However, the sensitivities of ozone to domestic emissions are small. In addition, influences of emissions in Japan to background ozone are marginal. I think influences of emissions of ozone precursors in Japan on oxidation of SO₂ and NO₂ are limited. The following sentence will be inserted in the line 382.

“while the sensitivities of ozone to domestic emissions are small.”

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