acp-2022-657: Global sensitivities of reactive N and S gas and particle concentrations and deposition to precursor emissions reductions
Ge et al.

Response to Reviewer #2

We thank the reviewer for their time spent reading our manuscript and for their recommendation for publication upon addressing their comments. Below we include all the reviewer's comments and provide in blue text our point-by-point responses. Please note that the line numbers mentioned in our responses refer to the clean revised manuscript (not the track-changed version).

Specific comments 1: Model set-up
Global 20% and 40% emission reductions were applied in this study. Some sentences are needed to justify the selection of 20% and 40%. Why a greater emission reduction (i.e., 60%) was not considered?

Response: We thank the reviewer for their suggestion. In this work, the design of modelling experiments involves balancing two factors. On one hand, the number of simulations is constrained by available computing resources and storage space. On the other hand, we aim to consider reductions that are both realistic and achievable, such as 20% and 40%, without being overly ambitious for certain regions. However, as discussed in the Discussion section, the non-one-to-one but relatively linear responses of N_r and S_r concentrations and deposition to 20% and 40% emissions reductions suggest that more ambitious reductions may be needed in the future.

In the revised manuscript, the following sentence (lines 139-141) is revised to justify our selection of reduction levels:

"Limited by available computational resources and storage space and taking the achievability of real-world emissions controls into account, the model experiments applied 20% and 40% reductions to global anthropogenic emissions of NH_3, NO_x, SO_x from all sectors both individually and collectively (i.e., reductions applied to all 3 species simultaneously)."

Specific comments 2: Section 3.1.2, Line 260
What are the sources of fine and coarse nitrate aerosol? Can you explain why coarse nitrate would increase associated with NH_3 emission reduction? Please clarify.

Response: The fine nitrate aerosol in EMEP MSC-W model is essentially NH_4NO_3 which is produced from the reaction between HNO_3 and NH_3. The coarse nitrate comes from reactions between HNO_3 and coarse particles (e.g., dust and sea salt). Reductions in NH_3 emissions cause the equilibrium between HNO_3 and NH_3 to shift away from NH_4NO_3 production and therefore free more HNO_3. As a result, more HNO_3 is available to produce coarse nitrate aerosol. In the revised manuscript, we revised this sentence to provide a clearer explanation (lines 263-265):

"Reductions in NH_3 emissions cause the equilibrium between HNO_3 and NH_3 to shift away from NH_4NO_3 production and therefore free more HNO_3 molecules. As a result, more HNO_3 is available to produce coarse nitrate aerosol, leading to a decrease in fine NO_3^- but an increase in coarse NO_3^- concentrations (Fig. S2)."

Specific comments 3: Page 18, Figure 8

Figure 8 showed the spatial sensitivity regimes based on 40% emission reductions and annual mean PM2.5 concentrations. Such sensitivity regimes shall have large seasonal variations. Do you have the model datasets to generate the seasonal maps? That shall provide valuable information to understand the SIA formation regimes.

Response: We do have monthly model outputs to generate seasonal maps. We calculated seasonal sensitivity regimes for the globe and noticed some seasonal variations in $PM_{2.5}$ sensitivity regimes in East Asia and Europe, which indeed reveals some interesting subtleties regarding the limiting factor in SIA formation in different regions. Considering this manuscript is already very long, we put this seasonal analysis into the supplement as Sect. S2:

"To reveal more details of temporal variations in global $PM_{2.5}$ sensitivity regimes, we compare $PM_{2.5}$ sensitivities to individual emissions reductions on a seasonal basis using the Northern Hemisphere calendar: spring (March, April, and May), summer (June, July, and August), autumn (September, October, and November), and winter (December, January, and February). Figure S7 presents the spatial distribution of dominant $PM_{2.5}$ sensitivity regimes in four seasons. In East Asia, the dominant regime shifts from NO_x-sensitive to SO_x-sensitive from spring to summer, while the NH_3-sensitive regime expands more and more from autumn to winter. Similar trends are observed across Europe as well, where NO_x-sensitive grids are prevalent during spring while NH_3-sensitive grids dominate during winter. The springtime NO_x-sensitive regime in these regions can be attributed to large NH_3 emissions from intensive agricultural activities in this season (Cheng et al., 2021; Dammers et al., 2019), which leads to the formation of NH_4NO_3 being primarily limited by the availability of HNO_3. Consequently, reductions in NO_x emissions decrease gaseous HNO_3 production which then decrease SIA concentrations. In the summer, NH_4NO_3 becomes less stable due to the generally higher temperature and sulfate aerosols remain a significant contributor to $PM_{2.5}$ in East Asia (Ianniello et al., 2011; Wang et al., 2013). Since the production of sulfate aerosols depends on the oxidation processes of SO_2 rather than the availability of NH_3, and NH_3 is in excess anyway, SO_x emissions

reductions become the most effective single-precursor control for $PM_{2.5}$ mitigation in this region. The wintertime NH_3-sensitive regime in both Europe and East Asia is caused by smaller NH_3 emissions (due to reduced agricultural activities) and relatively larger NO_x emissions (such as from increased domestic heating). Changes in meteorological factors (e.g., decreased vertical dispersion) may also contribute to higher NO_x surface concentrations in the winter. As a result, NH_3 becomes the limiting factor in NH_4NO_3 formation and therefore has the greatest impact on $PM_{2.5}$ sensitivities.

In contrast, North America and South Asia do not show significant seasonal variations in $PM_{2.5}$ sensitivity regimes. In the eastern US, $PM_{2.5}$ formation is NO_x-sensitive for most of the year, except for the summer when it is SO_x sensitive. This suggests that further reductions in NO_x emissions are necessary to decrease annual $PM_{2.5}$ levels in this region. In South Asia, the SO_x-sensitive regime dominates throughout the year, with the exception of northern India in the winter, which is more NO_x-sensitive. As discussed in the main paper, the extreme NH_3-richness and dominant contribution of sulfate aerosols to SIA in South Asia render $PM_{2.5}$ formation almost exclusively sensitive to SO_x emission reductions.

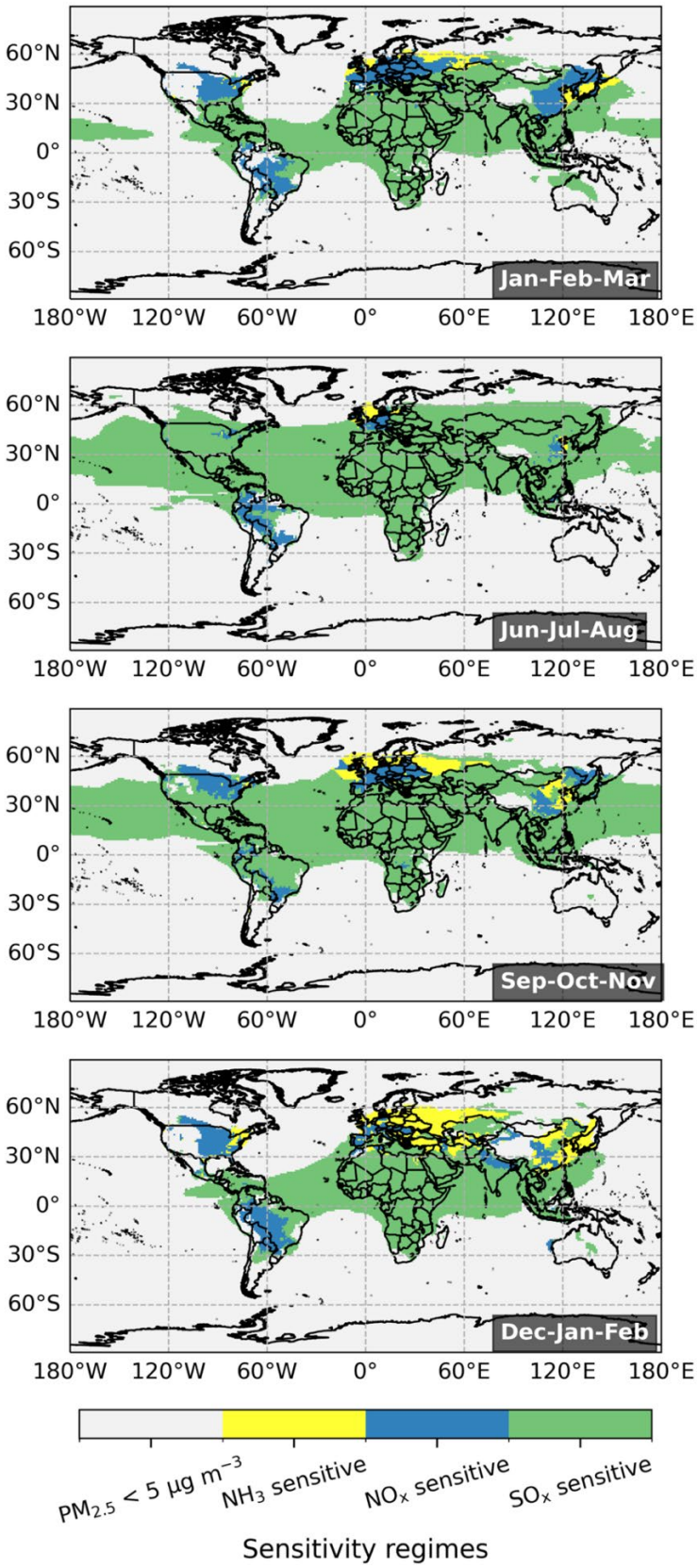

Figure S7: Spatial and seasonal variation in sensitivity regime of $PM_{2.5}$ mitigation based on data from 40% individual reductions in emissions of NH_3, NO_x, or SO_x. The regime is defined according to the precursor that yields the greatest decreases in grid seasonal average $PM_{2.5}$ concentration: NH_3 sensitive (yellow), NO_x sensitive (blue), SO_x sensitive (green). Model grids with baseline seasonal mean $PM_{2.5}$ concentrations <5 µg m^{-3} are masked out."

Specific comments 4:

For the green/red circles and stars in Figures 2, 4, 6, 9, and 11, in the main text, the symbols that are discussed as the maximum reductions (e.g., Page 11, Line 330-335) were denoted as "Min" in these figures. Please be consistent.

Response: The reason for denoting the maximum reduction as "Min" is because the actual differences are negative values. The "Max" and "Min" points in these figures represent the maximum and minimum differences between baseline and emissions reduction scenarios respectively, so if these differences are positive values, they are described as increases (rather than decreases) in the main text. Therefore, we would like to retain these figures in their current form.

Specific comments 5: Page 25, Line 708-710

It is not clear what "non-linearity" mean here in the text. We can see from Figures 3, 5, 7, and 10, the responses with respect to 20% vs. 40% emission reductions are rather linear. They deviate from the 1:1 line, however, the responses are linear. Please clarify.

Response: We thank the reviewer for pointing out this ambiguity. In the revised manuscript Sect. 4.4 (lines 752-781), we rephrased the discussion of "non-linearity" to "non-one-to-one linearity". The non-linearity is defined as a lack of one-to-one proportionality between an emission reduction and a species concentration or deposition change. Meanwhile, it is also recognised in the same section that a linearity in response to emissions reductions is apparent via the observation that the responses of $PM_{2.5}$, N_r, and S_r annual concentrations and deposition components remain essentially proportional to the precursor emissions reductions (20% and 40%) for a given precursor in a given region, albeit that the magnitude of the slope varies substantially with different precursors and regions.

Specific comments 6:

The writing of the manuscript is rather intensive. Many results are described in parallel, which makes the manuscript less focused. I understand that many results can be derived from the set of sensitivity simulations, still, the key findings of the study shall be better emphasized in the abstract and conclusions.

Response: We agree with the reviewer that the abstract and conclusion are too long. In the revised manuscript, we have shortened the two sections as requested to deliver a more focused message.

References

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