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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 #1

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:

"A main topic of this study is the non-linearity between sulfur dioxide emissions and sulfate mixing-ratios at surface and in the free troposphere. This aspect needs a more thorough analyses. Non-linearity is caused by low oxidant concentrations (OH and H2O2) in winter and by H2O2 limitations in regions with high SO2 emissions. As oxidation rate is the key process controlling the non-linearity, the vertical and seasonal

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distribution of the oxidation rates should be presented and the importance of the different chemical transformations depending on altitude, latitude, season, and magnitude of SO2 mixing-ratios should be discussed (e.g. E-W cross section over Asia and Pacific)."

We think the reviewer's comments and suggestions are very important. However, we didn't archive the oxidation rates between SO2 and the three oxidants (OH, H2O2, and O3). Given the importance of this topic, we tried to use what we have to analyze this issue. Figure A1 (in the supplementary material) shows the monthly concentration and its relative change (to baseline situation) of these oxidants (i.e., OH, O3 and H2O2) over the EA source as a result of changing EA SO2 emissions. We find that the change of EA SO2 emissions strongly influences H2O2 concentrations (particularly in summer) over EA, but has only a slight influence on OH and O3 concentrations over EA (mostly in winter, spring and fall). We add the discussion in Section 4 (p.14) of the revised paper: 'For example, in this study a 50% change (either increase or decrease) of EA sulfur emissions is associated with 5-10% (1-2%), 2-5% (0.5-2%), and 1-2% (0.5-1%) change in surface H2O2, OH, and O3 concentrations over EA (eastern Pacific), with the largest percentage changes occurring in winter for OH and O3 and in summer for H2O2 (plots are given in the supplementary material).'

"Minor comments: 1. Abstract: I miss, the conclusion that mainly local emissions not long-range transport affects air quality."

EA sulfate accounts for approximately 30-50% and 10-20% of North American background sulfate over the western and eastern U.S. However, the magnitude of EA sulfate concentrations is much smaller than the magnitude of domestic sulfate concentrations over the U.S. As shown in Figure 4(b), EA sulfate contributes up to 15% of the total sulfate concentrations over the western U.S. In addition, from [Liu and Mauzerall, 2007], the population weighted EA sulfate concentration only accounts for less than 0.5% of the population weighted US sulfate at the surface. We follow the reviewer's suggestion and add this point in the revised abstract.

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"2. page 5541 ln 25: same wet deposition rate for SO2 and H2O2: The solubility of SO2 is considerably lower than that of H2O2? In my opinion, the wet removal of SO2 is negligible."

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.

"3. page 5542 ln 15-25: The meteorology of the simulations covers the period 1990 to 1991, whereas the observations used for evaluation cover different periods. As regional sulfur dioxide emissions underwent significant changes between the 1980es and the early 2000es, this should be addressed in the discussion about the model's performance."

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 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.

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"4. chapter 3: "Global contribution of sulfate aerosol from EA" Kritz et al (Tellus B, 1990) reported fast transport of radon-222 from EA to North America in spring and explain this fast transport by convective activity in China and subsequent mixing of radon into the jet-stream. Please, refer to Kritz's study and discuss whether this mechanism might be important for sulfur dioxide and sulfate transports as well."

The mechanisms for transporting EA air pollution to the North Pacific differ significantly by season. A lot of studies have evaluated these mechanisms. We follow the reviewer's suggestion and add a discussion in Section 3 (p. 9).

"5. Typo: Please, replace throughout the paper "mPa" by "hPa"."

We have replaced all "mPa" by "hPa". Thank you for catching this.

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

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