

Interactive
Comment

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 #4

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 paper uses the MOZART model to analyze the source-receptor relationships between the sulfur dioxide emissions in East Asia and the sulfate concentrations in the Northern Hemisphere at the surface and 500 hPa. This is done by tagging the emissions at different regions and increasing/decreasing EA emissions. The "non-linearity" of SO₂ to sulfate conversion is also investigated. The paper is well written and focused.

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However there are several major issues that should be dealt with and clarified."

"1. The model simulation is done for year 1990 to 1991 using the "standard MOZART-2 inventories" representing the early 1990 emissions. Since all the simulations used in this work are based on the standard or changes from the standard simulations, it is necessary to show the standard emissions, either by a map or by a table, to give readers an idea about the distributions and magnitudes of the emissions."

We have added a table (see Table 1 in the revised paper) to show the magnitudes of SO₂ surface emissions from each region.

"2. The model evaluation is done in a very crude way - only multi-year averages of model and data are compared in Fig. 2, and the model and data are compared for different periods of time. I don't understand why not using data for the same time periods, at least for IMPROVE, EMEP, and probably RAMAS too, for 1990-1991? I understand the reason for using the 2000-2004 EANET data because of the data availability, but it seems that the model calculated sulfate (representing early 1990s) is generally much higher than the data measured in the 2000s. This means the model would have overestimate the EA sulfate even worse if there were 1990s data to compare with, since the Asian anthropogenic emission is believed to be much higher in the 2000s than in the 1990s. This overestimation of sulfate near Asia, together with the model underestimation of the sulfate over North America (e.g. comparison with IMPROVE data in Figure 2) implies that the model could have overestimated the Asian emission impact on NA. Since the EA influence is the theme of this paper, these problems have to clearly addressed and resolved."

We think the reviewer's suggestion is very important 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

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estimate to have increased between the early 1990s and the early 2000s (Klimont et al., 2001).¹

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.

"3. Global distribution and transport: I cannot see from Figure 3 what is described in section 3, that is the transport from EA and the EA influence on North America surface concentrations are the strongest in MAM and JJA; Figure 3 shows the transport is strongest in MAM and weakest in DJF, and the EA influence on NA surface concentration is highest in MAM. I also don't see the transport at 500 hPa is "very strong in summer" (Fig 3a) - the transport in summer is about the same as in the fall, and weaker than in MAM. And westerlies prevail in all seasons. Also, why is the seasonal significance changed from the previous study by the same authors? Is this due to the use of different meteorology, or something else?"

We have improved this discussion in the revised paper. We now say in Section 3 (pp.9-10):

'The contribution of EA to surface sulfate over the western U.S. is highest in Spring (MAM) (up to 0.15 $\mu\text{g}/\text{m}^3$) and lowest in Winter (DJF) ($<0.06 \mu\text{g}/\text{m}^3$). In our earlier work (Liu et al., 2005; Liu and Mauzerall, 2005), we found that trans-Pacific transport of an idealized tracer (with a fixed first-order decay lifetime of 2 weeks) to be strongest in winter-spring and weakest in summer. The results here differ from those of our earlier study because sulfate production is highly dependent on the abundance of oxidants, which have different seasonal variation than trans-Pacific transport.'¹

"4. Surface sulfate: The 0.1 $\mu\text{g}/\text{m}^3$ is chosen to exhibit the EA contribution to NA surface sulfate. This number should be put into some air quality context. How significant is this number? What is the USEPA standard for a "good" air quality? Will the transport

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of sulfate from EA ruin the US air quality? The 30-50% and 10-20% contributions to the "background"; sulfate sounded significant, but how does the EA sulfate compared with the NA sulfate (not the background)? Wouldn't it be more appropriate to compare local pollution with imported pollution to see the relative magnitude?"

Based on the reviewer's suggestion, we have compared 0.1 ug/m³ to the US EPA annual average PM_{2.5} standard in the revised paper (Section 3, p.11). We choose 0.1 ug/m³ because in our baseline simulation the spatial extent of AEA0.1 covers the western U.S. at the surface. If we use other values, for instance 1 ug/m³, its spatial extent only covers the western Pacific (see Figure 3b).

For PM, epidemiological studies have shown that the concentration-response relationship for long-term exposure to fine PM is linear and without a threshold. From Pope et al. (2002), each 10ug/m³ elevation in PM_{2.5} is associated with ~4% increased premature mortality. Our follow-up paper shows that EA sulfate alone could potentially lead to 300-400 annual premature deaths over the North America.

As shown in Figure 4(b), EA sulfate contributes up to 15% of the total sulfate concentrations over the western U.S., but less than 1% of the total sulfate concentrations over the eastern U.S. We also compare the EA sulfate with the local sulfate from NA in our 2007 ERL paper. Please see:

Liu, J. and D. L. Mauzerall, Evaluating the potential influence of inter-continental transport of sulfate aerosols on air quality, Environ. Res. Lett. 2 045029, doi:10.1088/1748-9326/2/4/045029, 2007

"5. The "non-linearity" issue: I don't think the non-linearity of sulfate formation from SO₂ is explained/demonstrated correctly. If there is no oxidant limitation, sulfate production should be linearly dependent on SO₂. Only when there is not enough oxidant to oxidize SO₂ then the sulfate production is proportional to the oxidant concentrations, not to the SO₂ amount. This means that the curve shown in Figure 7 should have a straight C_SO₄ line from O to C before bending over from C to F. The OEC curve is

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wrong - under no circumstances the sulfate concentration could be higher than that from a complete SO₂ conversion (straight line)."

Given the reviewer's helpful comments, we clarify our explanation for Figure 7. The x-axis should be 'SO₂ emission scale factor' (i.e., the SO₂ emission in each sensitivity run is scaled to the baseline emissions; this is why we put '1' to indicate the base situation). The Y-axis should be the 'sulfate concentration scale factor' (i.e., the sulfate concentration in each sensitivity run is scaled to the baseline concentrations). Therefore, the curve OEC in Figure 7 does not represent more than complete conversion of SO₂ to sulfate, and SO₄ is not really increasing 'faster' than SO₂ emissions. This is just an artifact of normalizing to present-day emissions/concentrations. Figure A2 (in the supplementary material) gives an example which shows the S-R relationship between EA SO₂ emission scale factor and EA sulfate concentration scale factor over (a) the EA source and (b) the NA receptor. This is the basis for Figure 7.

"6. I also feel the "non-linearity index"; is very confusing. How is L calculated from equation (1), that is, how are the values of S_{OCE}, S_{CFA}, S_{OAB} defined from the model results? Why is there no L=0 anywhere in Figure 8, which means that everywhere in the NH sulfate production is oxidant limited? This does not make sense at all."

We calculate the areas of OCE and CFA based on the actual OECF curve in each grid box as indicated in Figure A2 (in the supplementary material). Since we only conducted six runs, the values of these areas that we calculated could be smaller than the actual areas. The L values are in the range 10-20% for Figure A2a, and 2.5-5% for Figure A2b (Figure A2 is given in the supplementary material). Therefore, when L < 5%, the S-R relationship is very close to linear. Since sulfate is a secondary aerosol, the change of SO₂ emissions will more or less affect the oxidants and disturb the original photochemical reactions over both the source and downwind regions. Therefore, it is difficult to find a place where S-R relationship is perfectly linear (i.e., L=0).

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"7. More on the non-linearity index: It is said in Figure 8 caption "low numbers indicates approximate linearity". Quantitatively, how low is the "low number"? Shouldn't it be zero for linearity according to Figure 7?"

A value of $L=0$ would indicate perfect linearity, but this almost never occurs in practice. We find that when $L<5\%$, the relationship is close to linear. We include this in Section 4 (p.13-14).

"8. Furthermore, SO₂ oxidation should be more explicitly explained. SO₂ has very little effects on OH, so the gas-phase sulfate production should be pretty much linear to SO₂ concentration. The oxidant limitation is mainly from the H₂O₂ amount in the cloud/rain. Basically, the entire section 4 should be substantially re-worked."

We do quantify the effect of changing EA SO₂ emissions on the oxidation levels. We have also mentioned that the oxidation limitation is mainly due to the liquid oxidation of SO₂ by H₂O₂. Figure A1 (in the supplementary material) shows the plots which we used to quantify how changing EA SO₂ emissions affects OH, O₃ and H₂O₂ concentrations.

"9. This is certainly not the first paper studying the Asian sulfate pollution on large scale. To put the findings into some perspective, comparisons with other papers, in addition to Park et al 2004, are needed (e.g. Heald et al. 2006, Koch et al 2007, Chin et al 2007, etc.)"

We accept the reviewer's suggestion and make some additional comparisons between our work and other studies in Section 3 (p. 9-11).

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

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