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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.

Anonymous Referee #4

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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. 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 arebased 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.

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.

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?

4. Surface sulfate: The 0.1 ug/m3 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 of

ACPD

8, S2408–S2411, 2008

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?

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

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 defind 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.

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?

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.

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.)

ACPD
8, S2408–S2411, 2008

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8, S2408–S2411, 2008

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