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 have revised 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 has been changed as follows.

"Comprehensive analyses of source sensitivities and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques"

In addition, I have checked the main text, tables, figures, and supplemental material, to correct all the relevant parts. A grammar check has been 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 358-365 have been 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, the influence of the second-order term for a perturbation beyond 20% is 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 439-440 has been 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-NO_X 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 have been 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 NO_X but not NH_3, and s08, which emit NH_3 but not NO_X. 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 NO_3^- and NH_4^+ 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 because of the nonlinear responses. Similar features are evident in the sensitivities derived by HDDM-100 and HDDM-20, implying that HDDM is capable of representing 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 NO_3^- to s04, and NH_4^+ 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, NO_3^- and NH_4^+ concentrations are never apportioned but nonlinearly sensitive to s08 and s04, respectively."

The third paragraph of the Section 3.6.3 has been divided into two and the latter one has been revised as follows based on the results for s01-VOC and s01-NO_X with 100% reduction. Figure 8 (attached as Fig. 1 in this reply) has been 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 linearly related to emissions. The sensitivities of NO_X emissions derived by BFM-20 and BFM-100 are also almost identical when they are negative. That means titration of ozone by NO_X is also linearly related to emissions. In contrast, the sensitivities to NO_X emissions derived by BFM-100 are higher than those derived by BFM-20 when they are positive. That means ozone formation from NO_X is nonlinearly related 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 (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 NO_3^- and NH_4^+ in section 3.6.2. That implies effects on concentrations of ozone, NO_3^-, and NH_4^+ may be less than those inferred by BFM-100 and ISAM when reductions of emissions of NO_X and NH_3 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 have added 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 have revised 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 have been replaced by the following sentences in the lines 24-29 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 NH_3 emissions and NH_4^+ concentrations, NO_X emissions and NO_3^- concentrations, and NO_X emissions and ozone concentrations – the sensitivities did not reach the apportionments because there were 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 has been inserted to the lines 100-101 as follows.

"Horizontal resolutions of d01, d02, d03, and d04 are 45 × 45 km, 15 × 15 km, 5 × 5 km, and 5 × 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 avoided 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 has been added as suggested as follows to the lines 210-211.

"As OC is the second major component of $PM_{2.5}$ following SO_4^{2-}"

[Referee #1]

6) Lines 196-197: "Less overestimates dots: : :" Consider deleting this sentence.

[Reply]

This sentence has been 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 has been added to the previous paragraph in the line 224 to just show the fact of the negative sensitivities to sea salts.

"The sensitivity of $PM_{2.5}$ to s12 (sea salt) is negative."

Explanations in the lines 238-242 have been revised as follows. I hope they make discussions clearer.

"The sensitivities of NO_3^- and NH_4^+ to s12 (sea salt) are negative. Cl^- originated from sea salts and mostly involved in coarse particles tend to be replaced by NO_3^- because of the so-called chlorine loss caused by gas-aerosol partitioning (Pio and Lopes, 1998; Chen et al., 2016). Therefore, if sea salts are present, more HNO_3 gases are partitioned to coarse particles. That provides capacities for NO_3^- and associated NH_4^+ involved in $PM_{2.5}$ to evaporate to the gas phase, resulting in negative sensitivities of $PM_{2.5}$ including NO_3^- and NH_4^+ 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 have corrected 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 257-273 has been divided into three to make easier to follow. The latter two paragraphs, originally in the lines 257 and 270, have started with the following sentences, respectively.

"One of possible reasons for these elevated contributions is reduction of emissions in Japan"
"Besides the changes in Chinese emissions, there are other reasons for the higher contributions from sources outside Japan."

The explanation of the normalization has been added to the beginning of this paragraph in the lines 244-247 as follows.

"Table S3 in Supplementary Material lists the ratios of the source sensitivities of the annual mean ozone and $PM_{2.5}$ 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% because of the 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 have been 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 has been inserted in the lines 317-318 as follows.

"All the values shown in Fig. 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 in the lines 352-354 have been revised as follows.

"The simulated SOA concentrations were characterized 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 has been replaced as suggested.

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

[Reply]
It has been replaced as suggested.

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

[Reply]
The sentence in the lines 507-510 has been revised as follows.

"While $PM_{2.5}$ concentrations were lower than those simulated by previous studies for past years because of 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 has been inserted in the lines 519-522. This paragraph has been merged to the precedent one because both contain similar descriptions.

"For example, the sensitivities of SO_4^{2-} and NO_3^- to the transport from outside Japan encompassed at least two undistinguished influencing factors, including the direct transport of SO_4^{2-} and NO_3^-, which were evaluated by their corresponding apportionments, and oxidation of SO_2 and NO_X emitted from domestic sources by OH originating in ozone transported from outside Japan."

[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 has been added to show sensitivities of $PM_{2.5}$ uniformly scaled by the ratios of observed and simulated concentrations of $PM_{2.5}$ components. Discussions on this figure have been inserted in the lines 536-545 as follows.

"Figure s10 in the Supplementary Material shows source sensitivities of the annual mean $PM_{2.5}$ concentrations derived by BFM in the regions. The values shown in (b) were uniformly scaled by the ratios of observed and simulated concentrations of $PM_{2.5}$ components shown in Table S2. The scaled sensitivities of $PM_{2.5}$ to the transport from outside Japan are higher by 1.0–2.2 μg/m^3 (15–40%) because of their high contributions to underestimated POA and SOA. The scaled sensitivities of $PM_{2.5}$ to other sources are different by 0–0.5 μg/m^3. This case assumes that deviations between observed and simulated $PM_{2.5}$ 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 NH_4^+ and NO_3^- concentrations are nonlinearly sensitive to NH_3 and NO_X emissions. Uncertainties in NH_3 and NO_X emission sources could largely influence source sensitivities as well as model performance of NH_4^+ and NO_3^- concentrations. More studies are necessary to increase the

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 in the lines 551-552 has been replaced by the following one.

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

Dear Referee #2:

[Referee #2]
The paper by Chatani et al. is based on source sensitivities and apportionments of O3 and PM2.5 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 have revised the manuscript based on your comments.

[Referee #2]
I have only two minor comments for the authors.
Line 178-179.
According to the simulations and statement, "The PM2.5 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 lines 194-195.

"A possible reason is discussed in section 3.2."

The description in the lines 265-269 has been 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 the 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 overestimated."

[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 PM2.5 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 PM2.5 precursors (for example NOx, SOx or VOCs) but not releasing O3 precursors. All combustion sources are strong VOC emitters, and efforts are made/have been made to abate NOx and SOx. 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 led 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 O_3 precursors. Discussions were too generalized. They should focus on SO_4^{2-} and NO_3^- as target species and SO_2 and NO_X as precursors. Corresponding expressions in this paragraph in the lines 405-413 have been 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 SO_2 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), because of its long lifetime in the atmosphere, resulting in less contrast in the sensitivities of $PM_{2.5}$ to s11 between western and eastern Japan, whereas 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 SO_2 and NO_X emissions in Japan will not effectively contribute to the reduction in the concentrations of SO_4^{2-} and NO_3^- 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 SO_2 and NO_2 are limited. The following sentence has been inserted in the line 410.

"whereas the sensitivities of domestic emissions are small"

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 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 $PM_{2.5}$ 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 $PM_{2.5}$ 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 $PM_{2.5}$. 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 in other countries and regions.

The paragraph has been be added at the end of Section 1 in the lines 85-91 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 $PM_{2.5}$ (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. The findings of this study contribute not only to solving remaining issues involving ozone and $PM_{2.5}$ in Japan, but also to understanding on possible influences of nonlinear relationships in other countries and regions."

I have tried to explain the importance of this study throughout the manuscript. In addition, results and discussions of additional simulations, which were conducted based on the comments of another reviewer, have been added. Please check them in the revised manuscript. I hope these revisions are interesting for you and readers.

I have also revised 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 has been changed as follows.

"Comprehensive analyses of source sensitivities and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques"

In addition, I have checked the main text, tables, figures, and supplemental material, to correct all the relevant parts. A grammar check has been 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 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 $PM_{2.5}$ concentrations. Not only the former but also the latter aspects are discussed in the lines 265-269. Discussions for the latter aspect have been revised to make clearer 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 the 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 overestimated."

[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 have tried 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 has been corrected.

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

[Reply]
It has been removed.

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

Comprehensive analyses of source sensitivities ~~to~~ and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques

Satoru Chatani[1], Hikari Shimadera[2], Syuichi Itahashi[3], Kazuyo Yamaji[4]

[1]National Institute for Environmental Studies, Tsukuba, Ibaraki 305-8506, Japan
[2]Osaka University, Suita, Osaka 565-0871, Japan
[3]Central Research Institute of Electric Power Industry, Abiko, Chiba 270-1194, Japan
[4]Kobe University, Kobe, Hyogo 658-0022, Japan

Correspondence to: Satoru Chatani (chatani.satoru@nies.go.jp)

Abstract. Source sensitivity and source apportionment are two major indicators representing source–~~-~~receptor relationships, which serve as essential information when considering effective strategies to accomplish improved air quality. This study evaluated source sensitivities ~~to~~ and apportionments of ambient ozone and $PM_{2.5}$ concentrations over Japan with multiple numerical techniques embedded in regional chemical transport models, including a brute forth method (BFM), a high-order decoupled direct method (HDDM), and an integrated source apportionment method (ISAM), to update the source–~~-~~receptor relationships considering stringent emission controls recently implemented in Japan and surrounding countries. We also attempted to understand the differences among source sensitivities and source apportionments calculated by multiple techniques. While a part of ozone concentrations was apportioned to domestic sources ~~had certain source apportionments to ozone concentrations~~, their sensitivities were small or even negative; ozone concentrations ~~they~~ were exclusively sensitive to transport from outside Japan ~~dominated the source sensitivities~~. Although the simulated $PM_{2.5}$ concentrations ~~and absolute magnitudes of their source sensitivities~~ were significantly lower than those reported by previous studies, ~~the~~ their sensitivity to transport from outside Japan were still relatively ~~has relatively~~ large ~~contributions to $PM_{2.5}$ concentrations~~, implying that there has been a reduction in Japanese emissions, similar to surrounding countries including China, due to implementation of stringent emission controls. HDDM allowed us to ~~effectively~~ understand the importance of the nonlinear responses of $PM_{2.5}$ concentrations to precursor emissions. Apportionments derived by ISAM were useful in distinguishing various direct and indirect influences on ozone and $PM_{2.5}$ concentrations by combining with sensitivities. ~~It was suggested~~The results indicate that ~~that~~ ozone transported from outside Japan plays a key role in exerting various indirect influences on the formation of ozone and secondary $PM_{2.5}$ components. While the sensitivities become closer to the apportionments when perturbations in emissions are larger in highly nonlinear relationships – including those between NH_3 emissions and NH_4^+ concentrations, NO_X emissions and NO_3^- concentrations, and NO_X emissions and ozone concentrations – the sensitivities did not reach the apportionments because there were 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. ~~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.~~

1. Introduction

The air quality of Japan has gradually improved. However, ambient concentrations of fine particulate matter smaller than 2.5 micrometres ($PM_{2.5}$) and photochemical oxidants (predominantly ozone) exceed the Environmental Quality Standards (EQS). Therefore, we must develop effective strategies to suppress ambient $PM_{2.5}$ and ozone concentrations. Quantitative source–receptor relationships serve as essential information when considering effective strategies. There are two major indicators representing source–receptor relationships (Clappier et al., 2017). One is source sensitivity, which corresponds to a change in ambient pollutant concentrations caused by a certain perturbation in precursor emissions. The second is source apportionment, which corresponds to the contribution of precursor emissions to ambient pollutant concentrations. Receptor modelling, including Chemical Mass Balance (CMB) and Positive Matrix Factorization (PMF) methods, have been widely applied to evaluate source apportionments (Hopke, 2016). However, they have limitations when attempting to treat secondary pollutants, which form in the atmosphere via complex photochemical reactions. Moreover, receptor modelling cannot evaluate source sensitivities. Forward modelling using a regional chemical transport model is a powerful tool for evaluating both the source sensitivities and apportionments of~~to~~ primary and secondary pollutants.

Several numerical techniques have been developed for regional transport models to evaluate source sensitivities and apportionments (Dunker et al., 2002; Cohan and Napelenok, 2011). A simple technique for evaluating source sensitivities is the brute force method (BFM). Differences in the simulated pollutant concentrations between two simulation cases with and without perturbations in the input precursor emissions is considered as the sensitivity ~~of~~ to a given emission source based on the BFM. This technique can require significant computational ~~demand~~ resources when evaluating the sensitivities ~~of~~ to many emission sources. A decoupled direct method (DDM) is a numerical technique that simultaneously tracks the evolution of sensitivity coefficients, in addition to pollutant concentrations when solving model equations (Yang et al., 1997). This method has been extended to a high-order DDM (HDDM) to track high-order sensitivity coefficients (Hakami et al., 2003). The ozone source apportionment technology (OSAT) (Dunker et al., 2002) and particulate matter source apportionment technology (PSAT) (Wagstrom et al., 2008) are numerical techniques that evaluate the source apportionments of ozone and particulate matter concentrations, respectively, by tagging contributions of precursor emissions to simulated concentrations. An integrated source apportionment method (ISAM) is a similar numerical technique that evaluates source apportionments (Kwok et al., 2013). Each method has its strengths and weaknesses, such that it is important to appropriately interpret results that will be used to develop effective strategies.

Source sensitivities and apportionments of ambient pollutant concentrations over Japan have been evaluated using regional chemical transport models. Chatani et al. (2011) evaluated the sensitivities of ~~domestic sources and transboundary transport to~~ simulated $PM_{2.5}$ concentrations over three metropolitan areas in Japan to domestic sources and transboundary

transport in the 2005 fiscal year. Ikeda et al. (2015) evaluated the sensitivities of ~~source regions in Japan, Korea, and China to~~ simulated $PM_{2.5}$ concentrations over the nine receptor regions in Japan to source regions in Japan, Korea, and China in 2010. These two studies only employed the BFM to derive source sensitivities ~~to~~ of $PM_{2.5}$ concentrations. Itahashi et al. (2015) evaluated the sensitivities and apportionments of ~~sources in Japan, Korea, and China to~~ simulated ozone concentrations over East Asia to sources in Japan, Korea, and China. ~~This~~ That study presented a unique exercise discussing the differences in source sensitivities and apportionments derived by multiple techniques, including the BFM, HDDM, and OSAT, in Asia; these differences have only been discussed in limited studies targeting the United States and Europe (Koo et al., 2009; Burr and Zhang, 2011; Thunis et al., 2019). Expanding targets is key to obtaining a more comprehensive and appropriate understanding of the source sensitivities and apportionments of ~~derived by multiple techniques to~~ pollutant concentrations, including ozone and $PM_{2.5}$, across Asia, including Japan, derived by multiple techniques.

In addition, recent studies (Ronald et al., 2017; Wang et al., 2017; Zheng et al., 2018) suggest that stringent emission controls implemented in China have achieved improved air quality. These improvements should affect air quality not only in China but also across downwind regions including Japan. We must, therefore, update source sensitivities and apportionments when considering additional effective strategies aimed at further air quality improvement in Japan.

Mutual inter-comparisons of the source sensitivities and apportionments derived by multiple models and numerical techniques is one of the objectives of Japan's Study for Reference Air Quality Modelling (J-STREAM) (Chatani et al., 2018b). Model inter-comparisons conducted in earlier phases of J-STREAM have contributed to the derivation of model configurations and development of emission inventories, both of which have contributed to improved model performance (Chatani et al., 2020; Yamaji et al., 2020). As one of the subsequent activities of J-STREAM, this study evaluates the sources sensitivities ~~to~~ of ozone and $PM_{2.5}$ concentrations simulated over regions in Japan for a recent year using the outcomes obtained in earlier phases of J-STREAM. Comprehensive analyses from various perspectives were performed to evaluate the sensitivities ~~of~~ to eight domestic and two natural emission source groups, as well as foreign anthropogenic emission sources and transboundary transport throughout the entire 2016 fiscal year. In addition, we perform mutual comparisons of the source sensitivities and apportionments ~~to~~ of simulated ozone and $PM_{2.5}$ concentrations. Although the target periods were limited to two weeks in four seasons, we discuss notable characteristics with respect to the differences in the source sensitivities and apportionments derived by the BFM, HDDM, and ISAM.

There are well-known nonlinear relationships between ambient concentrations of secondary pollutants including ozone and secondary components involved in $PM_{2.5}$ (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. The findings of this study contribute not only to solving remaining issues involving ozone and $PM_{2.5}$ in Japan, but also to understanding on possible influences of nonlinear relationships in other countries and regions.

2. Methodology

2.1. Model configuration

The Community Multiscale Air Quality (CMAQ) modelling system (Byun and Schere, 2006) version 5.0.2, in which both the HDDM and ISAM are embedded, was selected to calculate the source sensitivities and apportionments, in addition to ambient pollutant concentrations. The carbon bond chemical mechanism with the updated toluene chemistry (CB05-TU) (Whitten et al., 2010) and aero6 aerosol module were employed. Input meteorological fields were simulated by the Weather Research and Forecasting (WRF) - Advanced Research WRF (ARW) version 3.7.1 (Skamarock et al., 2008).

Horizontal locations and resolutions of the four target domains, named as d01, d02, d03, and d04, remain unchanged since the first phase of J-STREAM (Chatani et al., 2018b), as shown in Fig. 1. Horizontal resolutions of d01, d02, d03, and d04 are 45 × 45 km, 15 × 15 km, 5 × 5 km, and 5 × 5 km, respectively. The top height of the model was lifted from 10,000 to 5,000 Pa to explicitly treat transport in the lower stratosphere (Itahashi et al., 2019a). The vertical layer heights were adjusted to be consistent with those of Chemical Atmospheric Global Climate Model for Studies of Atmospheric Environment and Radiative Forcing (CHASER) (Sudo et al., 2002), which was used to provide boundary concentrations, to avoid numerical diffusions to adjacent layers. Each vertical layer of CHASER from the ground to 80,000 Pa was further divided into two to simulate vertical variations in the lower atmosphere in more detail. The bottom layer height was approximately 28 m.

~~Following s~~Several changes were applied to the original WRF configuration employed in the first phase of J-STREAM described in Chatani et al. (2018b) based on the outcomes of the model inter-comparisons. The input land use dataset was replaced with one created from geographic information system (GIS) data based on the 6th and 7th Vegetation Surveys released by the Biodiversity Centre of Japan, Ministry of Environment, which yielded improved performance for multiple meteorological parameters over urban areas (Chatani et al., 2018a). Lakes were added to the dataset based on the National Land Numerical Information Lakes Data. The shortwave and longwave radiation schemes were replaced with the Rapid Radiative Transfer Model for General circulations models (RRTMG) schemes (Iacono et al., 2008) to use the climatological ozone and aerosol profiles with spatial, temporal, and compositional variations (Tegen et al., 1997). Microphysics and cumulus schemes had significant influences on the simulated pollutant concentrations in the model inter-comparisons. A Morrison double-moment microphysics scheme (Morrison et al., 2009) and Grell–~~–~~Devenyi ensemble cumulus scheme (Grell and Devenyi, 2002) were newly selected because they were ~~characterised~~ characterized by better performance during the sensitivity experiments. Analysis datasets were replaced with the finer ones, i.e., the NCEP GDAS/FNL 0.25 Degree Global Tropospheric Analyses and Forecast Grids (ds083.3) (National Centers for Environmental Prediction/National Weather Service/NOAA/U.S. Department of Commerce, 2015) and Group for High Resolution Sea Surface Temperature (GHRSST) (Martin et al., 2012), for the initial and boundary conditions, as well as grid nudging. Nudging coefficients are critical parameters for model performance (Spero et al., 2018), but forcing terms in the model equations may disturb physical consistencies. While nudging coefficients for winds were set to 1.0 ×~~*~~ 10^{-4} sec^{-1} for all domains and vertical layers, those for temperature and water vapour were reduced to 5.0 ×~~*~~ 10^{-5}, 3.0 ×~~*~~ 10^{-5}, 1.0 ×~~*~~ 10^{-5}, and 1.0 ×~~*~~ 10^{-5} sec^{-1} for d01, d02, d03, and d04, respectively. In addition,

nudging for the temperature and water vapour within the planetary boundary layer in d03 and d04 was turned off to avoid excessive nudging to finer spatial and temporal scales than the input analysis datasets, as well as to allow the simulated values to be in accordance with the physical equations.

2.2. Emission inputs

Various improvements were applied to the original emission inputs used in the first phase of J-STREAM described in Chatani et al. (2018b) based on the outcomes of the model inter-comparisons. Hemispheric Transport of Air Pollution (HTAP) emissions version 2.2 (Janssens-Maenhout et al., 2015) was used for anthropogenic sources and international shipping for Asian countries except for Japan. While ~~its~~ the target year of HTAP v2.2 is 2010, the ratios of sectoral annual emissions reported by Zheng et al. (2018) were multiplied for China, and those reported by the Clean Air Policy Support System (CAPSS) (Lee et al., 2011) were multiplied for ~~China and~~ South Korea, ~~respectively,~~ to represent the changes in the precursor emissions of recent years. Itahashi et al. (2018) suggested the importance of heterogeneous reactions involving Fe and Mn in sulphate formation. The speciation profiles of Fu et al. (2013) were applied to consider other components, including Fe and Mn, in addition to originally available black and organic carbon in $PM_{2.5}$ emissions. The $PM_{2.5}$ emission inventory developed by the Ministry of Environment for the 2015 fiscal year was used for on-road and other transportation sectors in Japan. Emissions from stationary sources in Japan developed in J-STREAM (Chatani et al., 2018b) were fully updated to the 2015 fiscal year with the following improvements. The emission database of large point sources discretized into sectors, facilities, and fuel types were newly developed by Chatani et al. (2019) based on the Research of Air Pollutant Emissions from Stationary Sources to represent emissions characteristics and speciation profiles including Fe and Mn. Missing fugitive volatile organic compound (VOC) emission sources, including the use of repellents, air fresheners, aerosols inhalers, cosmetic products, and products for car washing and repair, were added to be consistent with the Greenhouse Gas Inventory Office of Japan (2018). NH_3 emissions from fertilizer use and manure management were replaced by the values reported by the Greenhouse Gas Inventory Office of Japan (2018). Fugitive VOC and PM emissions from manure management were newly estimated based on the European Environment Agency (2016). Emission factors of other NH_3 sources, including human sweat, human breath, dogs, and cats, were replaced by those reported in Sutton et al. (2000). PM emissions from the abrasion of railways wires and rails were newly estimated as one of the major sources of Fe and Mn. The method to estimate emissions from open agricultural residue burning were replaced by that used by the Greenhouse Gas Inventory Office of Japan (2018). We applied the emission factors reported in Fushimi et al. (2017) and Hayashi et al. (2014), as well as the temporal variations from Tomiyama et al. (2017). Biogenic VOC emissions were estimated by Chatani et al. (2018a) using a detailed database of vegetation and emission factors specific to Japan. The surf zone, defined as zones adjacent to beaches in the National Land Numerical Information Land Use Fragmented Mesh Data, was newly added to estimate higher sea salt emissions from these areas (Gantt et al., 2015) in the CMAQ.

2.3. Simulation setup

Ambient pollutant concentrations in d01, d02, d03, and d04 were simulated for the entire 2016 fiscal year (from April 2016 to March 2017). Simulations for the preceding month (March 2016) were treated as spin-up. Sensitivities ~~of~~ to the emission source groups, classified as listed in Table 1, were evaluated by the BFM, in which the emissions of each source group were reduced by 20% for the entire fiscal year in d02 and two selected weeks in spring (from May 6 to 20), summer (from July 21 to August 4), autumn (from October 20 to November 3), and winter (from January 19 to February 2 of 2017) in d03 and d04. These two weeks in the four seasons were the periods in which the monitoring campaigns for the ambient concentrations of the $PM_{2.5}$ components were conducted throughout Japan. The reason for choosing 20% reduction as a perturbation range in BFM is that it is a typical range of emission reduction by potential emission controls. For s11 (transport through the boundaries of d02), the boundary concentrations of all species for d02 were reduced by 20%. Differences in the concentrations scaled by five between the simulations with and without 20% perturbations were treated as sensitivities in this study. In addition, source sensitivities and apportionments ~~of~~ to all the emission source groups listed in Table 1 were evaluated by the HDDM and ISAM, respectively, using consistent inputs for the two coincident weeks in the four seasons in d02. The first- and second-order sensitivity coefficients ~~for~~ to gaseous precursors of a single emission source group were calculated using HDDM. We note that the HDDM results were missing for the seasons other than winter because the simulations were not successfully completed ~~due to~~because of numerical convergence problems. Table S1 in the Supplementary Material lists the annual total emission amounts for each source group in d02.

3. Results and discussion

3.1. Model performance on ozone and $PM_{2.5}$

We evaluated the model performance for the ozone and $PM_{2.5}$ concentrations in d02 for the entire 2016 fiscal year. Table S2 in the Supplementary Material lists the statistics for the model performance of the maximum daily 8-h average ozone (MDA8O3) and daily mean $PM_{2.5}$ concentrations. Table S2 includes the normalized mean bias (NMB), normalized mean error (NME), and correlation coefficient (R) (Emery et al., 2017) for entire Japan (JP), six regions (Kyushu-Okinawa, KO; Chugoku-Shikoku, CS; Kansai, KS; Tokai-Hokuriku, TH; Kanto-Koshinetsu, KK; Hokkaido-Tohuku, HT), and three areas designated by the Automobile NO_X-PM law as polluted urban areas (Osaka-Hyogo, OH; Aichi-Mie, AM; Shuto, ST). Figure 1 denotes the locations and abbreviations of the six regions and three designated areas. Automatic continuous monitoring data obtained at the ambient air pollution monitoring stations (APMSs) were used. Figure S1 in the Supplementary Material compares the observed and simulated monthly mean MDA8O3 and $PM_{2.5}$ concentrations averaged at all stations in the regions.

The MDA8O3 were slightly overestimated in all regions. The observed MDA8O3 was the highest in May and lowest in December. There was another peak in August in western Japan. The model consistently reproduced these monthly variations. Overestimation occurred from the peak in May to the ~~valley~~ low in December. Values from December to March were slightly

underestimated. The overestimation in summer in this study is less evident than that reported in Chatani et al. (2020), who summarized the performance of the models that participated in the model inter-comparisons conducted in the first phase of J-STREAM. The improved performance obtained in this study may be due to the various improvements in the configurations described in ~~Section~~ section 2, as well as differences in the meteorological conditions. Kitayama et al. (2019) shows that CB05-TU, which was employed in this study, tends to yield lower ozone concentrations among major chemical mechanisms. All the criteria proposed by Emery et al. (2017) were attained in all regions.

The $PM_{2.5}$ concentrations were underestimated in all regions. The statistics tended to be worse in eastern Japan as opposed to western Japan. The observed $PM_{2.5}$ concentrations fluctuated with a peak in May and valley near September. Although the simulations reproduced these monthly variations, the absolute values were consistently underestimated. A possible reason is discussed in section 3.2. The criteria proposed by Emery et al. (2017) were attained for NME and R, but not for NMB due to persistent underestimation.

As mentioned in ~~Section~~ section 2, monitoring campaigns for the ambient concentrations of the $PM_{2.5}$ components were conducted throughout Japan for the two target weeks in spring, summer, autumn, and winter. The components of the particulates collected on filters for ~~throughout one day~~24 hours were analysed. These data are useful for the further validation of model performance for the $PM_{2.5}$ components. Figure S2 in the Supplementary Material shows scatter plots of the observed and simulated daily concentrations of the $PM_{2.5}$ components (SO_4^{2-}, NO_3^-, NH_4^+, elemental carbon (EC), and organic carbon (OC)) at all locations throughout Japan during the monitoring campaigns in all four seasons. Table S1 summarizes their statistics for entire Japan and the four seasons. The simulated average concentrations of SO_4^{2-} and NH_4^+ are similar to the observed values. Their observed and simulated values have significant correlations with R, i.e., approximately 0.7. The NO_3^- concentrations were overestimated with NME of over 100%. The R between the observed and simulated values is 0.441, which is significantly lower than SO_4^{2-} and NH_4^+. A number of biased dots for NO_3^- occur in the scatter plot. While excessively higher simulated values appeared in summer, the model underestimated several of the higher values mainly observed in winter. Although previous studies have discussed issues of poor model performance associated with reproducing the NO_3^- concentrations in Japan (Shimadera et al., 2014; Shimadera et al., 2018), they have not yet been solved even after the application of various improvements. Both the EC and OC concentrations were underestimated. As OC is the second major component of $PM_{2.5}$ following SO_4^{2-}, its underestimation is one of the major causes of $PM_{2.5}$ underestimation. ~~Less overestimated dots are found in their scatter plots, indicating their persistent underestimation.~~ Shimadera et al. (2018) also discussed the issues of poor model performance associated with reproducing OC concentrations in Japan, suggesting condensable organic matter as a key factor for this poor performance. Although studies on this issue have been conducted by Morino et al. (2018), they remain unsolved.

We note that it is important to recognize that source sensitivities and apportionments introduced in the subsequent sections may be affected by the model performance described in this section.

3.2. Source sensitivities ~~on~~ of the annual mean ozone and $PM_{2.5}$

Figure 2 shows the source sensitivities ~~to~~ of the annual mean ozone and $PM_{2.5}$ concentrations derived by the BFM in all regions. ~~The sensitivity of~~ Ozone is overwhelmingly sensitive to ~~of~~ s11 (transport through the boundaries of d02) ~~is overwhelming to ozone~~. The sensitivities of ozone to domestic sources, including s01 (on-road vehicles) and s04 (stationary combustion), are negative in the three designated areas, which is caused by the titration of ozone ~~due to~~because of the higher NO_X emissions in urban areas. While ~~s11 has~~ the ~~highest~~ sensitivit~~ies~~ of~~to~~ $PM_{2.5}$ to s11 is the highest, $PM_{2.5}$ is also somewhat sensitive to ~~those of~~ domestic anthropogenic sources, including s01, s02 (ships), s04, and s08 (agriculture and fugitive ammonia)~~, are also evident~~. The sensitivities ~~of~~ to domestic anthropogenic sources are higher in the three designated areas with higher precursor emissions. The sensitivity of $PM_{2.5}$ to s12 (sea salt) is negative. The s~~S~~ums of the sensitivities of ozone to~~of~~ all the source groups ~~to ozone and $PM_{2.5}$~~ are lower ~~and higher~~ than their simulated concentrations, and the sums of the sensitivities of $PM_{2.5}$ are higher than their simulated concentrations~~respectively~~, due to the nonlinear relationships between their concentrations and precursor emissions.

The sensitivities ~~to~~ of $PM_{2.5}$ reflect the characteristics of the sensitivities ~~to~~ of individual $PM_{2.5}$ components. Figure S3 in the Supplementary Material shows the source sensitivities ~~to~~ of the annual mean concentrations of the $PM_{2.5}$ components derived by the BFM in all regions. The EC and primary organic aerosol (POA) are primary components. Sums of the sensitivities of these primary components to all the source groups ~~to these primary components~~ are consistent with the simulated concentrations. In the three designated areas (OH, AM and ST), ~~Higher sensitivities of~~ EC ~~and POA~~ are specifically sensitive to~~for specific source groups, including~~ s03 (non-road transport), and POA is specifically sensitive to ~~and~~ s05 (biomass combustion)~~, to EC and POA, respectively, are evident in the three designated areas~~. Sums of the sensitivities of ~~all the source groups to~~ SO_4^{2-}, which is mainly a secondary component but almost non-volatile, to all the source groups are also equivalent to the simulated concentrations. SO_4^{2-} is highly sensitive ~~The sensitivities of~~ to s09 (natural) ~~are higher~~ in western Japan, i.e., the location of several active volcanoes. Significant nonlinearities exist in the sensitivities ~~to~~ of NO_3^- and NH_4^+, which are mainly secondary components. Specifically, although s08 mainly emits NH_3 and no NO_X, ~~it is highly sensitive to~~ NO_3^- concentrations are highly sensitive to it ~~due to~~because of the indirect influences. Details of these nonlinearities are discussed in section 3.6, which compares the source sensitivities and apportionments. The sensitivities of NO_3^- and NH_4^+ to~~of~~ s12 (sea salt) are negative ~~to NO_3^- and NH_4^+~~. Cl^- originated from sea salts and mostly involved in coarse particles tend to be replaced by NO_3^- ~~due to~~because of the so-called chlorine loss caused by gas-aerosol partitioning (Pio and Lopes, 1998; Chen et al., 2016). Therefore, if sea salts are present, more HNO_3 gases are partitioned to coarse particles. That provides capacities for NO_3^- and associated NH_4^+ involved in $PM_{2.5}$ to evaporate to the gas phase, resulting in negative sensitivities of $PM_{2.5}$ including NO_3^- and NH_4^+ to sea salts. ~~If sea salts are absent, NO_3^- is more likely to be kept in $PM_{2.5}$ with NH_4^+.~~ Nonlinearities are also significant to secondary organic aerosol (SOA). SOA are specifically sensitive to b~~B~~iogenic VOC emissions included in s09 ~~are specifically sensitive to SOA~~.

Table S3 in Supplementary Material lists the ratios ~~(normalized or not)~~ of the source sensitivities ~~to~~ of the annual mean ozone and $PM_{2.5}$ 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% because of the 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. The annual mean $PM_{2.5}$ concentrations simulated in this study for the three designated areas are 6–9 μg/m^3, which is significantly lower than approximately 16 μg/m^3 simulated by Chatani et al. (2011) for the corresponding areas in the 2005 fiscal year. However, their ratios of~~for~~ the sensitivities ~~of~~ to foreign anthropogenic sources ~~are~~ were 48% in OH, 41% in AM, and 31% in ~~OH, AM, and~~ ST~~, respectively~~;, ~~which~~ these are lower than the approximately 65% calculated in this study as the sums of the sensitivities ~~for~~ to s10 (anthropogenic sources in other countries in d02) and s11. The normalized ratios for the sensitivities ~~of~~ to the sources in Korea and China for 2010 were 71% in Kyushu, 57% in Kinki, and 39% in ~~Kyushu, Kinki, and~~ Kanto, ~~respectively,~~ reported in Ikeda et al. (2015), whereas in this study the sensitivities ~~of~~ to s10 and s11 are 68% in KO (equivalent to Kyushu), 65% in KS (equivalent to Kinki), and 59% in KK (equivalent to Kanto)~~, which are equivalent to Kyushu, Kinki, and Kanto, were~~ are ~~68, 65, and 59%, respectively, in this study~~. Relative contributions of foreign sources evaluated in this study are even higher than previous studies for most areas of Japan despite the stringent emission controls implemented in China.

One of possible reasons ~~There are two possible reasons~~ for these elevated contributions is reduction of emissions in Japan. Zheng et al. (2018) showed that the emissions of $PM_{2.5}$, SO_2, and NO_X in China decreased by 31, 52, and 15%, respectively, from the 2010 to 2016 ~~due to~~as a result of the stringent emission controls. If we compare the emissions reported in Chatani et al. (2011) with those used in this study, which reflected changes in energy consumption and emission controls implemented since 2005, the emissions of $PM_{2.5}$, SO_2, and NO_X in Japan decreased by 29, 48, and 33%, respectively, from fiscal years 2005 to 2015. Therefore, the relative emission reductions in Japan may be larger than those in China if we assume certain ~~changes~~ increases in the emissions from 2005 to 2010. In particular, stringent emission controls implemented on diesel vehicles by the central and local governments were quite effective in suppressing $PM_{2.5}$ emissions and ambient concentrations in urban areas (Kondo et al., 2012). A reduction in the activity of the Miyakejima volcano in recent years has also resulted in lower SO_2 emissions. However, w~~W~~e 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 the 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 overestimated.~~Less contrasts in the sensitivities of foreign sources evaluated in this study between western Japan, which is more affected by westerly wind transport, and eastern Japan imply excessive estimates of emission reductions in Japan.~~

Besides the changes in Chinese emissions, there are other reasons for the higher contributions from sources outside Japan. ~~Another reason of the higher contributions of foreign sources owes to~~is ~~other factors than emissions in China, the fact that~~ s11 includes all the components that pass through the boundaries of d02, such that it is affected not only by anthropogenic sources in China, but also anthropogenic sources in other countries, natural sources, and background concentrations.

Ozone concentrations have been ~~nearly~~ relatively stable in Japan in recent years, while the NO_X and VOC concentrations have been supressed (Wakamatsu et al., 2013). Sensitivities derived in this study suggest that a continuous reduction in the NO_X emissions, ~~due to~~because of the stringent emission controls implemented in Japan, has resulted in increases in the annual mean ozone concentrations caused by less titration of the ozone in urban areas. Suppressing the annual mean ozone concentrations further is difficult because they ~~domestic sources~~ are practically insensitive to domestic sources. Trends in the transboundary transport of ozone likely have a significant effect on the mean annual ozone concentrations (Kurokawa et al., 2009; Chatani and Sudo, 2011). In contrast, the annual mean $PM_{2.5}$ concentrations are sensitive to domestic sources~~,~~ as well as transport from outside Japan~~, are sensitive to the annual mean $PM_{2.5}$ concentrations~~. The stringent emission controls implemented in Japan and surrounding countries appear to have contributed to their decreasing trends in Japan. Additional efforts to reduce emissions may produce further improvements in the annual mean $PM_{2.5}$ concentrations, whereas further validations of the emissions in Japan are necessary.

3.3. Monthly variations in source sensitivities ~~on~~ of ozone and $PM_{2.5}$

Figure 3 shows the source sensitivities ~~to~~ of the monthly mean ozone and $PM_{2.5}$ concentrations derived by BFM simulated ~~in entire~~for the whole of Japan (JP) and ST, which is one of the three designated areas, including the Tokyo metropolitan area. Figure S4 in the Supplementary material shows the sensitives ~~to~~ of the $PM_{2.5}$ components. Ozone is negatively sensitive ~~Negative sensitivities of the ozone concentration~~ to the domestic sources, including s01 (on-road vehicles) and s04 (stationary combustion), ~~to the ozone concentrations are evident~~ in winter ~~due to~~because of the titration of ozone by higher NO_X emissions and inactive photochemical reactions in urban areas. The sensitivity of ozone to s11 (transport through the boundaries of d02) is higher than the simulated concentrations, indicating that more ozone is transported from ~~the~~ outside Japan and titrated by NO_X emissions in Japan. In contrast, negative sensitivities ~~of~~ to domestic sources are less evident in summer even in the ST. Reductions in the ozone by titration are compensated by ozone formation from precursor emissions originating from domestic sources ~~due to~~because of the more active photochemical reactions. Differences can be observed in the major source groups, which have positive sensitivities in summer in JP and ST. While the sensitivities of s02 (ships) and s04, which mainly emit NO_X, are higher in JP, those of s07 (fugitive VOC) and s09 (natural), which mainly emit VOC, are higher in ST.

The sensitivity of $PM_{2.5}$ to ~~of~~ s11 ~~to $PM_{2.5}$~~ is the highest in May due to transport by dominant westerly winds in this season. The sensitivity ~~to~~ of POA is predominantly high, suggesting that it is affected by variable sources, such as open biomass burning. $PM_{2.5}$ in summer is highly sensitive ~~The sensitivities of~~ to s02, s04, and s09, which are mainly located in the southern sides of Japan, ~~are higher in the summer, caused by~~because of dominant southerly winds, as well as active secondary formation, which are clearly reflected in their sensitivities ~~to~~ of SO_4^{2-}. $PM_{2.5}$ in winter is highly sensitive ~~The sensitivities of~~ to s01 and s08 (agriculture and fugitive ammonia)~~ are high in winter~~. A colder and more stable atmosphere in winter favours the accumulation of emissions from local sources and the partitioning of NO_3^- and NH_4^+ to the aerosol phase, as reflected in their sensitivities.

As discussed for the annual mean concentrations, suppressing the monthly mean ozone concentrations is difficult because the sensitivities ~~of~~ to s11 are ~~overwhelming~~ dominant in all months. In particular, the sensitivities ~~of~~ to s01 and s04 are largely negative~~ly large~~ in urban areas in autumn and winter. Further reductions in their NO_X emissions may result in additional increases in the monthly mean ozone concentrations in these seasons. ~~We note that the ozone concentrations simulated in these seasons are lower than other seasons. Returning to the background concentration levels via reduced titration is inevitable.~~ In contrast, the negative sensitivities are less evident in spring and summer. Reductions in the precursor emissions for ~~certain~~ domestic sources have the possibility to suppress, to a certain extent, the monthly mean ozone concentrations. Effective sources may be different in urban and other areas ~~due to~~because of differences in ozone formation regimes (Inoue et al., 2019). The effects that strategies have on various sources of precursor emissions for $PM_{2.5}$ may vary seasonally ~~due to~~because of differences in meteorological and photochemical conditions.

3.4. Source sensitivities per unit precursor emissions

Air quality standards are defined in terms of ambient concentrations, ~~while~~ whereas targets for emission controls are defined in terms of emission amounts. Therefore, it is important to understand~~ing~~ ~~if~~ whether the sensitivities of ambient concentrations per equal emission amounts of different sources ~~to ambient concentrations~~ are consistent ~~for different sources is important~~or not. Figure 4 shows the sensitivities of the annual mean ambient concentrations of $PM_{2.5}$ components per annual total amount of corresponding precursor emissions of~~for~~ domestic anthropogenic sources (s01–s08) ~~in d02 to the annual mean ambient concentrations of the corresponding $PM_{2.5}$ components~~ in all of Japan. All the values shown in Fig. 4 were normalized by the EC value for s01, which is inert and emitted only in the bottom layer. The horizontal and vertical locations of the emissions have an effect on the differences in the values for the primary components (EC and POA). Here, s02 includes ship emissions in surrounding oceans in d02, whose values suggest that approximately 40% of the ship emissions in d02 affect the concentrations of primary $PM_{2.5}$ components over Japan. The values of s03 (non-road transport) and s04 (stationary combustion) are slightly lower because they include elevated sources, such as airplanes and large point sources. Slight differences among s01 (on-road vehicles), s05 (biomass combustion), and s06 (residential combustion), whose emissions were ingested only in the bottom layer, may be caused by differences in their horizontal distributions. Sources located in coastal areas may have lower influences as their emissions are transported beyond the land. Additional differences caused by photochemical reactions were observed for secondary components. The value ~~of~~ to s05 include agricultural residue burning, which has large spatial and temporal variations, such that its emissions may be high where secondary formation is relatively active. The value ~~for~~ of NH_4^+ ~~in~~ to s01 is significantly higher than that ~~of~~ to s08 (agriculture and fugitive ammonia) because s01 co-emits NO_X and NH_3, which have a mutual correlation.

The effectiveness of equal reduction amounts of the precursor emissions may be different among sources ~~due to~~because of photochemical reactions, as well as the locations of emissions.~~,~~ ~~which are~~These factors ~~that~~ may need to be considered when exploring effective strategies.

3.5. Differences in source sensitivities among domains

Nesting is a technique in air quality simulations aimed at obtaining improved model performance using finer meshes over target regions, as well as representing large-scale transport in coarser meshes in a computationally effective manner. This study employed d03 and d04 with finer 5 ×~~x~~ 5 km meshes over OH, AM, and ST, which ~~are~~ include all the major target urban areas. We emphasize the importance of observing how much the sensitivities evaluated in d03 and d04 are different from those in d02 using coarser 15 ×~~x~~ 15 km meshes. Figure 5 shows the sensitivities ~~of~~ to all the source groups over OH, AM, and ST evaluated in d02, d03, and d04 averaged for the two target weeks during the four seasons. The ozone concentrations simulated for the summer in d02 and d03 or d04 are slightly different. Negative sensitivities ~~of~~ to s01 (on-road vehicles) and s04 (stationary combustion) are correspondingly higher. Finer meshes tend to result in slightly larger influences of ozone titration. ~~While~~ Although the simulated $PM_{2.5}$ concentrations are slightly different in different domains, the relative contributions of the source groups to the sensitivities are consistent. These results suggest that differences in horizontal resolutions between d02 and d03 or d04 do not cause critical differences in the sensitivities when they are spatially and temporally averaged over the target areas and two weeks. They also support the validity of the discussions in this study, which are mostly based on the results obtained in d02.

3.6. Mutual comparisons of source sensitivities and apportionments derived by BFM, HDDM, and ISAM

3.6.1. Overall differences among techniques

Figure 6 shows the apportionments derived by ISAM and sensitivities derived by BFM and HDDM of ~~all the source groups to~~ the simulated ozone and $PM_{2.5}$ concentrations to all the source groups ~~in entire~~for the whole of Japan (JP) and ST averaged for the two target weeks during the four seasons. We used the following treatments in Fig. 6. Only the sensitivities ~~of~~ to the gaseous precursor emissions were calculated by HDDM. The sensitivities ~~of~~ to emissions and boundary concentrations of primary aerosol components (EC, POA, and other primary components) calculated by BFM were also used for HDDM. The simulated SOA concentrations were characterized as apportionments of "OTHR" in ISAM in this study because~~.~~ ~~Apportionments~~ apportionments ~~to~~ of SOA concentrations were not calculated by ISAM embedded in CMAQ version 5.0.2. ~~The simulated SOA concentrations were characterised as apportionments of "OTHR" in ISAM.~~ The HDDM sensitivities were evaluated using first- and second-order sensitivity coefficients ($S^{(1)}$ and $S^{(2)}$) based on the following Taylor expansion (Eq. (1)):

$$C(+\Delta\varepsilon) = C(0) + \Delta\varepsilon S^{(1)}(0) + \frac{\Delta\varepsilon^2}{2} S^{(2)}(0), \quad (1)$$

where $C(+\Delta\varepsilon)$ and $C(0)$ are the simulated concentrations with and without the perturbations, respectively~~,~~; $\Delta\varepsilon$ is a perturbation ratio~~,~~; and $S^{(1)}(0)$ and $S^{(2)}(0)$ are the first- and second-order sensitivity coefficients, respectively. The HDDM-20 corresponds to the value calculated by applying $\Delta\varepsilon = -0.2$ and multiplication by 5~~, which is equivalent to the value obtained by the BFM~~. If a sensitivity is represented by a second-order polynomial function, HDDM-20 is equivalent to the

value obtained by BFM. However, the influence of the second-order term for a perturbation beyond 20% is not reflected in HDDM-20 because the value at a 20% perturbation is just linearly extrapolated. They are reflected in t~~T~~he 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 d~~D~~ifferences between HDDM-20 and HDDM-100 correspond to the influences of the second-order term for a perturbation beyond 20%~~nonlinear responses against changes in the precursor emissions~~. Sums of the apportionments of all the source groups derived by ISAM represent, in principle, the simulated concentrations.

Not only the sensitivities described in previous sections, but also the apportionments of ozone to s11 (transport through the boundaries of d02) ~~to ozone concentrations~~ are ~~overwhelming~~dominant, suggesting that ozone over Japan is predominantly transported from outside Japan. There are certain positive apportionments of ozone to d~~D~~omestic sources, including s01 (on-road vehicles), s02 (ships), and s04 (stationary combustion), ~~have certain positive apportionments to ozone concentrations~~ in the spring and summer, indicating that a certain amount of ozone ~~forms~~ originates from precursors emitted from these sources. Nevertheless, ~~domestic sources have small or even negative~~ sensitivities ~~to~~ of ozone ~~concentrations~~ to domestic sources are small or even negative. Let us consider a simple example. Ozone transported from outside Japan reacts with NO emitted in Japan and forms NO_2 (step 1). Next, NO_2 is photochemically decomposed to NO and O, followed by ozone regeneration via a rapid reaction between O and O_2 (step 2). Potential ozone (ozone + NO_2) is preserved in these two steps (Itahashi et al., 2015). Regenerated ozone is apportioned to NO sources in Japan by ISAM in this case. However, if ozone transported from outside Japan increases and enough NO is available, there is a subsequent equivalent increase in NO_2 formation and ozone regeneration. This indicates that regenerated ozone is sensitive to transport from outside Japan. In contrast, if NO emissions in Japan increase, ozone concentrations decrease after step 1 or remain unchanged after step 2. This suggests that the sensitivities ~~of~~ to NO sources in Japan are negative after step 1 or ~~none~~ zero after step 2. Their sensitivities cannot become positive in this example. In reality, a certain amount of the NO is oxidized by other species, including RO_2 that originates from VOCs emitted in Japan. They result in net ozone formation and positive sensitivities, which compensates~~, to a certain extent,~~ negative sensitivities to a certain extent. The apportionment of ozone concentrations to s11 ~~to ozone concentrations~~ is smaller than their sensitivities in autumn and winter in ST. The apportionments ~~of~~ to domestic sources are negligible in these seasons. Ozone is titrated by high NO emissions in urban areas in step 1, whereas~~ile~~ step 2 is not fully reached ~~due to~~because of the inactive photochemical reactions.

There are differences in the source apportionments and sensitivities ~~to~~ of $PM_{2.5}$, which reflect those ~~to~~ of ~~the concentrations of~~ the $PM_{2.5}$ components, shown in Fig. S5 in the Supplementary Material. Sensitivities ~~to~~ of gaseous HNO_3 and NH_3 ~~concentrations~~, which are counterparts of NO_3^- and NH_4^+ in the gas phase, are also shown in Fig. S5. The source apportionments and sensitivities ~~to~~ of primary components (EC and POA) are consistent. While the sums of the source apportionments and sensitivities of SO_4^{2-} to ~~of~~ all the sources ~~to SO_4^{2-}~~ are also consistent, there are differences in the relative contributions of the source groups. The apportionment ~~of~~ to s11 corresponds to the concentrations of SO_4^{2-} transported from outside Japan. The higher sensitivities are affected by additional indirect influences, i.e., SO_2 is oxidized to H_2SO_4 via gaseous

and aqueous reactions, and is then predominantly partitioned to SO_4^{2-}. Gaseous ~~oxidation of~~ SO_2 is oxidized by ~~occurs due to~~ OH, a part of which originates in ozone. Therefore, s11, which has an overwhelmingly high sensitivity to ozone, also has higher sensitivities ~~to~~ of SO_4^{2-} oxidized from SO_2. In contrast, if SO_2 emissions are reduced under fixed OH, other SO_2 remaining in the atmosphere has the opportunity to be oxidized to SO_4^{2-}. Therefore, the sensitivities ~~of~~ to downwind domestic sources are smaller than their apportionments. Similar discussions are applicable to NO_3^-. The apportionments of ~~s11 to~~ NO_3^- and HNO_3 to s11 are lower than ~~its~~ their sensitivities, indicating that a certain amount of the NO_3^- and HNO_3 is not directly transported from outside Japan. Ozone overwhelmingly affected by s11 enhances the oxidation of NO_X to HNO_3 through OH, followed by a smaller amount that is further partitioned to NO_3^-. This causes indirect influences on the sensitivities ~~of~~ to s11. Such influences are apparent in the horizontal distributions of the apportionments and sensitivities of ~~s11 to~~ concentrations of related species to s11 for the two target weeks of spring shown in Fig. S6 in the Supplementary Material. The sensitivities ~~to~~ of SO_4^{2-} and NO_3^- are higher than their apportionments over Japan. The sensitivities of SO_2 and NO_2 over Japan are correspondingly negative, suggesting that they are oxidized by OH that originated in ozone transported from outside Japan. The isolated higher sensitivities over Japan, particularly visible for those ~~to~~ of NO_3^-, clearly suggest that they are not directly transported from outside Japan.

Section 3.2 discussed higher relative contributions than previous studies and less contrasts between western and eastern Japan for the sensitivities of ~~s11 to the~~ $PM_{2.5}$ ~~concentrations~~ to s11 obtained in this study. Oxidation of ~~precursors~~ SO_2 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~~because of its long lifetime in the atmosphere, resulting in less contrast~~s~~ in the sensitivities of $PM_{2.5}$ to s11 between western and eastern Japan, whereas the sensitivities of domestic emissions are small. Ozone governs ~~an~~ the oxidative capacity of the atmosphere (Prinn, 2003). If ozone transported from outside Japan is not as reduced in future, efforts to reduce ~~precursor~~ SO_2 and NO_X emissions in Japan will not effectively contribute to the reduction in the concentrations of ~~secondary $PM_{2.5}$ components~~SO_4^{2-} and NO_3^- because OH that originated in ozone transported from outside Japan affects their formation.

There is no apportionment of NO_3^- to s08 (agriculture and fugitive ammonia), which emits NH_3 and no NO_X, ~~has no apportionments to NO_3^-~~ in accordance to the principle. Nevertheless, NO_3^- is highly sensitive to s08~~it has high sensitivities to of NO_3^-~~, ~~which are~~and is affected by the relationships between NH_4^+ and NO_3^-. Here, NH_4^+ and NO_3^- are mutual counter ions in NH_4NO_3, whose formation is enhanced when both are available. More NH_3 emissions can induce the partitioning of HNO_3 to NH_4NO_3. These influences can be observed in the correspondingly negative sensitivities of ~~s08 to~~ gaseous HNO_3 to s08. While the apportionments ~~to~~ of NH_4^+ are dominated by s08 in ST, its sensitivities are significantly smaller than the apportionments. Both $(NH_4)_2SO_4$ and NH_4NO_3 are major forms of NH_4^+, where, as discussed above, NH_4NO_3 formation is sensitive to NH_3 emissions. In contrast, the sensitivities of ~~s08 to~~ SO_4^{2-} ~~concentrations~~ to s08 are negligible (Fig. S5(c2)), suggesting that $(NH_4)_2SO_4$ formation is predominantly limited by SO_2 sources, including s02 and s04. Their influences are reflected in the sensitivities of ~~s02 and s04 to~~ NH_4^+ ~~concentrations~~ to s02 and s04 (Fig. S5(e2)). These results are consistent

with Clappier et al. (2017), who discuss the differences between apportionments and sensitivities in different regimes involving SO_2, NO_X, and NH_3 using idealized example cases.

There is the certain degree of sensitivity of $PM_{2.5}$ ~~concentrations~~ to s08 ~~has certain sensitivities to $PM_{2.5}$ concentrations~~, as shown in Fig. 2(b), which are indirectly caused by the interactions between NH_4^+ and NO_3^-. There ~~are~~ have been several studies that ~~insist on~~have highlighted the importance of NH_3 emission controls to reduce $PM_{2.5}$ concentrations (Pinder et al., 2007; Wu et al., 2016; Guo et al., 2018). Such discussions are applicable to Japan. However, Liu et al. (2019) suggested that NH_3 emission control could worsen acid rain because nitric acid is not neutralized and remains in the atmosphere. When seeking strategies to achieve sustainable developments, it is necessary to consider ~~Not only reducing $PM_{2.5}$ concentrations, but also~~ other environmental aspects, including acid rain and nitrogen cycles, as well as the reduction of $PM_{2.5}$ concentrations~~, should be considered to develop strategies effective at accomplishing sustainable developments~~.

3.6.2. Nonlinear responses in sensitivities

Differences among the sensitivities derived by BFM, HDDM-20, and HDDM-100 are mostly small, suggesting that, in most cases, HDDM is able to calculate sensitivities consistent with BFM. Slight differences were found in the sensitivities ~~to~~ of NO_3^- ~~concentrations~~ derived by them. Figure 7(a) shows the sensitivities of the daily NO_3^- concentrations to the source groups located within d02 (s01–s10) derived by BFM, HDDM-20, and HDDM-100 ~~to the daily NO_3^- concentrations~~ for the two target weeks in winter in ST. The sensitivities derived by BFM are slightly higher than those derived by HDDM-20. While the sensitivities derived by HDDM-20 are only affected by gaseous precursor emissions, those derived by BFM contain minor contributions of primary emitted NO_3^-. They are one of the factors that may result in higher sensitivities derived by BFM. However, differences were found even in the sensitivities ~~of~~ to s08 (agriculture and fugitive ammonia), which mostly emits NH_3. Differences should be recognized as difficulties in representing sensitivities only with first- and second-order sensitivity coefficients derived by HDDM~~uncertainties that originated from two principally different methodologies~~.

Sums of the sensitivities derived by HDDM-100 are higher than those derived by HDDM-20 for all days, indicating nonlinear responses of NO_3^- concentrations against precursor emissions. Daily variations of two additional indicators are shown in Fig. 7(b). One is a nonlinear index (Cohan et al., 2005), which is calculated as follows:

$$Nonlinear\ index = \left|\frac{0.5S^{(2)}}{S^{(1)}}\right|. \quad (2)$$

This corresponds to an absolute ratio of the second- to first-order sensitivity terms when a perturbation is $\Delta\varepsilon = \pm 1.0$, indicating the strength of the nonlinearities. Another indicator is an available NH_3 ratio, which corresponds to a ratio of NH_3 + NH_4^+ (those stoichiometrically equivalent to SO_4^{2-} are subtracted) to HNO_3 + NO_3^-, indicating an abundance of potential NH_4^+ that can be combined with NO_3^-. Here, s08 has the highest nonlinear indices that cause the overall nonlinearities, implying that the NO_3^- concentrations have nonlinear responses to NH_3 emissions. Daily variations in the nonlinear indices of s08 and available NH_3 ratios are well correlated; nonlinearities are higher when available NH_3 ratios are lower. The formation of NH_4NO_3 tends to be more constrained by NH_3 with less available NH_3, as shown by Xing et al. (2011). A typical situation

485 occurred on 30 January ~~30th~~. Negative sensitivities of s04 (stationary combustion) suggest that SO_2 emissions of s04 remove NH_3 to form $(NH_4)_2SO_4$ and prevent NH_4NO_3 formation. The HDDM can represent such complex nonlinear relationships involving multiple species.

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 NO_X but not NH_3, and s08, which
490 emit NH_3 but not NO_X. 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 NO_3^- and NH_4^+ 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 because of the nonlinear responses. Similar features are evident in the sensitivities derived by HDDM-100 and HDDM-20, implying that HDDM is capable of representing directions of nonlinear responses beyond 20% perturbation. It is notable that
495 the sensitivities derived by BFM with a larger perturbation become closer to the apportionments for NO_3^- to s04, and NH_4^+ 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, NO_3^- and NH_4^+ concentrations are never apportioned but nonlinearly sensitive to s08 and s04, respectively.

3.6.3. Dependence of ozone formation on NO_X and VOC

500 ISAM has the capability to separately calculate apportionments of ozone to NO_X and VOC emissions of a given source ~~to ozone concentrations~~ based on ozone formation conditions (Kwok et al., 2013). It is important to understand relationships between apportionments and sensitivities of ozone to NO_X and VOC emissions. ~~Understanding their relationships with the corresponding sensitivities is important.~~ Additional simulations were conducted to separately derive the sensitivities of ozone to NO_X and VOC emissions of s01 (on-road vehicles) ~~to ozone concentrations~~ by BFM with 20% (BFM-20) and
505 100% (BFM-100) perturbations.

Figure 8 shows the sensitivities derived by BFM-20 and BFM-100, and apportionments derived by ISAM ~~and sensitivities~~ of daily ozone concentrations to the NO_X and VOC emissions of s01 ~~derived by ISAM and BFM to daily ozone concentrations~~ for the two target weeks in summer in ST. The apportionment ~~of~~ to the NO_X emissions is higher than ~~that~~ the apportionment ~~of~~ to the VOC emissions. While there are differences in the magnitudes of the apportionments and sensitivities
510 ~~of~~ to the VOC emissions, the~~y have consistent~~ir daily variations are consistent. The sensitivity ~~of~~ to the NO_X emissions is mostly negative, but became positive on 25 July ~~25th~~ when the apportionments, as well as the ozone concentrations, were the highest. The dominant winds were northerly until 24 July ~~24th~~ and switched to southerly on 25 July ~~25th~~. Precursors and the ozone formed from them were transported to the south and returned to ST. Therefore, ~~a relatively~~the aged airmass passed over ST on 25 July 25th. Influences of ozone formation from NO_X emissions were higher than the immediate titration by them for
515 this condition.

Figure ~~S7~~S8 in the Supplementary Material shows the sensitivities derived by BFM-20 and BFM-100, and apportionments derived by ISAM ~~and sensitivities~~ of the hourly ozone concentrations to the NO_X and VOC emissions of s01

~~derived by ISAM and BFM to the hourly ozone concentrations~~ on 25 July ~~25th~~ in ST. Hourly variations in the apportionments and sensitivities ~~of~~ to the VOC emissions are consistent. ~~While~~ Whereas the sensitivities ~~of~~ to the NO_X emissions during the night are slightly negative ~~due to~~because of titration, their higher positive sensitivities during the daytime indicate the contribution of the NO_X emissions to the high ozone concentrations.

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 linearly related to emissions. The sensitivities of NO_X emissions derived by BFM-20 and BFM-100 are also almost identical when they are negative. That means titration of ozone by NO_X is also linearly related to emissions. In contrast, the sensitivities to NO_X emissions derived by BFM-100 are higher than those derived by BFM-20 when they are positive. That means ozone formation from NO_X is nonlinearly related to emissions. ~~the sensitivities of the NO_X and VOC emissions calculated by BFM with 20% perturbations are still lower than their apportionments in this condition, which may be caused by nonlinear responses of ozone concentrations against precursor 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 ~~of~~ 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 NO_3^- and NH_4^+ in section 3.6.2. That implies effects on concentrations of ozone, NO_3^-, and NH_4^+ may be less than those inferred by BFM-100 and ISAM when reductions of emissions of NO_X and NH_3 are small.

Figure ~~S8~~S9 in the Supplementary Material shows the horizontal distributions of the apportionments and sensitivities of the ozone concentrations to the s01 NO_X and VOC emissions ~~to the ozone concentrations~~ averaged for the two target weeks in summer. The sensitivity ~~of~~ to NO_X emissions is negative in urban and coastal areas where NO_X emissions from on-road vehicles are high. There are consistencies in the horizontal distributions of the positive sensitivities and apportionments ~~of~~ to NO_X and VOC emissions. While there are quantitative differences in the magnitudes of the sensitivities and apportionments due to nonlinear influences, ISAM ~~can~~ provides ~~the~~ spatial and temporal variations in the apportionments ~~of~~ to NO_X and VOC emissions consistent with the sensitivities derived by BFM.

4. Conclusions

Sensitivities and apportionments of ~~emissions from twelve source groups to~~ ozone and $PM_{2.5}$ concentrations over regions in Japan for the 2016 fiscal year to emissions from twelve source groups were evaluated by the BFM, HDDM, and ISAM using emissions data that take into account the latest stringent emission controls. Ozone was predominantly sensitive to t~~T~~ransport from outside Japan ~~dominated the source sensitivities to of ozone concentrations~~. While $PM_{2.5}$ concentrations ~~and their absolute sensitivities of all the sources~~ were lower than those ~~calculated~~ simulated by previous studies for past years ~~due to~~because of 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 ~~similar~~ to surrounding countries, including China. Moreover, ~~their~~ sensitivities of $PM_{2.5}$ ~~calculated in this study~~ included indirect influences of ozone predominantly transported from outside Japan via the oxidation of precursors by OH to secondary $PM_{2.5}$ components. There was a certain sensitivity of $PM_{2.5}$ to d~~D~~omestic sources ~~had certain sensitivities to $PM_{2.5}$~~, but the sensitivity of ozone to domestic sources was significantly smaller or even negative ~~sensitivities to ozone due to~~ because of titration and nonlinear responses against precursor emissions.

Sensitivities and apportionments ~~for~~ of primary species were consistent. Fundamental differences were found between them for secondary species. ~~While~~ Whereas apportionments represent direct contributions, sensitivities include indirect influences. Clappier et al. (2017) and Thunis et al. (2019) have suggested that sensitivities can provide more useful information than apportionments when considering effective strategies. This study indicates that apportionments simultaneously evaluated with sensitivities can be useful in distinguishing direct and indirect influences, i.e. they cannot be distinguished only by sensitivities. For example, the sensitivities of SO_4^{2-} and NO_3^- to the transport from outside Japan encompassed at least two undistinguished influencing factors, including the direct transport of SO_4^{2-} and NO_3^-, which were evaluated by their corresponding apportionments, and oxidation of SO_2 and NO_X emitted from domestic sources by OH originating in ozone transported from outside Japan. In addition, ~~Comparisons between apportionments and sensitivities can help distinguishing direct and indirect influences. Various indirect influences on ozone and $PM_{2.5}$ were identified in this study, including~~ the titration of ozone by NO_X emissions~~, oxidation of precursors by OH that originated in ozone,~~ and inter-correlations between NH_4^+ and NO_3^- in their partitioning were also identified as key indirect influences on ozone and $PM_{2.5}$.

~~, all of which could occur everywhere in the world.~~ Sensitivities of $PM_{2.5}$ derived by BFM and HDDM were mostly consistent except for NO_3^- and NH_4^+. There were differences between the sensitivities of NO_3^- and NH_4^+ calculated with the first- and second-order sensitivity coefficients derived by HDDM and those derived by BFM. ~~The~~ HDDM ~~also~~ revealed possibilities to ~~represent~~ indicate directions of nonlinear responses to larger perturbations in emissions. ~~of concentrations to precursor emissions.~~ The sensitivities derived by BFM become closer to the apportionments derived by ISAM when perturbations in emissions are larger in highly nonlinear relationships, including those between NH_3 emissions and NH_4^+ concentrations, NO_X emissions and NO_3^- concentrations, and NO_X emissions and ozone concentrations. However, the sensitivities did not reach the apportionments because of the various indirect influences, including other sectors, complex photochemical reactions, and gas-aerosol partitioning. The dependence of ozone formation on the NO_X and VOC emissions derived by ISAM was spatially and temporally consistent with sensitivities derived by BFM.

~~Quantitative source receptor relationships serve as essential information when considering effective strategies, as described in the introduction.~~ Clappier et al. (2017) ~~and~~ Thunis et al. (2019) ~~suggested that sensitivities can provide more useful information than apportionments when considering effective strategies. This study indicates that apportionments simultaneously evaluated with sensitivities are also helpful to distinguish direct and indirect influences, i.e., they cannot be distinguished only by sensitivities. For example, the sensitivities of SO_4^{2-} and NO_3^- concentrations to the transport from outside Japan encompassed at least two undistinguished influencing factors, including the direct transport of SO_4^{2-} and NO_3^-, which were evaluated by their corresponding apportionments, and oxidation of SO_2 and NO_X emitted from domestic sources by ozone~~

~~transported from outside.~~ Understanding the influences that various factors have on sensitivities can contribute to the establishment of effective strategies. However, accurate sensitivities and apportionments depend on model performance. Uncertainties remain in model performance, as discussed in section 3.1. If specific emission sources affect overall model performance, source sensitivities and apportionments derived by models may be skewed. Figure s10 in the Supplementary Material shows source sensitivities of the annual mean $PM_{2.5}$ concentrations derived by BFM in the regions. The values shown in (b) were uniformly scaled by the ratios of observed and simulated concentrations of $PM_{2.5}$ components shown in Table S2. The scaled sensitivities of $PM_{2.5}$ to the transport from outside Japan are higher by 1.0–2.2 $\mu g/m^3$ (15–40%) because of their high contributions to underestimated POA and SOA. The scaled sensitivities of $PM_{2.5}$ to other sources are different by 0–0.5 $\mu g/m^3$. This case assumes that deviations between observed and simulated $PM_{2.5}$ 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 NH_4^+ and NO_3^- concentrations are nonlinearly sensitive to NH_3 and NO_X emissions. Uncertainties in NH_3 and NO_X emission sources could largely influence source sensitivities as well as model performance of NH_4^+ and NO_3^- concentrations. ~~—~~More studies are necessary to ~~obtain~~ increase the~~more~~ confidence in source sensitivities and apportionments as well as model performance. In addition, sensitivities obtained by the BFM with a single perturbation may be inappropriate for applications to different perturbation ranges when nonlinearities are higher. High-order sensitivity coefficients calculated by the HDDM could help evaluate the importance of nonlinear responses.

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~~ these are vital for the development of effective strategies for improved air quality, using consistent model configurations and inputs. However, model configurations and inputs may not necessarily be consistent. ~~In reality, different model configurations and inputs may be used to consider strategies.~~ Itahashi et al. (2019b) reported that source sensitivities can be changed by the regional chemical transport model with improved treatments for aqueous reactions. Uncertainties in the sensitivities and apportionments caused by different model configurations and inputs should be explored as the next step of J-STREAM.

Data availability

The input datasets are available upon request at http://www.nies.go.jp/chiiki/jstream.html. The output datasets are available upon request to the authors.

Author contribution

SC designed this study, conducted BFM simulations, and prepared the manuscript. HS ~~and SI~~ conducted ISAM simulations, and SI conducted HDDM simulations~~, respectively~~. KY prepared the meteorology and additional inputs.

Competing interests

There are no conflicts of interest to declare.

Acknowledgements

This study was supported by the Environment Research and Technology Development Fund (5-1601JPMEERF20165001 and JPMEERF2018500255-1802) of the Environmental Restoration and Conservation Agency of Japan. The data based on the 6th and 7th Vegetation Surveys was obtained from the Biodiversity Centerre of Japan, Ministry of the Environment (http://gis.biodic.go.jp/webgis/sc-006.html). The National Land Numerical Information data was obtained from the National Land Numerical Information download service (http://nlftp.mlit.go.jp/ksj/index.html). Data from the Research of Air Pollutant Emissions from Stationary Sources was provided by the Ministry of the Environment. Automatic continuous monitoring data of the ambient air pollution monitoring stationsAPMSs was obtained from the National Institute for Environmental Studies (http://www.nies.go.jp/igreen/). The data associated with the monitoring campaigns for the ambient concentrations of the $PM_{2.5}$ components was obtained from the Ministry of the Environment (http://www.env.go.jp/air/osen/pm/monitoring.html).

References

Burr, M. J., and Zhang, Y.: Source apportionment of fine particulate matter over the Eastern U.S. Part II: source apportionment simulations using CAMx/PSAT and comparisons with CMAQ source sensitivity simulations, Atmospheric Pollution Research, 2, 318-336, doi:10.5094/apr.2011.037, 2011.

Byun, D., and Schere, K. L.: Review of the governing equations, computational algorithms, and other components of the models-3 Community Multiscale Air Quality (CMAQ) modeling system, Appl. Mech. Rev., 59, 51-77, doi:10.1115/1.2128636, 2006.

Chatani, S., Morikawa, T., Nakatsuka, S., and Matsunaga, S.: Sensitivity analyses of domestic emission sources and transboundary transport on $PM_{2.5}$ concentrations in three major Japanese urban areas for the year 2005 with the three-dimensional air quality simulation, J. Jpn. Soc. Atmos. Environ., 46, 101-110, doi:10.11298/taiki.46.101, 2011.

Chatani, S., and Sudo, K.: Influences of the variation in inflow to East Asia on surface ozone over Japan during 1996-2005, Atmos. Chem. Phys., 11, 8745-8758, doi:10.5194/acp-11-8745-2011, 2011.

Chatani, S., Okumura, M., Shimadera, H., Yamaji, K., Kitayama, K., and Matsunaga, S.: Effects of a detailed vegetation database on simulated meteorological fields, biogenic VOC emissions, and ambient pollutant concentrations over Japan, Atmosphere, 9, 179, 2018a.

Chatani, S., Yamaji, K., Sakurai, T., Itahashi, S., Shimadera, H., Kitayama, K., and Hayami, H.: Overview of model inter-comparison in Japan's Study for Reference Air Quality Modeling (J-STREAM), Atmosphere, 9, 19, 2018b.

Chatani, S., Cheewaphongphan, P., Kobayashi, S., Tanabe, K., Yamaji, K., and Takami, A.: Development of Ambient Pollutant Emission Inventory for Large Stationary Sources Classified by Sectors, Facilities, and Fuel Types in Japan, J. Jpn. Soc. Atmos. Environ., 54, 62-74, doi:10.11298/taiki.54.62, 2019.

Chatani, S., Yamaji, K., Itahashi, S., Saito, M., Takigawa, M., Morikawa, T., Kanda, I., Miya, Y., Komatsu, H., Sakurai, T., Morino, Y., Nagashima, T., Kitayama, K., Shimadera, H., Uranishi, K., Fujiwara, Y., Shintani, S., and Hayami, H.: Identifying key factors influencing model performance on ground-level ozone over urban areas in Japan through model inter-comparisons, Atmos. Environ., 223, 117255, doi:https://doi.org/10.1016/j.atmosenv.2019.117255, 2020.

Chen, Y., Cheng, Y. F., Ma, N., Wolke, R., Nordmann, S., Schuttauf, S., Ran, L., Wehner, B., Birmili, W., van der Gon, H., Mu, Q., Barthel, S., Spindler, G., Stieger, B., Muller, K., Zheng, G. J., Poschl, U., Su, H., and Wiedensohler, A.: Sea salt emission, transport and influence on size-segregated nitrate simulation: a case study in northwestern Europe by WRF-Chem, Atmos. Chem. Phys., 16, 12081-12097, doi:10.5194/acp-16-12081-2016, 2016.

Clappier, A., Belis, C. A., Pernigotti, D., and Thunis, P.: Source apportionment and sensitivity analysis: two methodologies with two different purposes, Geosci. Model Dev., 10, 4245-4256, doi:10.5194/gmd-10-4245-2017, 2017.

Cohan, D. S., Hakami, A., Hu, Y. T., and Russell, A. G.: Nonlinear response of ozone to emissions: Source apportionment and sensitivity analysis, Environ. Sci. Technol., 39, 6739-6748, doi:10.1021/es048664m, 2005.

Cohan, D. S., and Napelenok, S. L.: Air quality response modeling for decision support, Atmosphere, 2, 407-425, doi:10.3390/atmos2030407, 2011.

Dunker, A. M., Yarwood, G., Ortmann, J. P., and Wilson, G. M.: Comparison of source apportionment and source sensitivity of ozone in a three-dimensional air quality model, Environ. Sci. Technol., 36, 2953-2964, doi:10.1021/es011418f, 2002.

Emery, C., Liu, Z., Russell, A. G., Odman, M. T., Yarwood, G., and Kumar, N.: Recommendations on statistics and benchmarks to assess photochemical model performance, J. Air Waste Manage. Assoc., 67, 582-598, doi:10.1080/10962247.2016.1265027, 2017.

European Environment Agency: EMEP/EEA air pollutant emission inventory guidebook 2016, 2016.

Fu, X., Wang, S. X., Zhao, B., Xing, J., Cheng, Z., Liu, H., and Hao, J. M.: Emission inventory of primary pollutants and chemical speciation in 2010 for the Yangtze River Delta region, China, Atmos. Environ., 70, 39-50, doi:10.1016/j.atmosenv.2012.12.034, 2013.

Fushimi, A., Saitoh, K., Hayashi, K., Ono, K., Fujitani, Y., Villalobos, A. M., Shelton, B. R., Takami, A., Tanabe, K., and Schauer, J. J.: Chemical characterization and oxidative potential of particles emitted from open burning of cereal straws and rice husk under flaming and smoldering conditions, Atmos. Environ., 163, 118-127, doi:10.1016/j.atmosenv.2017.05.037, 2017.

Gantt, B., Kelly, J. T., and Bash, J. O.: Updating sea spray aerosol emissions in the Community Multiscale Air Quality (CMAQ) model version 5.0.2, Geosci. Model Dev., 8, 3733-3746, doi:10.5194/gmd-8-3733-2015, 2015.

Greenhouse Gas Inventory Office of Japan: National Greenhouse Gas Inventory Report of Japan, 2018.

Grell, G. A., and Devenyi, D.: A generalized approach to parameterizing convection combining ensemble and data assimilation techniques, Geophys. Res. Lett., 29, doi:10.1029/2002gl015311, 2002.

Guo, H. Y., Otjes, R., Schlag, P., Kiendler-Scharr, A., Nenes, A., and Weber, R. J.: Effectiveness of ammonia reduction on control of fine particle nitrate, Atmos. Chem. Phys., 18, 12241-12256, doi:10.5194/acp-18-12241-2018, 2018.

Hakami, A., Odman, M. T., and Russell, A. G.: High-order, direct sensitivity analysis of multidimensional air quality models, Environ. Sci. Technol., 37, 2442-2452, doi:10.1021/es020677h, 2003.

Hayashi, K., Ono, K., Kajiura, M., Sudo, S., Yonemura, S., Fushimi, A., Saitoh, K., Fujitani, Y., and Tanabe, K.: Trace gas and particle emissions from open burning of three cereal crop residues: Increase in residue moistness enhances emissions of carbon monoxide, methane, and particulate organic carbon, Atmos. Environ., 95, 36-44, doi:10.1016/j.atmosenv.2014.06.023, 2014.

Hopke, P. K.: Review of receptor modeling methods for source apportionment, J. Air Waste Manage. Assoc., 66, 237-259, doi:10.1080/10962247.2016.1140693, 2016.

Iacono, M. J., Delamere, J. S., Mlawer, E. J., Shephard, M. W., Clough, S. A., and Collins, W. D.: Radiative forcing by long-lived greenhouse gases: Calculations with the AER radiative transfer models, J. Geophys. Res.-Atmos., 113, doi:10.1029/2008jd009944, 2008.

Ikeda, K., Yamaji, K., Kanaya, Y., Taketani, F., Pan, X., Komazaki, Y., Kurokawa, J.-i., and Ohara, T.: Source region attribution of $PM_{2.5}$ mass concentrations over Japan, Geochem. J., 49, 185-194, doi:10.2343/geochemj.2.0344, 2015.

Inoue, K., Tonokura, K., and Yamada, H.: Modeling study on the spatial variation of the sensitivity of photochemical ozone concentrations and population exposure to VOC emission reductions in Japan, Air Quality Atmosphere and Health, 12, 1035-1047, doi:10.1007/s11869-019-00720-w, 2019.

Itahashi, S., Hayami, H., and Uno, I.: Comprehensive study of emission source contributions for tropospheric ozone formation over East Asia, J. Geophys. Res.-Atmos., 120, 331-358, doi:10.1002/2014jd022117, 2015.

Itahashi, S., Yamaji, K., Chatani, S., and Hayami, H.: Refinement of Modeled Aqueous-Phase Sulfate Production via the Fe- and Mn-Catalyzed Oxidation Pathway, Atmosphere, 9, doi:10.3390/atmos9040132, 2018.

Itahashi, S., Mathur, R., Hogrefe, C., and Zhang, Y.: Modeling Trans-Pacific Transport using Hemispheric CMAQ during April 2010: Part 1. Model Evaluation and Air Mass Characterization for the Estimation of Stratospheric Intrusion on Tropospheric Ozone, Atmos. Chem. Phys. Discuss., 2019, 1-43, doi:10.5194/acp-2019-203, 2019a.

Itahashi, S., Yamaji, K., Chatani, S., and Hayami, H.: Differences in Model Performance and Source Sensitivities for Sulfate Aerosol Resulting from Updates of the Aqueous- and Gas-Phase Oxidation Pathways for a Winter Pollution Episode in Tokyo, Japan, Atmosphere, 10, doi:10.3390/atmos10090544, 2019b.

Janssens-Maenhout, G., Crippa, M., Guizzardi, D., Dentener, F., Muntean, M., Pouliot, G., Keating, T., Zhang, Q., Kurokawa, J., Wankmuller, R., van der Gon, H. D., Kuenen, J. J. P., Klimont, Z., Frost, G., Darras, S., Koffi, B., and Li, M.: HTAP_v2.2: a mosaic of regional and global emission grid maps for 2008 and 2010 to study hemispheric transport of air pollution, Atmos. Chem. Phys., 15, 11411-11432, doi:10.5194/acp-15-11411-2015, 2015.

Kitayama, K., Morino, Y., Yamaji, K., and Chatani, S.: Uncertainties in O_3 concentrations simulated by CMAQ over Japan using four chemical mechanisms, Atmos. Environ., 198, 448-462, doi:10.1016/j.atmosenv.2018.11.003, 2019.

Kondo, Y., Ram, K., Takegawa, N., Sahu, L., Morino, Y., Liu, X., and Ohara, T.: Reduction of black carbon aerosols in Tokyo: Comparison of real-time observations with emission estimates, Atmos. Environ., 54, 242-249, doi:10.1016/j.atmosenv.2012.02.003, 2012.

Koo, B., Wilson, G. M., Morris, R. E., Dunker, A. M., and Yarwood, G.: Comparison of Source Apportionment and Sensitivity Analysis in a Particulate Matter Air Quality Model, Environ. Sci. Technol., 43, 6669-6675, doi:10.1021/es9008129, 2009.

Kurokawa, J., Ohara, T., Uno, I., Hayasaki, M., and Tanimoto, H.: Influence of meteorological variability on interannual variations of springtime boundary layer ozone over Japan during 1981-2005, Atmos. Chem. Phys., 9, 6287-6304, doi:10.5194/acp-9-6287-2009, 2009.

Kwok, R. H. F., Napelenok, S. L., and Baker, K. R.: Implementation and evaluation of $PM_{2.5}$ source contribution analysis in a photochemical model, Atmos. Environ., 80, 398-407, doi:10.1016/j.atmosenv.2013.08.017, 2013.

Lee, D., Lee, Y. M., Jang, K. W., Yoo, C., Kang, K. H., Lee, J. H., Jung, S. W., Park, J. M., Lee, S. B., Han, J. S., Hong, J. H., and Lee, S. J.: Korean National Emissions Inventory System and 2007 Air Pollutant Emissions, Asian J. Atmos. Environ., 5, 278-291, doi:10.5572/ajae.2011.5.4.278, 2011.

Liu, M. X., Huang, X., Song, Y., Tang, J., Cao, J. J., Zhang, X. Y., Zhang, Q., Wang, S. X., Xu, T. T., Kang, L., Cai, X. H., Zhang, H. S., Yang, F. M., Wang, H. B., Yu, J. Z., Lau, A. K. H., He, L. Y., Huang, X. F., Duan, L., Ding, A. J., Xue, L. K., Gao, J., Liu, B., and Zhu, T.: Ammonia emission control in China would mitigate haze pollution and nitrogen deposition, but worsen acid rain, Proceedings of the National Academy of Sciences of the United States of America, 116, 7760-7765, doi:10.1073/pnas.1814880116, 2019.

Martin, M., Dash, P., Ignatov, A., Banzon, V., Beggs, H., Brasnett, B., Cayula, J. F., Cummings, J., Donlon, C., Gentemann, C., Grumbine, R., Ishizaki, S., Maturi, E., Reynolds, R. W., and Roberts-Jones, J.: Group for High Resolution Sea Surface temperature (GHRSST) analysis fields inter-comparisons. Part 1: A GHRSST multi-product ensemble (GMPE), Deep-Sea Research Part Ii-Topical Studies in Oceanography, 77-80, 21-30, doi:10.1016/j.dsr2.2012.04.013, 2012.

Morino, Y., Chatani, S., Tanabe, K., Fujitani, Y., Morikawa, T., Takahashi, K., Sato, K., and Sugata, S.: Contributions of Condensable Particulate Matter to Atmospheric Organic Aerosol over Japan, Environ. Sci. Technol., 52, 8456-8466, doi:10.1021/acs.est.8b01285, 2018.

Morrison, H., Thompson, G., and Tatarskii, V.: Impact of Cloud Microphysics on the Development of Trailing Stratiform Precipitation in a Simulated Squall Line: Comparison of One- and Two-Moment Schemes, Mon. Weather Rev., 137, 991-1007, doi:10.1175/2008mwr2556.1, 2009.

National Centers for Environmental Prediction/National Weather Service/NOAA/U.S. Department of Commerce: NCEP GDAS/FNL 0.25 Degree Global Tropospheric Analyses and Forecast Grids, in, Research Data Archive at the National Center for Atmospheric Research, Computational and Information Systems Laboratory, Boulder, CO, 2015.

Pinder, R. W., Adams, P. J., and Pandis, S. N.: Ammonia emission controls as a cost-effective strategy for reducing atmospheric particulate matter in the eastern United States, Environ. Sci. Technol., 41, 380-386, doi:10.1021/es060379a, 2007.

Pio, C. A., and Lopes, D. A.: Chlorine loss from marine aerosol in a coastal atmosphere, J. Geophys. Res.-Atmos., 103, 25263-25272, doi:10.1029/98jd02088, 1998.

Prinn, R. G.: The cleansing capacity of the atmosphere, Annual Review of Environment and Resources, 28, 29-57, doi:10.1146/annurev.energy.28.011503.163425, 2003.

Ronald, J. V., Mijling, B., Ding, J., Koukouli, M. E., Liu, F., Li, Q., Mao, H. Q., and Theys, N.: Cleaning up the air: effectiveness of air quality policy for SO_2 and NO_X emissions in China, Atmos. Chem. Phys., 17, 1775-1789, doi:10.5194/acp-17-1775-2017, 2017.

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

Shimadera, H., Hayami, H., Chatani, S., Morino, Y., Mori, Y., Morikawa, T., Yamaji, K., and Ohara, T.: Sensitivity analyses of factors influencing CMAQ performance for fine particulate nitrate, J. Air Waste Manage. Assoc., 64, 374-387, doi:10.1080/10962247.2013.778919, 2014.

Shimadera, H., Hayami, H., Chatani, S., Morikawa, T., Morino, Y., Mori, Y., Yamaji, K., Nakatsuka, S., and Ohara, T.: Urban Air Quality Model Inter-Comparison Study (UMICS) for Improvement of $PM_{2.5}$ Simulation in Greater Tokyo Area of Japan, Asian J. Atmos. Environ., 12, 139-152, doi:10.5572/ajae.2018.12.2.139, 2018.

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Barker, D. M., Duda, M. G., Huang, X. Y., Wang, W., and J. G., P.: A Description of the Advanced Research WRF Version 3NCAR/TN-475+STR, 2008.

Spero, T. L., Nolte, C. G., Mallard, M. S., and Bowden, J. H.: A Maieutic Exploration of Nudging Strategies for Regional Climate Applications Using the WRF Model, J. Appl. Meteorol. Climatol., 57, 1883-1906, doi:10.1175/jamc-d-17-0360.1, 2018.

Sudo, K., Takahashi, M., Kurokawa, J., and Akimoto, H.: CHASER: A global chemical model of the troposphere - 1. Model description, J. Geophys. Res.-Atmos., 107, doi:10.1029/2001jd001113, 2002.

Sutton, M. A., Dragosits, U., Tang, Y. S., and Fowler, D.: Ammonia emissions from non-agricultural sources in the UK, Atmos. Environ., 34, 855-869, doi:10.1016/s1352-2310(99)00362-3, 2000.

Tegen, I., Hollrig, P., Chin, M., Fung, I., Jacob, D., and Penner, J.: Contribution of different aerosol species to the global aerosol extinction optical thickness: Estimates from model results, J. Geophys. Res.-Atmos., 102, 23895-23915, doi:10.1029/97jd01864, 1997.

Thunis, P., Clappier, A., Tarrason, L., Cuvelier, C., Monteiro, A., Pisoni, E., Wesseling, J., Belisa, C. A., Pirovano, G., Janssen, S., Guerreiro, C., and Peduzzi, E.: Source apportionment to support air quality planning: Strengths and weaknesses of existing approaches, Environment International, 130, doi:10.1016/j.envint.2019.05.019, 2019.

Tomiyama, H., Tanabe, K., Chatani, S., Kobayashi, S., Fujitani, Y., Furuyama, A., Sato, K., Fushimi, A., Kondo, Y., Sugata, S., Morino, Y., Hayasaki, M., Oguma, H., Ide, R., Kusaka, H., and Takami, A.: Observation for Temporal Open Burning Frequency and Estimation for Daily Emissions caused by Open Burning of Rice Residue, J. Jpn. Soc. Atmos. Environ., 52, 105-117, doi:10.11298/taiki.52.105, 2017.

Wagstrom, K. M., Pandis, S. N., Yarwood, G., Wilson, G. M., and Morris, R. E.: Development and application of a computationally efficient particulate matter apportionment algorithm in a three-dimensional chemical transport model, Atmos. Environ., 42, 5650-5659, doi:10.1016/j.atmosenv.2008.03.012, 2008.

Wakamatsu, S., Morikawa, T., and Ito, A.: Air pollution trends in Japan between 1970 and 2012 and impact of urban air pollution countermeasures, Asian J. Atmos. Environ., 7, 177-190, doi:10.5572/ajae.2013.7.4.177, 2013.

Wang, J. D., Zhao, B., Wang, S. X., Yang, F. M., Xing, J., Morawska, L., Ding, A. J., Kulmala, M., Kerminen, V. M., Kujansuu, J., Wang, Z. F., Ding, D. A., Zhang, X. Y., Wang, H. B., Tian, M., Petaja, T., Jiang, J. K., and Hao, J. M.: Particulate matter pollution over China and the effects of control policies, Science of the Total Environment, 584, 426-447, doi:10.1016/j.scitotenv.2017.01.027, 2017.

Whitten, G. Z., Heo, G., Kimura, Y., McDonald-Buller, E., Allen, D. T., Carter, W. P. L., and Yarwood, G.: A new condensed toluene mechanism for Carbon Bond CB05-TU, Atmos. Environ., 44, 5346-5355, doi:10.1016/j.atmosenv.2009.12.029, 2010.

Wu, Y. Y., Gu, B. J., Erisman, J. W., Reis, S., Fang, Y. Y., Lu, X. H., and Zhang, X. M.: $PM_{2.5}$ pollution is substantially affected by ammonia emissions in China, Environ. Pollut., 218, 86-94, doi:10.1016/j.envpol.2016.08.027, 2016.

Xing, J., Wang, S. X., Jang, C., Zhu, Y., and Hao, J. M.: Nonlinear response of ozone to precursor emission changes in China: a modeling study using response surface methodology, Atmos. Chem. Phys., 11, 5027-5044, doi:10.5194/acp-11-5027-2011, 2011.

Yamaji, K., Chatani, S., Itahashi, S., Saito, M., Takigawa, M., Morikawa, T., Kanda, I., Miya, Y., Komatsu, H., Sakurai, T., Morino, Y., Kitayama, K., Nagashima, T., Shimadera, H., Uranishi, K., Fujiwara, Y., Hashimoto, T., Sudo, K., Misaki, T., and Hayami, H.: Model Inter-Comparison for PM2.5 Components over urban Areas in Japan in the J-STREAM Framework, Atmosphere, 11, 222, 2020.

Yang, Y. J., Wilkinson, J. G., and Russell, A. G.: Fast, direct sensitivity analysis of multidimensional photochemical models, Environ. Sci. Technol., 31, 2859-2868, doi:10.1021/es970117w, 1997.

Zheng, B., Tong, D., Li, M., Liu, F., Hong, C. P., Geng, G. N., Li, H. Y., Li, X., Peng, L. Q., Qi, J., Yan, L., Zhang, Y. X., Zhao, H. Y., Zheng, Y. X., He, K. B., and Zhang, Q.: Trends in China's anthropogenic emissions since 2010 as the consequence of clean air actions, Atmos. Chem. Phys., 18, 14095-14111, doi:10.5194/acp-18-14095-2018, 2018.

810

Table 1. Emission source groups to which~~whose~~ sensitivities and apportionments were evaluated in this study.

Group	Included emission sources
s01	On-road vehicles
s02	Ships
s03	Non-road transport (machineries, railways, and airplanes)
s04	Stationary combustion (power plants, industries, and commercial)
s05	Biomass combustion (smoking, cooking, and agricultural residue burning)
s06	Residential combustion
s07	Fugitive volatile organic compounds
s08	Agriculture (except for agricultural residue burning) and fugitive ammonia
s09	Natural (volcanoes, biogenic, and soil)
s10	Anthropogenic sources in other countries in d02
s11	Transport through boundaries of d02
s12	Sea salt

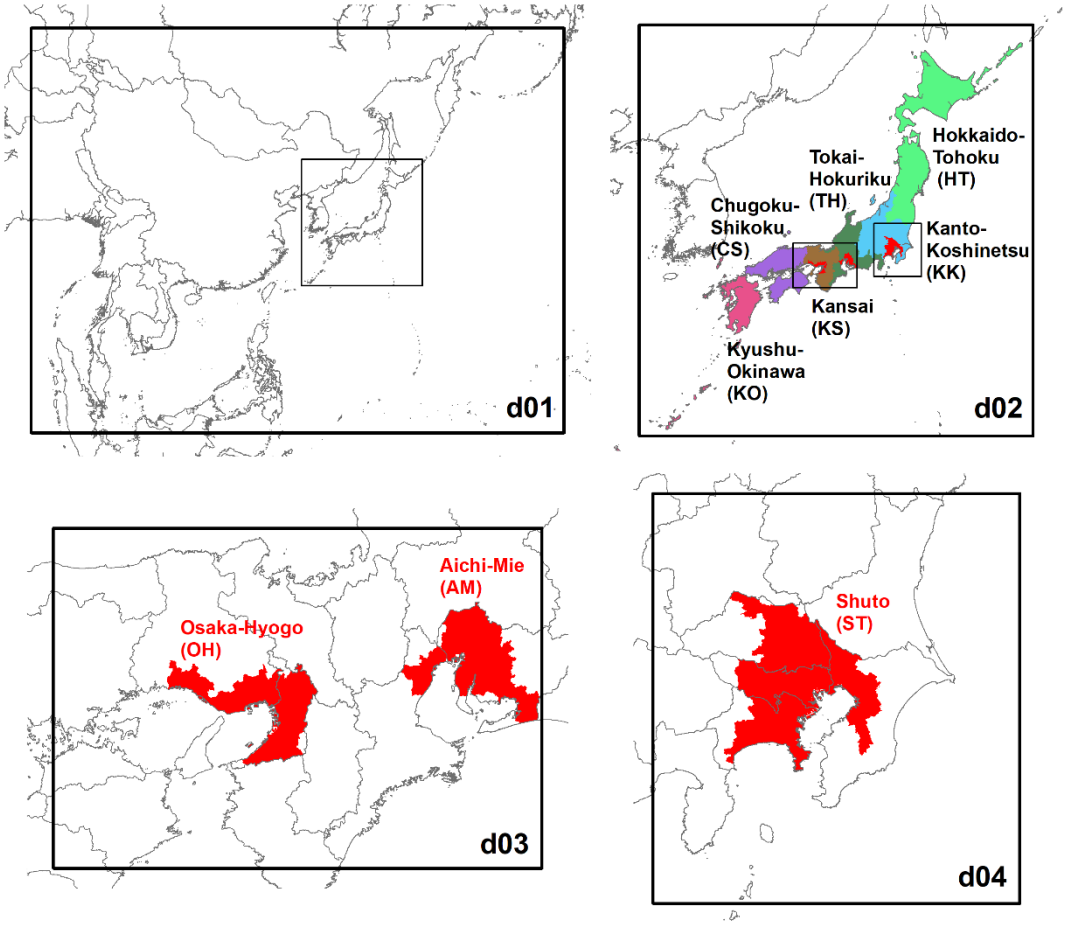


Figure 1. Target domains for the simulations in this study. Results are summarized for six colour coded regions in d02 and three designated areas shown in red in d02, d03, and d04. Their abbreviations are shown in parentheses.

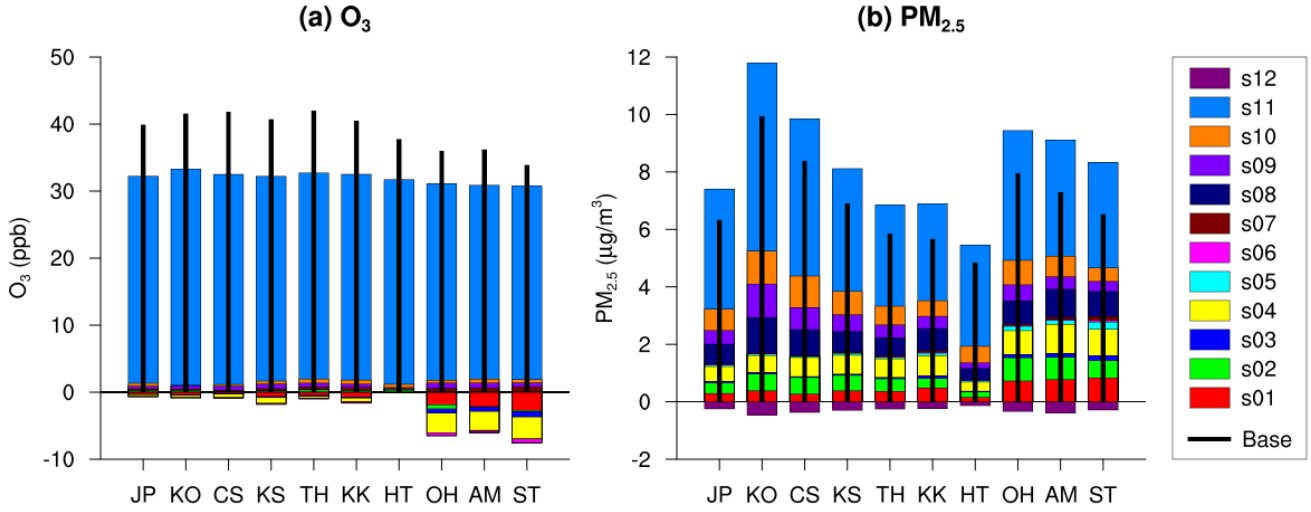


Figure 2. Source sensitivities ~~to~~ of the annual mean ozone and $PM_{2.5}$ concentrations derived by BFM in the regions. Thick black lines represent the simulated concentrations.

825

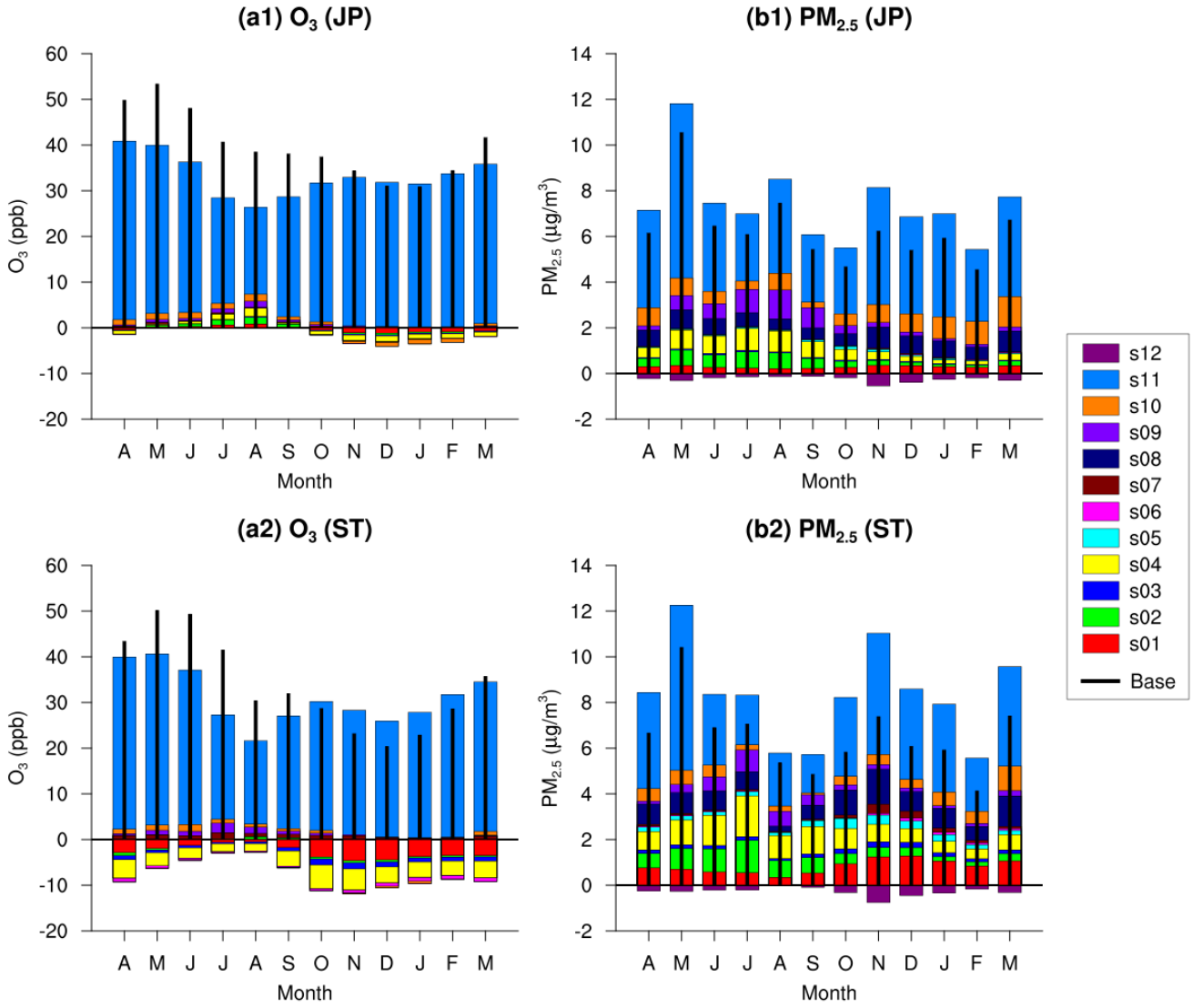


Figure 3. Source sensitivities ~~to~~ of the monthly mean ozone and $PM_{2.5}$ concentrations derived by BFM in entire Japan (JP) and ST. Thick black lines represent the simulated concentrations.

830

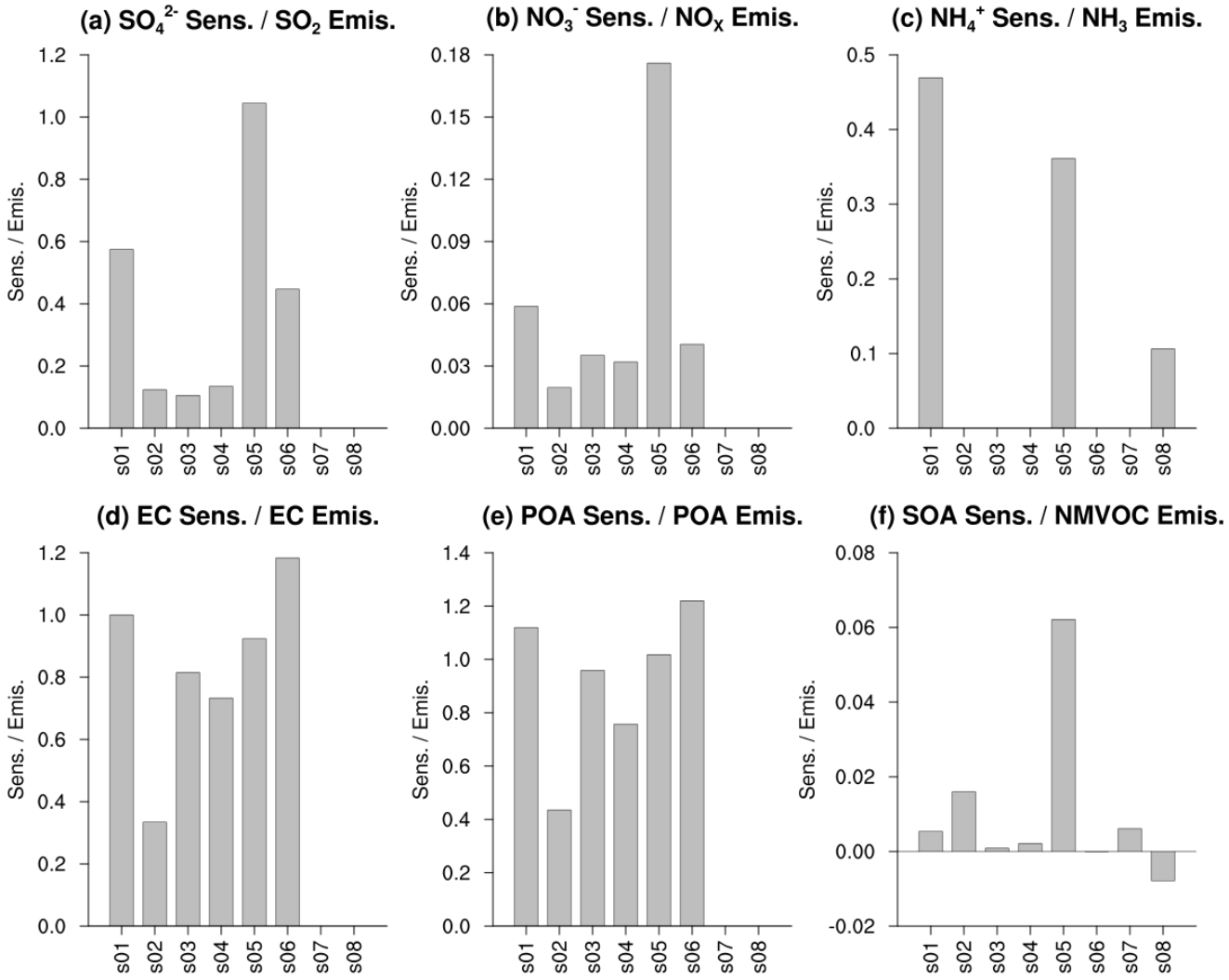


Figure 4. Sensitivities of the annual mean ambient concentrations of $PM_{2.5}$ components in d02 per total annual amounts of corresponding precursor emissions from domestic anthropogenic sources (s01–s08) ~~in d02 to the annual mean ambient concentrations of corresponding $PM_{2.5}$ components~~ in entire Japan. All of them are normalized by the EC value for s01.

835

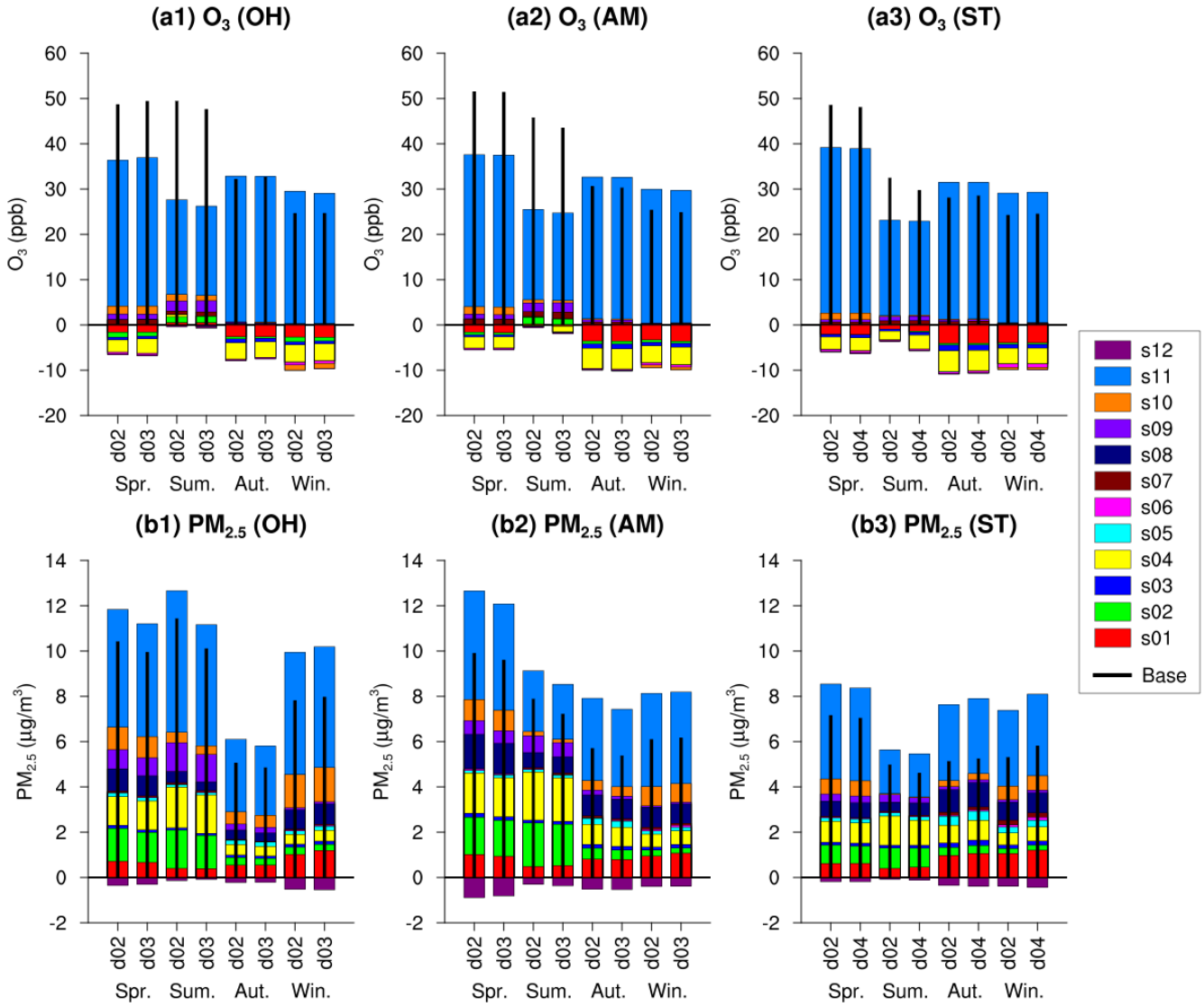
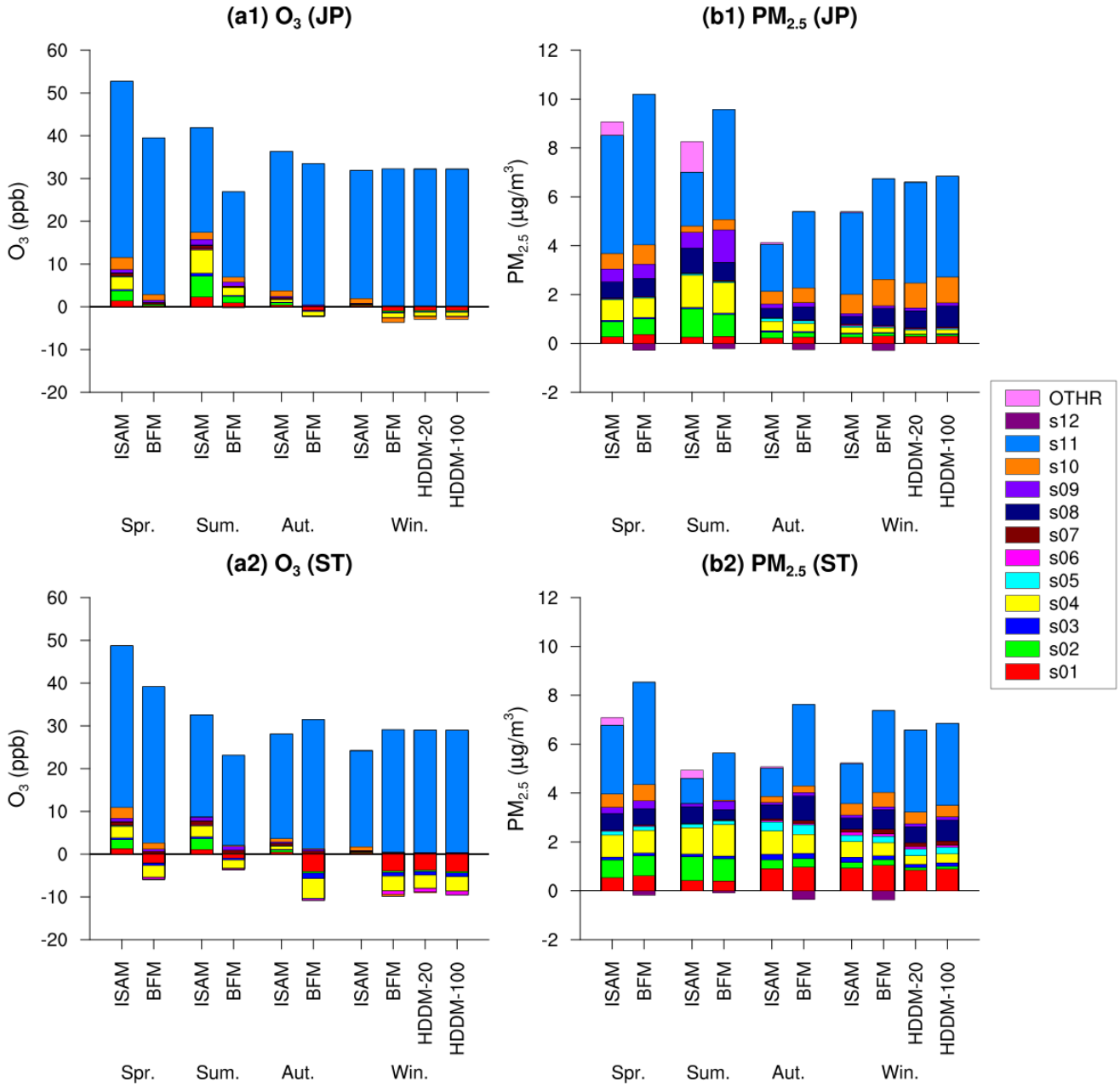
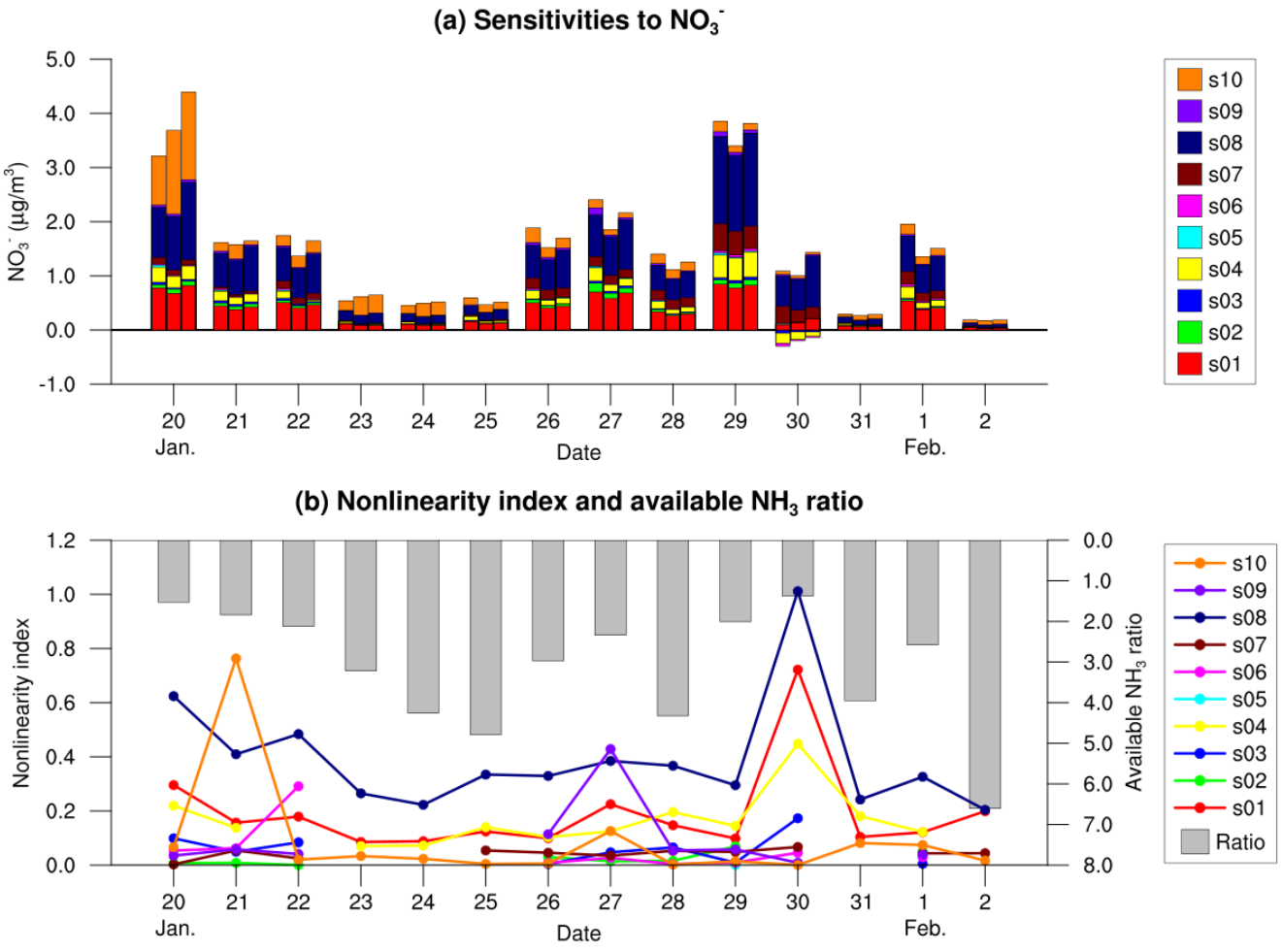


Figure 5. Sensitivities of the simulated ozone and $PM_{2.5}$ concentrations to all source groups over OH, AM, and ST evaluated in d02, d03, and d04 for the two target weeks in the four seasons.



840

Figure 6. Apportionments derived by ISAM and sensitivities derived by BFM and HDDM of ~~all source groups to~~ the simulated ozone and $PM_{2.5}$ concentrations to all source groups in JP and ST for the two target weeks in the four seasons.


(a) Sensitivities to NO_3^-
NO_3^- (μg/m^3)
5.0
4.0
3.0
2.0
1.0
0.0
-1.0
20
Jan.
21
22
23
24
25
26
27
28
29
30
31
1
Feb.
2
Date
s10
s09
s08
s07
s06
s05
s04
s03
s02
s01
(b) Nonlinearity index and available NH_3 ratio
Nonlinearity index
1.2
1.0
0.8
0.6
0.4
0.2
0.0
Available NH_3 ratio
0.0
1.0
2.0
3.0
4.0
5.0
6.0
7.0
8.0
Ratio

845

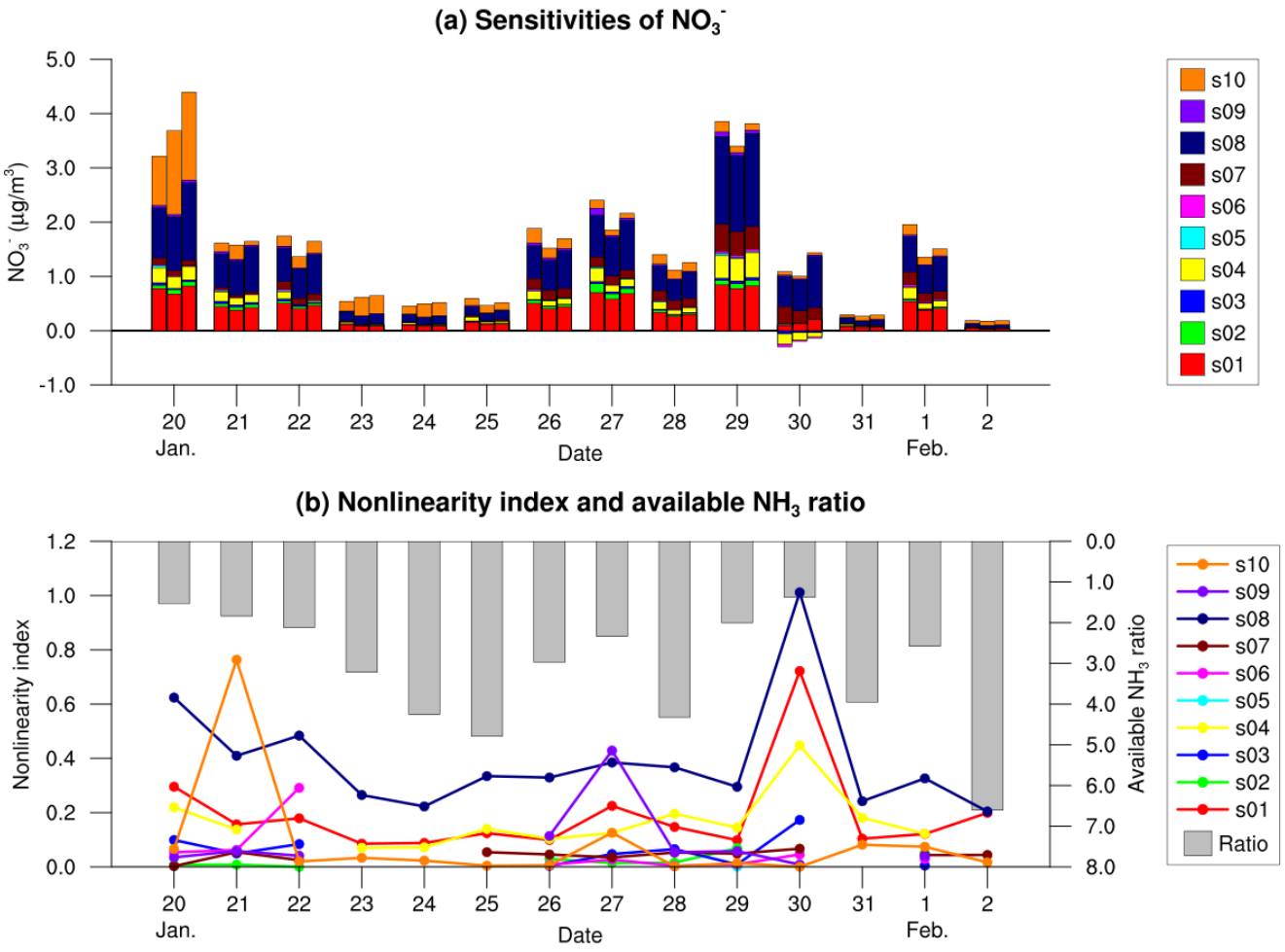


Figure 7: (a) Sensitivities of the daily NO_3^- concentrations to the source groups located within d02 (s01–s10) derived by BFM (left), HDDM-20 (middle), and HDDM-100 (right) ~~to the daily NO_3^- concentrations~~ and (b) daily nonlinearity index and available NH_3 ratios for the two target weeks in winter in ST. Nonlinearity indices for first-order sensitivity coefficients less than 0.001 μg/m^3 are not shown as they are likely to be affected by numerical noise.

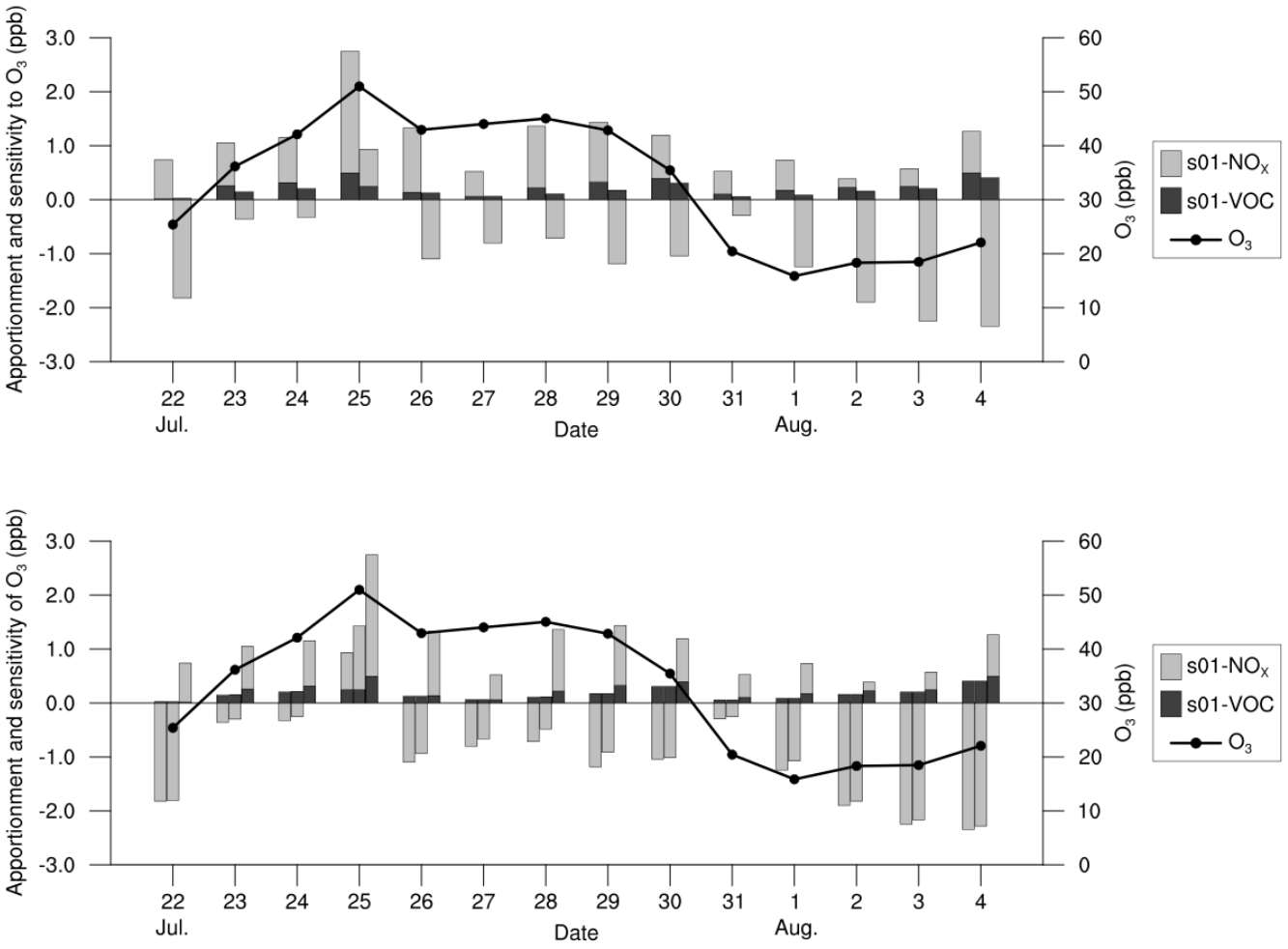


Figure 8. Sensitivities derived by BFM-20 (left) and BFM-100 (middle), and apportionments derived by ISAM (~~left) and sensitivities (~~right) of daily ozone concentrations (shown by a line with markers) ~~for~~ to the NO_X and VOC emissions of s01 ~~derived by ISAM and BFM to daily ozone concentrations (shown by a line with markers)~~ for the two target weeks during the summer in ST.

Comprehensive analyses of source sensitivities ~~to~~ and apportionments of $PM_{2.5}$ and ozone over Japan via multiple numerical techniques

Satoru Chatani[1], Hikari Shimadera[2], Syuichi Itahashi[3], Kazuyo Yamaji[4]

[1]National Institute for Environmental Studies, Tsukuba, Ibaraki 305-8506, Japan
[2]Osaka University, Suita, Osaka 565-0871, Japan
[3]Central Research Institute of Electric Power Industry, Abiko, Chiba 270-1194, Japan
[4]Kobe University, Kobe, Hyogo 658-0022, Japan

Correspondence to: Satoru Chatani (chatani.satoru@nies.go.jp)

Supplementary material

Table S1: Annual total emission amounts (Mg/year) of each source group for the 2016 fiscal year in d02.

Group	CO	SO_2	NO_X	NH_3	NMVOC[*1]	$PM_{2.5}$	EC[*2]	OC[*3]
s01	1,171,555	910	411,047	15,576	122,712	23,850	6,091	5,361
s02	59,862	279,102	682,708	0	20,593	55,988	10,766	16,045
s03	164,110	3,057	97,754	0	11,404	4,479	2,503	1,421
s04	1,230,684	356,649	761,388	0	226,562	19,196	1,661	3,459
s05	69,836	369	4,924	943	8,016	10,028	586	6,997
s06	46,034	840	33,421	0	11,938	1,770	190	619
s07	0	0	0	0	587,827	0	0	0
s08	0	0	0	389,915	55,714	1,811	0	0
s09[*4]	404,322	1,534,513	58,700	21,732	3,954,064	0	0	0
	132,359	1,534,513	9,940	21,732	1,302,798	0	0	0
s10	13,152,939	1,168,210	2,990,211	649,985	2,108,940	736,968	112,112	241,057

[*1] Non-methane volatile organic compound
[*2] Elemental carbon
[*3] Organic carbon
[*4] Lower values indicate amounts within Japan only

Table S2: Statistics of the model performance on the MDA8O3 and daily mean $PM_{2.5}$ concentrations for the entire 2016 fiscal year 2016 in the regions.

Species	Region	Number[*1]	Obs.[*2]	Sim.[*3]	NMB[*4]	NME[*5]	R[*6]
MDA8O3 (ppb)	JP	1150	42.7	46.0	7.79%	20.0%	0.860
	KO	151	43.4	48.4	11.3%	24.0%	0.820
	CS	143	44.4	46.8	5.27%	20.7%	0.834
	KS	176	43.1	46.2	7.14%	20.0%	0.868
	TH	201	43.6	47.4	8.69%	18.9%	0.869
	KK	369	41.5	44.3	6.94%	19.5%	0.872
	HT	110	39.4	44.1	11.7%	21.2%	0.797
	OH	108	42.6	45.2	6.10%	20.7%	0.864
	AM	75	42.6	45.6	6.89%	19.3%	0.874
	ST	196	40.8	42.9	5.21%	19.7%	0.880
$PM_{2.5}$ ($\mu g/m^3$)	JP	820	11.9	7.62	-35.9%	41.6%	0.852
	KO	127	14.2	10.3	-27.2%	36.6%	0.860
	CS	113	13.6	9.26	-30.3%	38.9%	0.853
	KS	134	12.0	7.77	-35.5%	39.5%	0.862
	TH	132	10.7	6.62	-38.2%	42.0%	0.855
	KK	243	11.3	6.48	-42.8%	46.2%	0.836
	HT	71	9.01	5.41	-39.9%	46.0%	0.827
	OH	71	12.5	8.30	-33.8%	38.2%	0.863
	AM	43	11.6	7.40	-36.3%	40.0%	0.855
	ST	156	12.0	6.68	-44.5%	46.8%	0.839
SO_4^{2-} ($\mu g/m^3$)	JP	154	2.73	2.64	-3.29%	40.9%	0.710
NO_3^- ($\mu g/m^3$)	JP	154	0.641	1.01	57.1%	121%	0.441
NH_4^+ ($\mu g/m^3$)	JP	154	1.11	1.08	-3.07%	41.1%	0.704
EC ($\mu g/m^3$)	JP	136	0.757	0.345	-54.4%	58.3%	0.477
OC ($\mu g/m^3$)	JP	151	2.58	0.958	-62.8%	66.0%	0.487

[*1] Number of monitoring stations
[*2] Observed values
[*3] Simulated values
[*4] Normalized Mean Bias
[*5] Normalized Mean Error
[*6] Correlation coefficient

Table S3: Source sensitivities ~~to~~ of the annual mean ozone and $PM_{2.5}$ concentrations simulated in the regions. The upper table shows ratios (%) against the simulated concentrations. The lower table shows their normalized ratios (%).

(a1) O_3 (not normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	-0.8	-1.0	-0.5	-1.6	-1.1	-1.7	-0.1	-5.2	-5.9	-8.0
s02	0.6	0.7	-0.1	0.5	0.8	0.7	0.8	-1.7	-0.3	-0.5
s03	-0.2	-0.1	-0.1	-0.4	-0.3	-0.5	0.0	-1.7	-1.7	-2.5
s04	-0.6	-0.7	-1.2	-2.0	-0.6	-1.2	0.4	-8.3	-7.8	-9.6
s05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
s06	-0.2	-0.1	-0.1	-0.3	-0.2	-0.3	-0.1	-1.0	-0.8	-1.6
s07	0.6	0.5	0.7	0.9	1.0	1.1	0.2	1.5	1.8	2.3
s08	0.0	-0.1	0.0	0.0	0.0	-0.1	0.0	0.0	0.0	-0.1
s09	1.0	1.2	1.5	1.6	1.4	1.2	0.4	2.3	2.0	1.9
s10	1.1	0.2	0.6	1.1	1.4	1.5	1.3	1.2	1.5	1.5
s11	77.5	77.7	75.1	75.1	73.3	75.9	81.0	81.7	80.0	85.3
s12	-0.2	-0.2	-0.2	-0.2	-0.2	-0.2	-0.1	-0.3	-0.3	-0.3
Sum	79.0	78.0	75.6	74.6	75.4	76.3	83.8	68.4	68.5	68.4

(a2) O_3 (normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	-1.0	-1.3	-0.7	-2.1	-1.5	-2.3	-0.1	-7.6	-8.6	-11.7
s02	0.8	0.9	-0.1	0.7	1.1	0.9	1.0	-2.5	-0.4	-0.8
s03	-0.2	-0.2	-0.1	-0.6	-0.4	-0.6	0.0	-2.5	-2.5	-3.6
s04	-0.7	-0.9	-1.6	-2.7	-0.9	-1.6	0.4	-12.1	-11.4	-14.0
s05	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
s06	-0.2	-0.1	-0.1	-0.4	-0.2	-0.4	-0.1	-1.5	-1.1	-2.4
s07	0.8	0.6	0.9	1.2	1.3	1.4	0.2	2.2	2.6	3.3
s08	0.0	-0.1	0.0	0.0	0.0	-0.1	0.0	0.0	0.0	-0.1
s09	1.3	1.5	2.0	2.1	1.8	1.6	0.5	3.4	2.9	2.7
s10	1.4	0.3	0.8	1.4	1.9	1.9	1.5	1.7	2.2	2.2
s11	98.2	99.6	99.3	100.6	97.1	99.5	96.6	119.4	116.9	124.7
s12	-0.2	-0.3	-0.3	-0.3	-0.3	-0.3	-0.2	-0.4	-0.4	-0.4

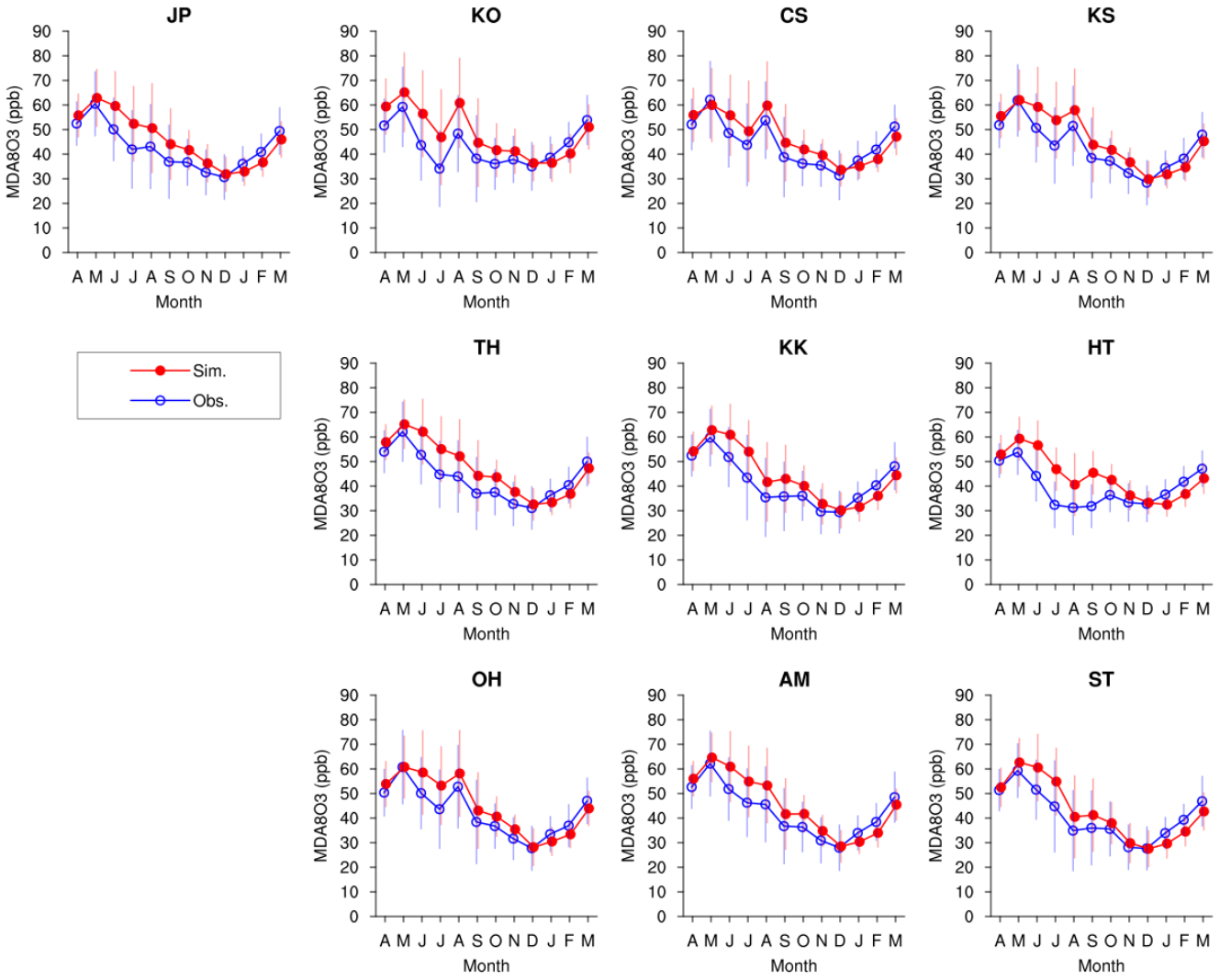
Table S3: ***Cont'd.***

(b1) $PM_{2.5}$ (not normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	4.6	3.8	3.2	5.5	6.1	8.3	3.1	9.1	10.6	12.6
s02	5.9	5.9	6.9	7.8	7.5	6.2	4.0	10.1	10.7	9.4
s03	0.7	0.5	0.4	0.8	1.0	1.5	0.5	1.5	1.8	2.6
s04	8.2	6.0	8.0	9.4	11.0	12.2	6.6	10.5	13.9	14.2
s05	0.9	0.6	0.4	1.0	1.0	1.7	1.0	2.0	2.0	3.8
s06	0.3	0.2	0.1	0.2	0.3	0.6	0.4	0.3	0.5	0.8
s07	0.3	0.1	0.3	0.4	0.5	0.9	0.1	0.7	1.0	2.0
s08	10.8	12.5	10.7	10.4	10.6	13.6	8.5	10.0	13.3	13.5
s09	7.6	11.7	9.1	8.4	7.9	7.6	3.9	7.1	6.1	5.4
s10	11.6	11.6	13.2	11.9	11.0	9.6	11.8	10.7	9.6	7.2
s11	66.2	65.9	65.4	61.8	60.4	59.7	73.1	56.8	55.6	56.4
s12	-3.9	-4.7	-4.4	-4.4	-4.3	-4.2	-2.8	-4.3	-5.4	-4.4
Sum	113.2	114.0	113.3	113.4	113.0	117.6	110.3	114.5	119.6	123.4

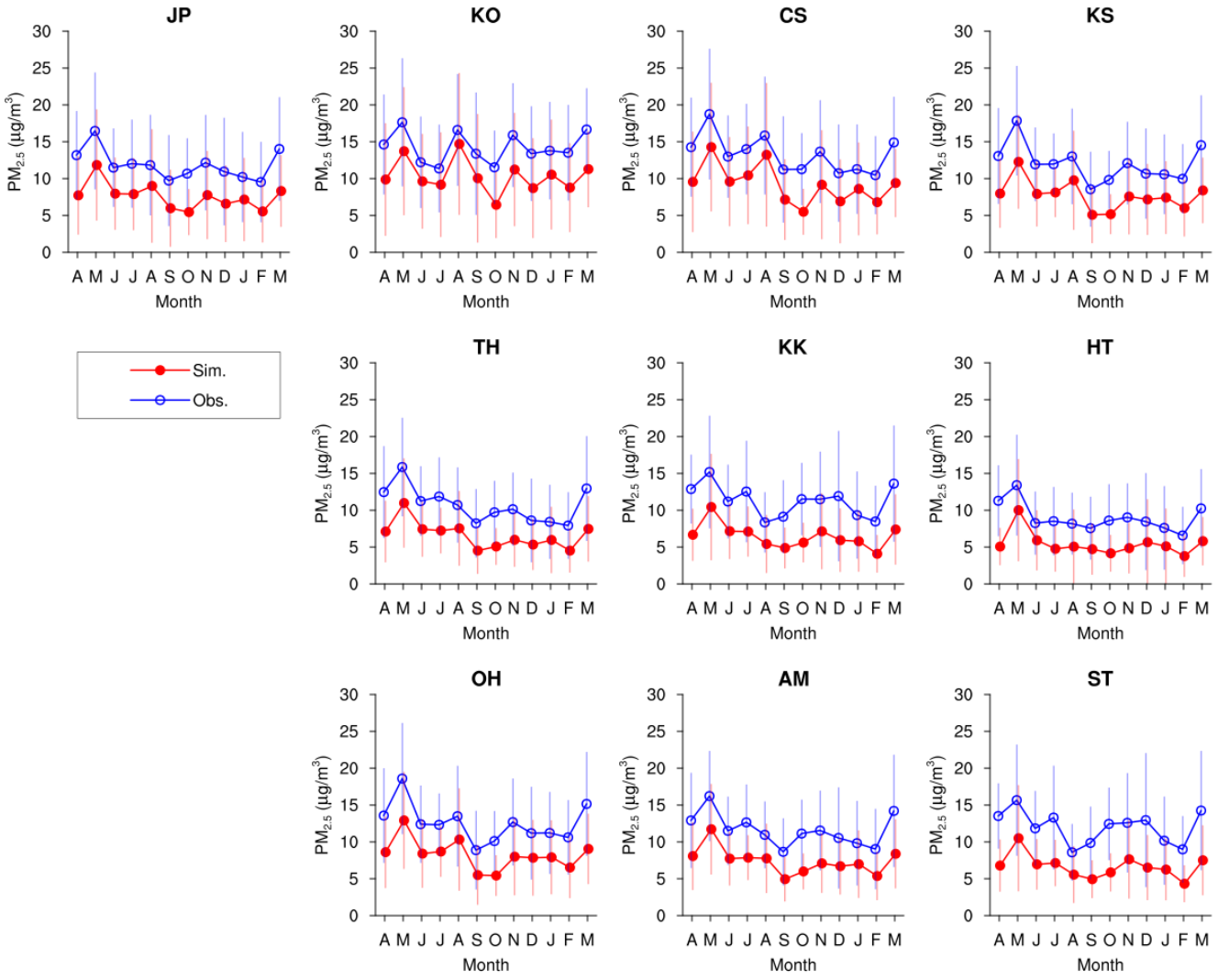
(b2) $PM_{2.5}$ (normalized)

Group	JP	KO	CS	KS	TH	KK	HT	OH	AM	ST
s01	4.0	3.4	2.8	4.9	5.4	7.1	2.8	8.0	8.8	10.2
s02	5.2	5.2	6.1	6.9	6.6	5.2	3.6	8.8	8.9	7.6
s03	0.6	0.4	0.3	0.7	0.9	1.3	0.4	1.3	1.5	2.1
s04	7.3	5.2	7.0	8.3	9.7	10.4	6.0	9.1	11.6	11.5
s05	0.8	0.5	0.4	0.9	0.9	1.4	0.9	1.7	1.7	3.1
s06	0.3	0.1	0.1	0.2	0.3	0.5	0.4	0.2	0.4	0.7
s07	0.3	0.1	0.2	0.4	0.5	0.8	0.1	0.6	0.8	1.6
s08	9.5	11.0	9.5	9.2	9.4	11.6	7.7	8.8	11.1	10.9
s09	6.7	10.2	8.0	7.4	7.0	6.4	3.6	6.2	5.1	4.4
s10	10.3	10.2	11.7	10.5	9.7	8.1	10.7	9.4	8.1	5.9
s11	58.5	57.8	57.7	54.5	53.4	50.8	66.3	49.7	46.5	45.7
s12	-3.5	-4.1	-3.9	-3.8	-3.8	-3.6	-2.5	-3.7	-4.5	-3.6



(a) MDA8O3

Figure S1: Comparisons of the observed and simulated monthly mean MDA8O3 and $PM_{2.5}$ concentrations at all stations in the regions. Markers and error bars represent mean values and standard deviations, respectively, of the daily concentrations at all monitoring stations for each month.



(b) $PM_{2.5}$

Figure S1: ***Cont'd***

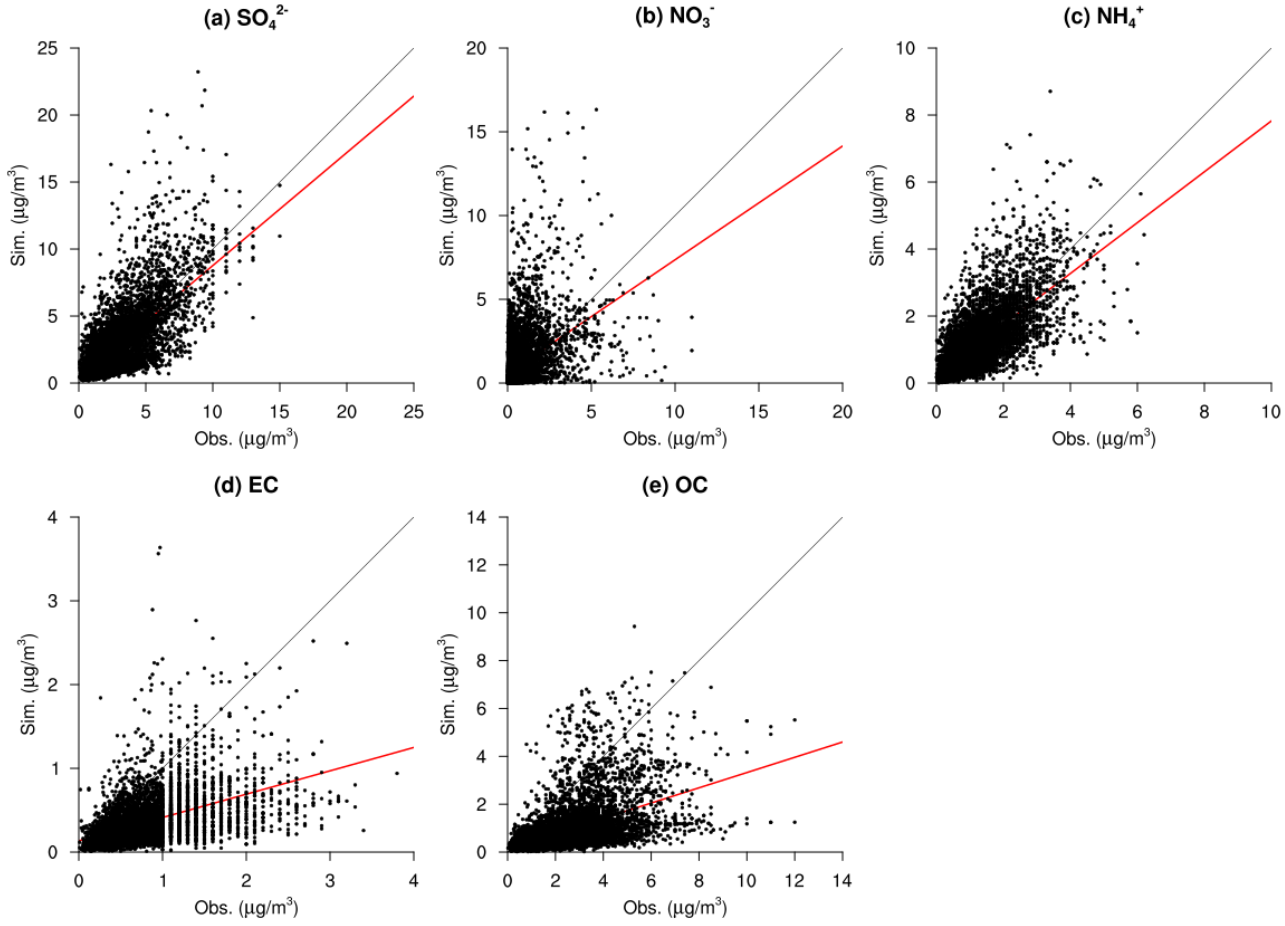


Figure S2: Scatter plots of the observed and simulated concentrations of $PM_{2.5}$ components during the monitoring campaigns of ambient concentrations of $PM_{2.5}$ components at all the locations throughout Japan in all four seasons. Regression lines are represented by red lines.

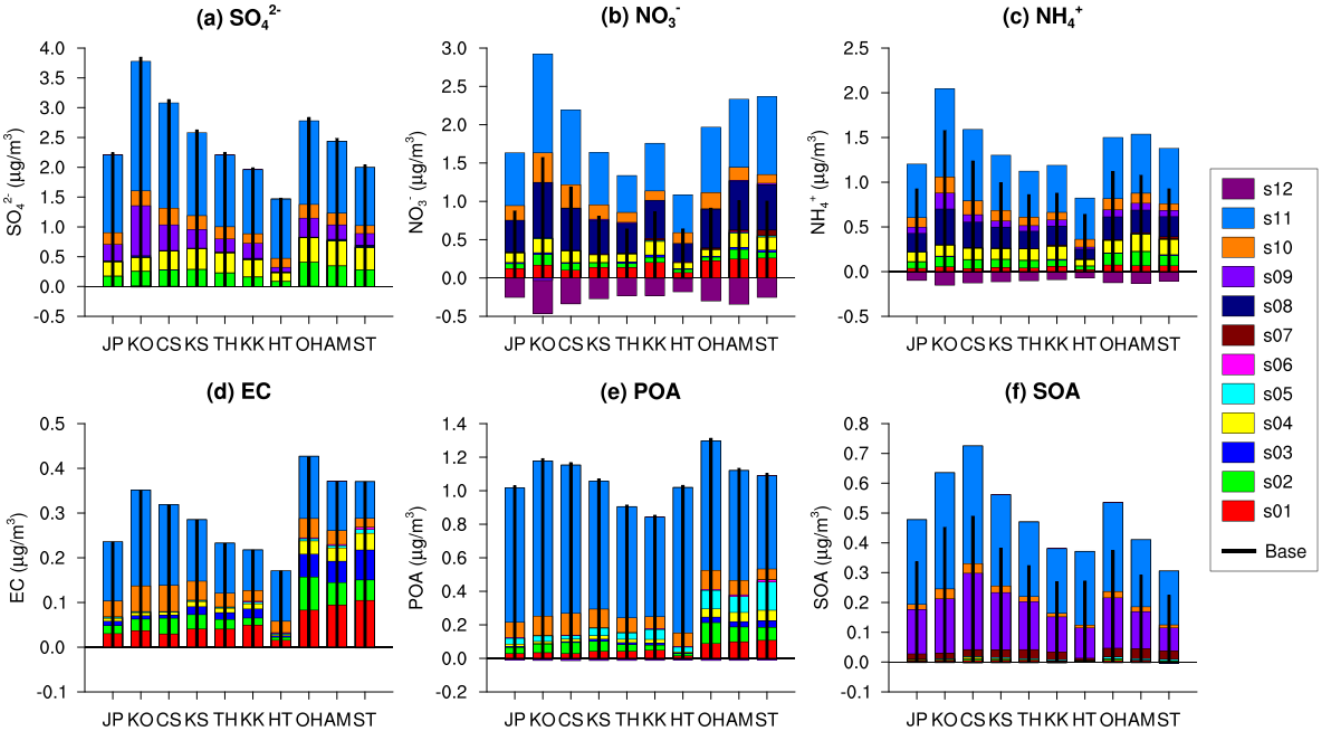


Figure S3: Source sensitivities ~~to~~ of the annual mean concentrations of $PM_{2.5}$ components derived by BFM in the regions. Thick black lines represent the simulated concentrations.

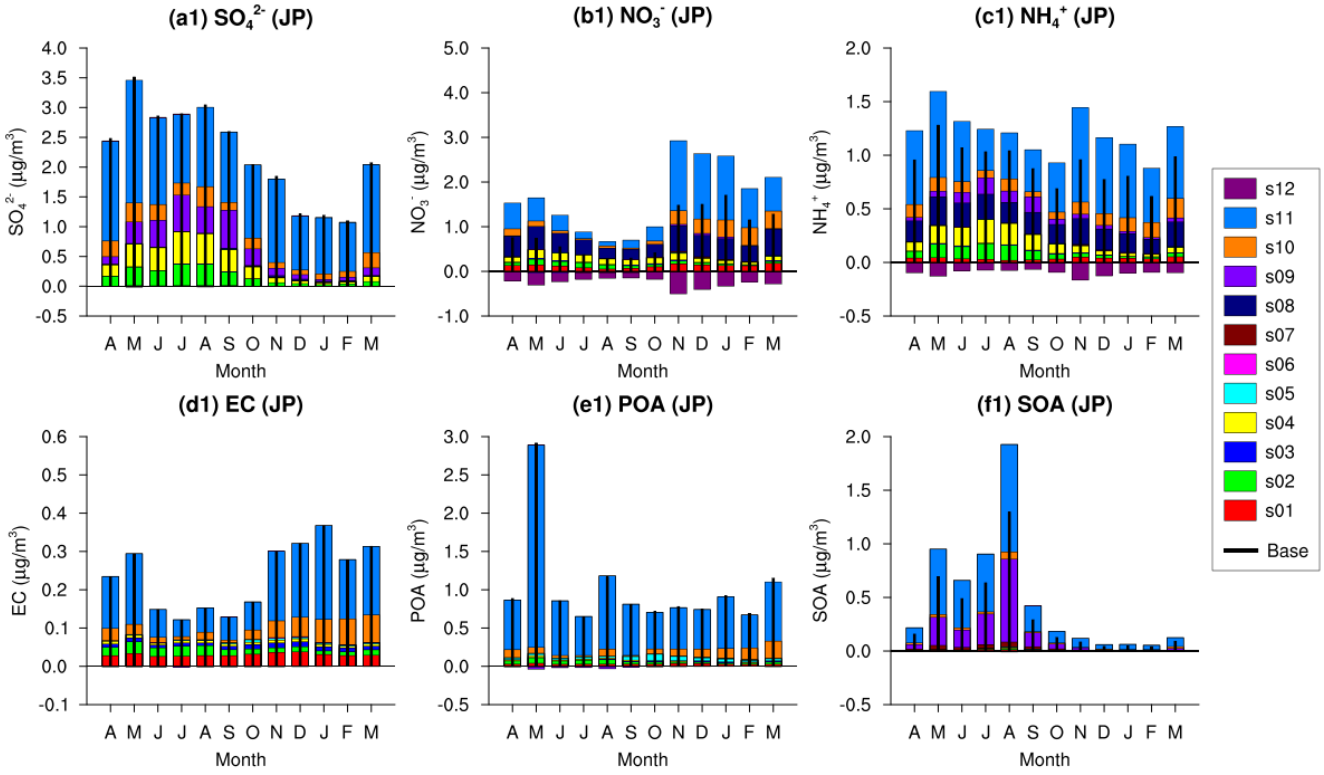


Figure S4: Source sensitivities ~~to~~ of the monthly mean concentrations of $PM_{2.5}$ components derived by BFM in entire Japan (JP) and ST. Thick black lines represent the simulated concentrations.

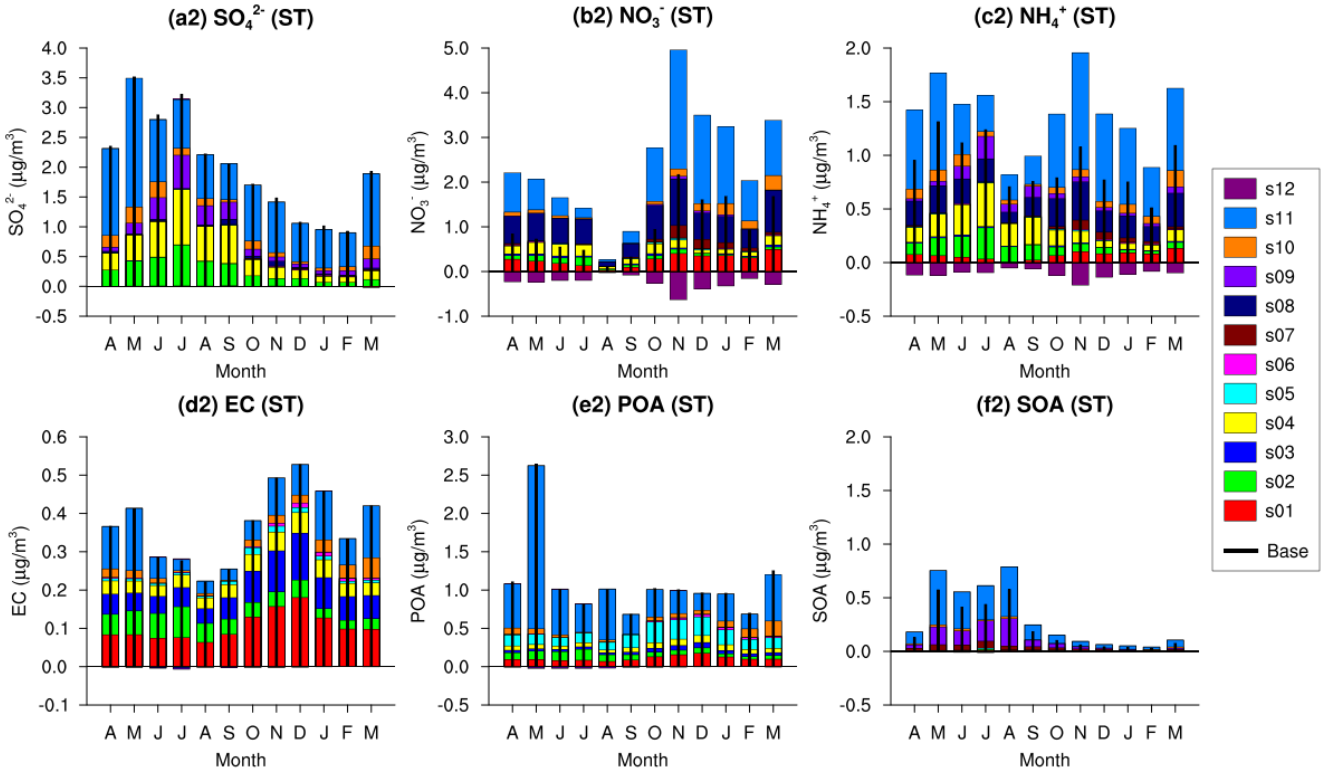


Figure S4: ***Cont'd***

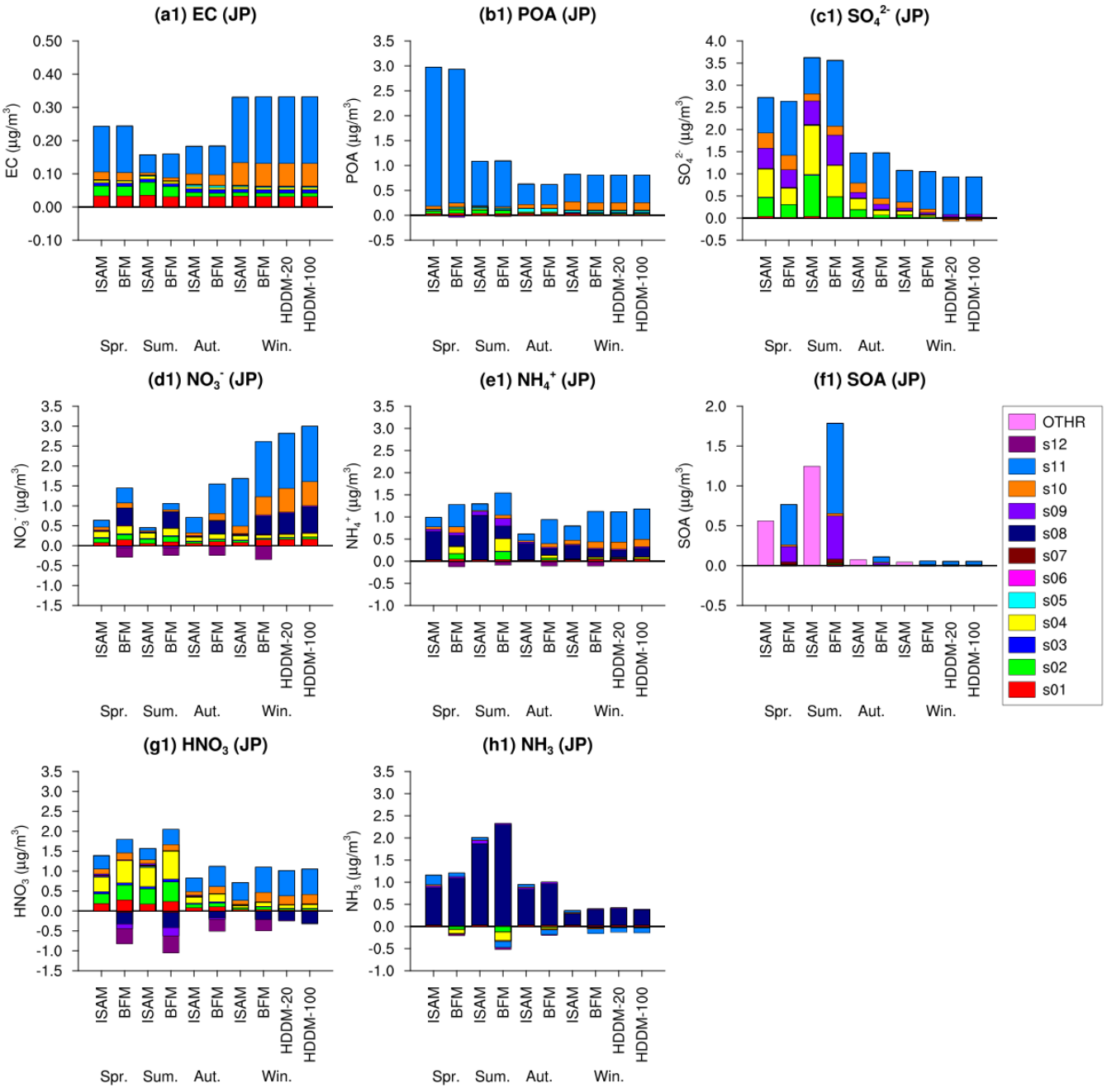


Figure S5: Apportionments derived by ISAM and sensitivities derived by BFM and HDDM of ~~all source groups to~~ the simulated concentrations of $PM_{2.5}$ components to all source groups in JP and ST for the two target weeks in the four seasons.

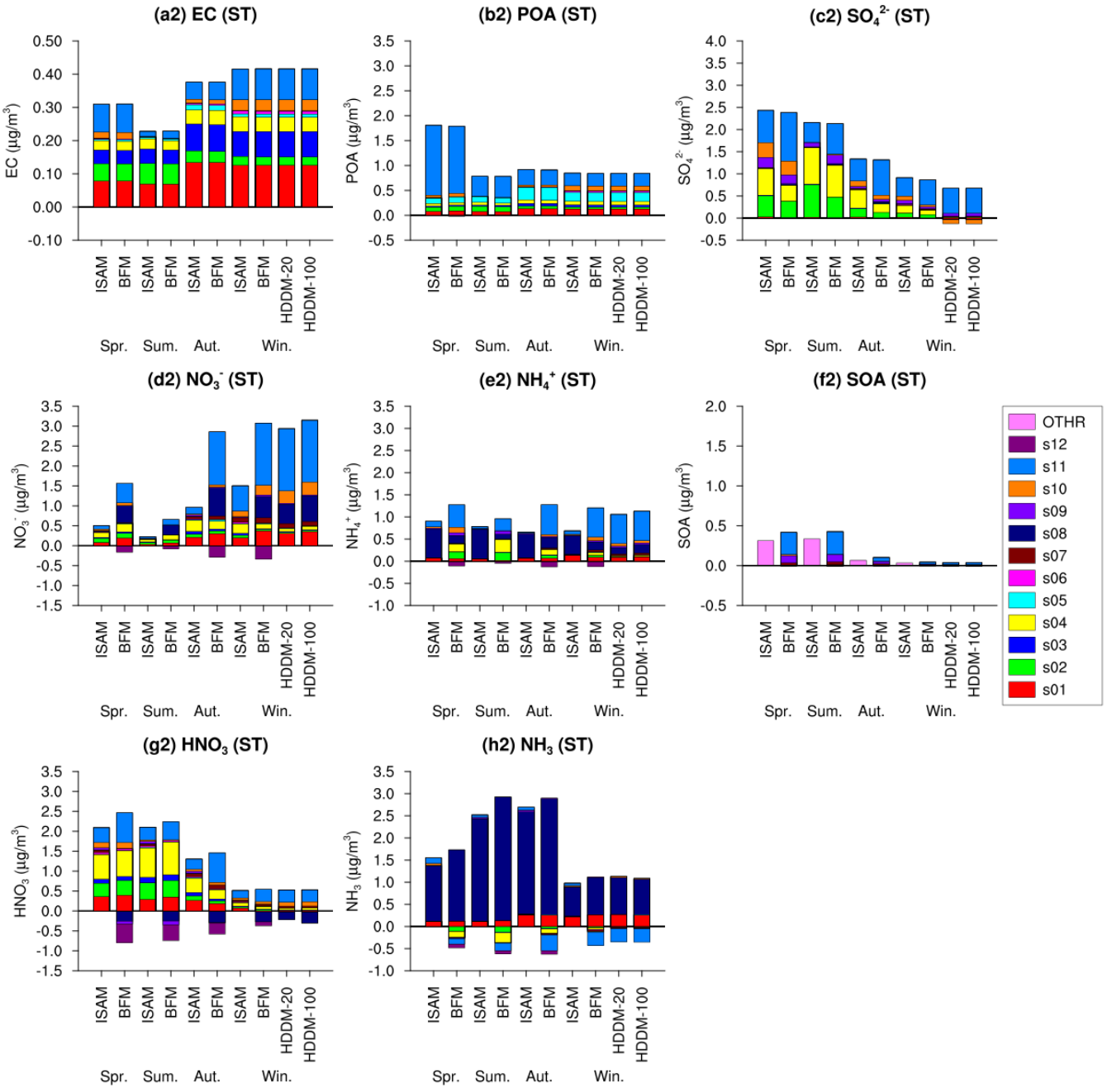


Figure S5: ***Cont'd***

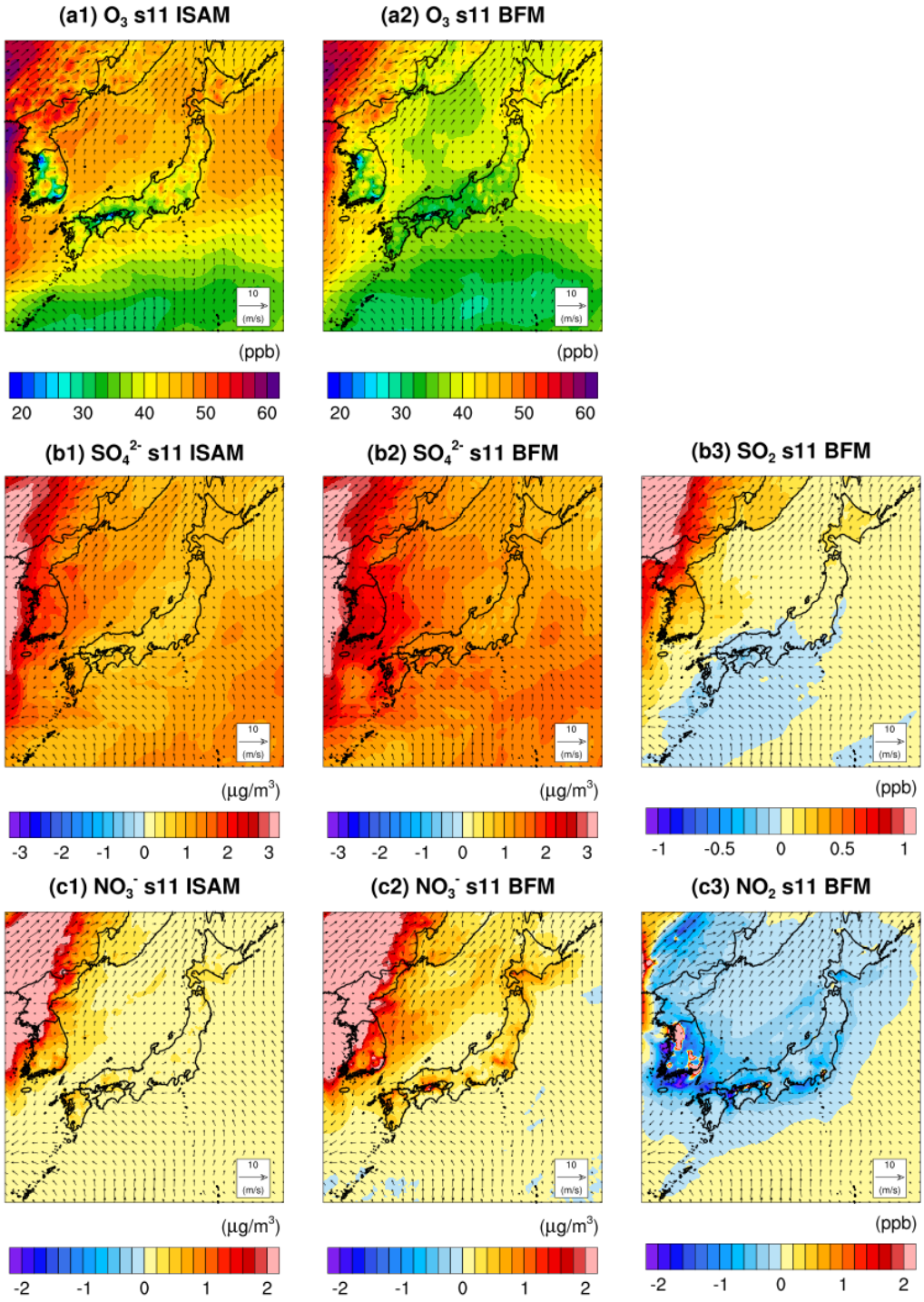


Figure S6: Horizontal distributions of the apportionments and the sensitivities ~~of~~ to s11 for the target two weeks of the spring.

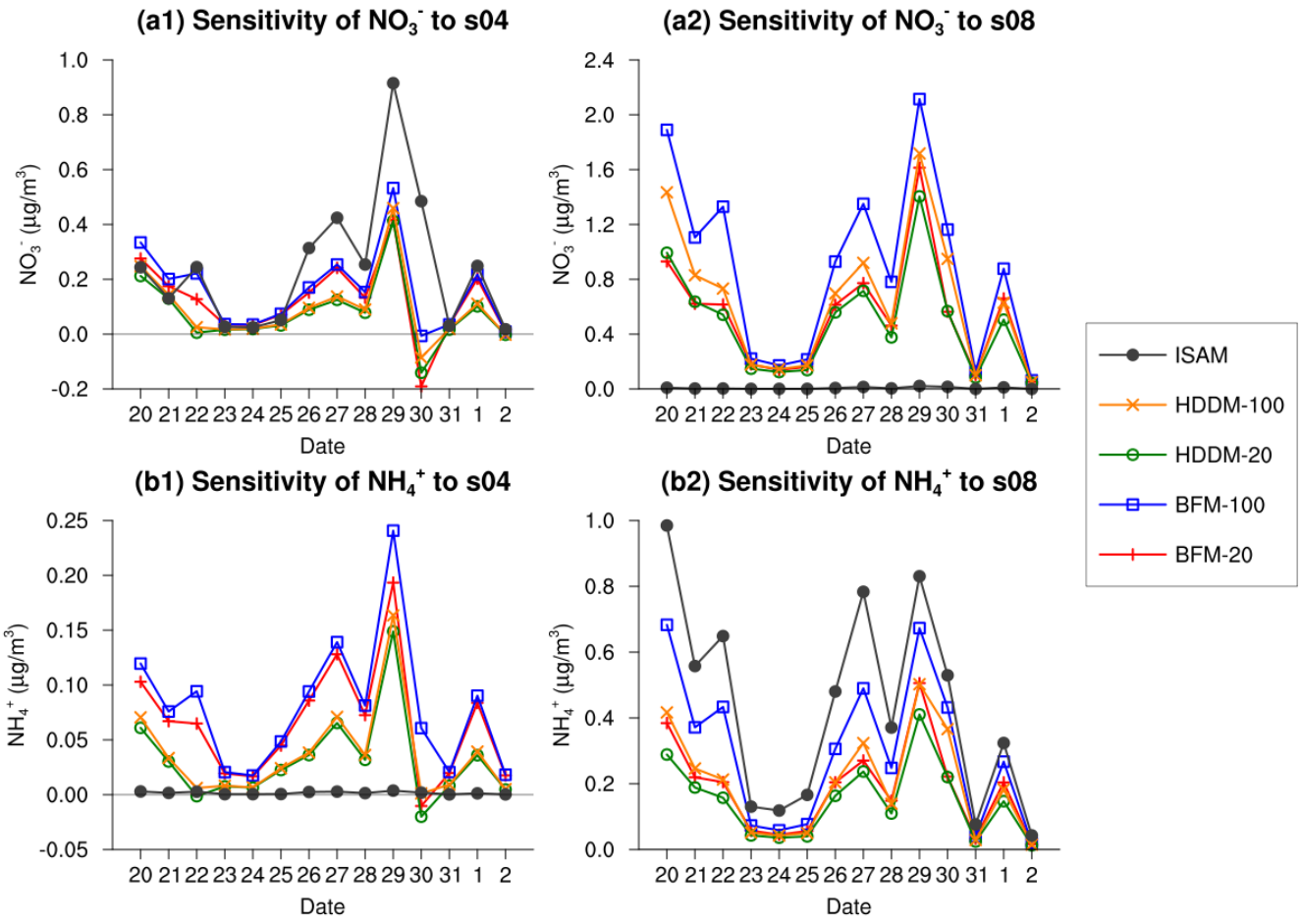


Figure S7: Sensitivities derived by BFM-20, BFM-100, HDDM-20, and HDDM-100, and apportionments derived by ISAM of the daily NO_3^- and NH_4^+ concentrations to s04 and s08 for the two target weeks in winter in ST.

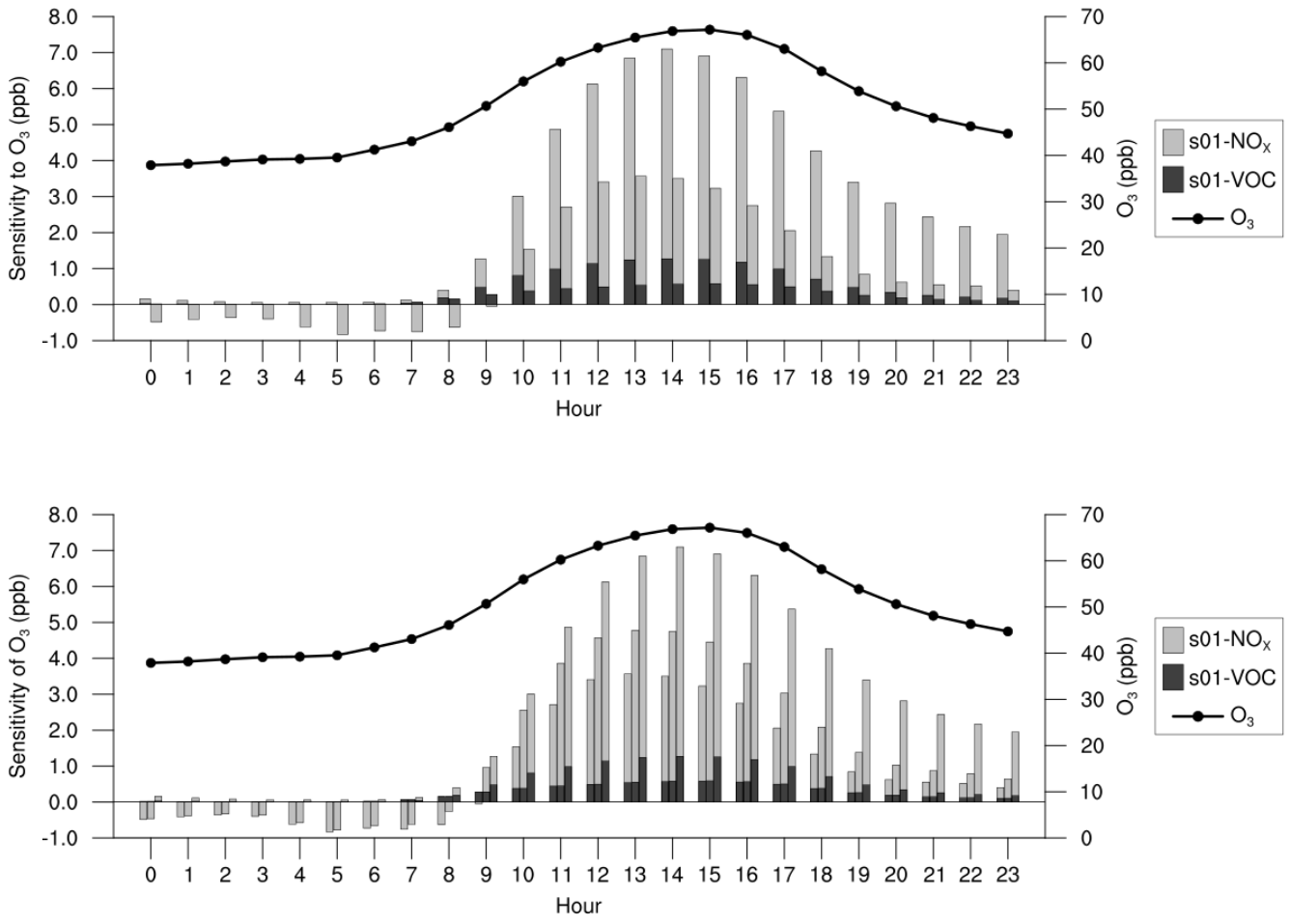


Figure ~~S7~~S8: Sensitivities derived by BFM-20 (left) and BFM-100 (middle), and a~~A~~pportionments derived by ISAM (~~left) and sensitivities (~~right) of the hourly ozone concentrations (shown by a line with markers) to NO_X and VOC emissions of s01 ~~derived by ISAM and BFM to the hourly ozone concentrations (shown by a line with markers)~~ on July 25th in ST.

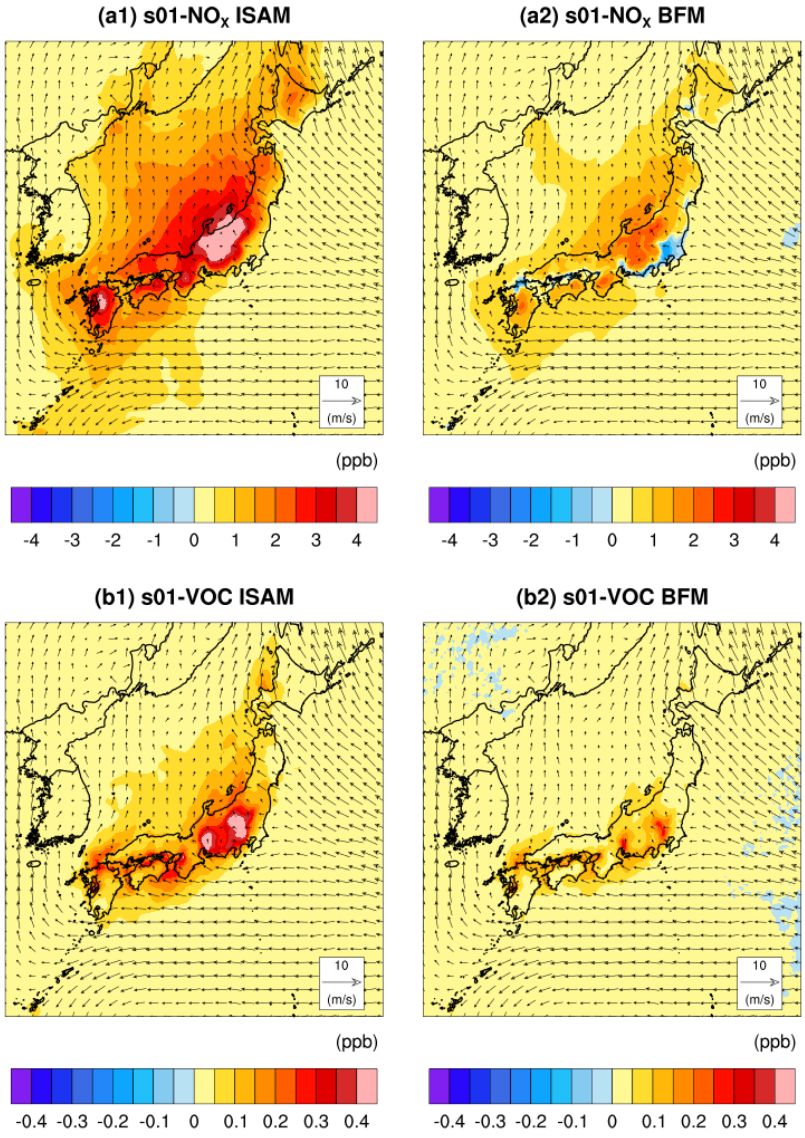


Figure ~~S8~~S9: Horizontal distributions of the apportionments and sensitivities of ~~the s01 NO_X and VOC emissions to~~ the ozone concentrations to the s01 NO_X and VOC emissions averaged for the two target weeks in summer.

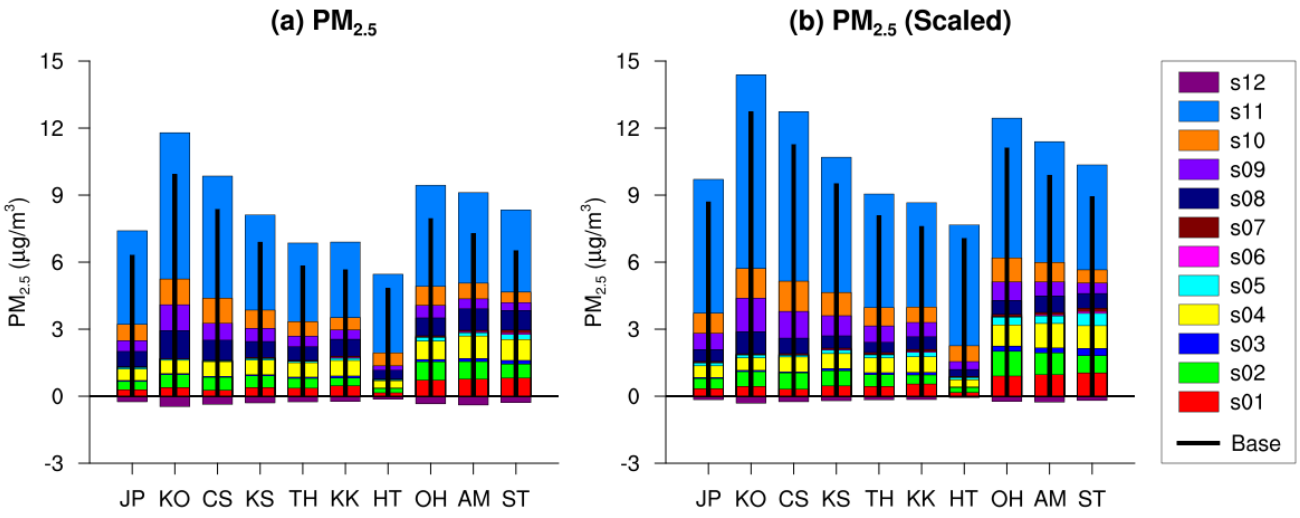


Figure s10. Source sensitivities of the annual mean $PM_{2.5}$ concentrations derived by BFM in the regions. Thick black lines represent the simulated concentrations. The values shown in (b) were scaled by ratios of observed and simulated concentrations of $PM_{2.5}$ components.