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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Source-receptor relationships between East Asian sulfur dioxide emissions and Northern Hemisphere sulfate concentrations" *by* J. Liu et al.

J. Liu et al.

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Response to Reviewer #2

We thank the reviewer for helpful comments and suggestions. We have incorporated most comments into our revised paper. Please see below for our response to each suggestion:

"This study considers the export of sulfate from East Asian (EA) emissions and the oxidant-limitation effects of changing EA SO2 emissions on local and exported sulfate. They have done this exactly by performing a single experiment with tagged emissions.By comparing the results of this tagged experiment with "sensitivity experiments", i.e. using the difference between a full experiment and an experiment with regional

emissions turned off to determine export, the degree of error from excess oxidant availability in the latter method was evaluated. The study has some unique ways of visualizing the effects of changing SO2 emissions on sulfate export, and of assessing the degree of non-linearity. Below are major and minor points that should be addressed prior to publication in ACP."

"Specific comments: 1. The study uses emissions from the early 1990s. I was surprised at the amount of agreement of model using 1990s emissions with EANET observations from after 2003. Does this suggest that EA emissions have not changed in the past decade?"

We follow the reviewer's suggestion and add a short discussion in Section 2 (p.9) of the revised paper: 'While the model shows little mean bias with respect to the EANET observations (from the early 2000s), it is likely that the model would overestimate sulfate concentrations over EA during the early 1990s (the period for which our emissions were estimated) because SO2 emissions from East Asia are estimate to have increased between the early 1990s and the early 2000s (Klimont et al., 2001).'

However, given the fact that most EANET observations are collected from Japan, it is also possible that the agreement between model and observations reflects a balance between the increased SO2 emissions from the mainland EA and the decreased local SO2 emissions from Japan.

"2. The SO2 emission inventory used here should be identified and referenced."

We are using the MOZART-2 standard emissions, which has been described by Horowitz 2006. In particular, anthropogenic emissions of SO2 are from the Emission Database for Global Atmospheric Research (EDGAR) v2.0 (Olivier et al., 1996). We add a table in the revised paper (Table 1) to show the annual sulfur emissions from each continental region.

"3. P 5541, lines 25-28, these seem incorrect: "the wet deposition rate for SO2 is set

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equal to that of H2O2. For sulfate, the wet deposition rates are set to 20% of that for the highly soluble gas HNO3". SO2 is much less soluble than H2O2. And sulfate is highly soluble."

The detailed description of sulfate simulation in MOZART-2 is given by Tie, et al [2001, 2005]. As we now discuss in Section 2 (p.7), while the Henry's law constant of SO2 is indeed low: 'In the case of SO2, its dissolution in cloud droplets and precipitation is enhanced considerably over its physical solubility by acid dissociation and by rapid aqueous-phase oxidation from S(IV) to S(VI) (primarily by H2O2). To reflect this enhanced solubility, wet removal of SO2 is calculating using an increased effective Henry's law constant, equal to that of H2O2, as assumed by Tie et al. (2001, 2005). '

The approach used here is a somewhat approximate attempt to account for the enhancements to solubility. A unified treatment of SO2 dissolution and oxidation in clouds and precipitation would certainly be better.

"4. The term "background" to refer to sulfate not derived from the local region is misleading, since background often means natural. "Non-local" might be better."

In this study, the background sulfate includes both "non-local" anthropogenic sulfate and total natural sulfate. We have clarified this in Section 3 (p. 11) and include an explicit definition.

"5. P 5544 Lines 21-end of page. As SO2 increases, maybe the larger increase in sulfate at higher altitudes and smaller increase near the surface is due to greater oxidant availability aloft. H2O2 typically peaks at mid-altitudes of the troposphere."

From Figure 8, it is true that the oxidation capacity to oxidizing SO2 is relatively stronger at higher altitude than at the surface over the EA source. But over the downwind region, the S-R relationship is approximately linear at both the surface and 500hPa. This indicates that the limitation of oxidants may not be the only reason to explain the spatial variability of AEA0.1 at different altitudes over the eastern Pacific and the U.S.

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Based on the reviewer's suggestion, we have made some clarification in Section 3 (p.11-12).

"6. P 5546. In addition to considering the degree of non-linearity to increased SO2, it would be worthwhile to consider also the amount of linearity if emissions are reduced, i.e. at what point of the curve is EA now?"

As the reviewer suggests, we consider the linearity of the emissions-concentration relationship at SO2 emission rates greater and less than the standard emissions. The Linearity index shows the cumulative linearity of SO2 emissions versus sulfate concentrations as the EA SO2 emissions range from 0 to 1.5 times the standard emissions. The linearity index (or the shape of line OECF in Figure 7) therefore is determined by the oxidant availability changes across a range of SO2 emissions.

"7. A more realistic simulation would include deposition of SO2 on particle surfaces. Discuss how including this in the model would affect the results."

This is a very important point. While MOZART-2 includes SO2 deposition and oxidation on cloud droplets, its deposition and oxidation on aerosol surfaces are not included. Since aerosol surfaces provide another pathway for SO2 oxidation, including this mechanism will somewhat reduce the non-linearity over the source regions. However, oxidant supply (rather than the availability of surfaces) is the primary limiter of SO2 oxidation, so this effect is likely to be small.

"Technical comments: 1. 5538, Lines 9-10, specify that this is in the column."

Revised.

"2. 5538 Line 20, the meaning of "sensitivity" should be clarified here, also on page 5540 lines 14-15."

The meaning of sensitivity is clarified.

"3. Several periods are missing: p5543 L 25, p5544 L 9, p5544 L 24"

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Added.

"4. P 5548 Sentence starting at line 5 is poorly constructed. The model transport findings do not depend upon the health effects: "Since sulfate aerosol is... harmful to human health, ...summertime trans-Pacific transport.. is important""

We have improved this sentence in the revised paper.

Interactive comment on Atmos. Chem. Phys. Discuss., 8, 5537, 2008.

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Interactive Comment