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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Impact of Chinese SO_2 emissions on submicron aerosol concentration at Mt. Tateyama, Japan" *by* K. Osada et al.

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We thank referee #3 for very constructive comments with respect to the overall clarity of the article. Modified words or sentences are highlighted as yellow in the text.

Comment: The paper 'Impact of Chinese SO2 emissions on submicron aerosol concentration at Mt. Tateyama, Japan' explains seasonally resolved trends of SO42- and mass concentration at the Japanese Tateyama mountain observatory with Chinese SO2 emission developments. The observatory represents free-tropospheric conditions during nighttime and is affected by seasonally changing airmass transport from China. Transport, chemical transformation and the relative Chinese contribution to SO42- immisions at Mt. Tateyama are simulated with a 3-D regional-scale CTM covering the east-Asian domain. The paper is topical, well written and reaches substantial novel

conclusions about pollution export from one of the world's largest and most emerging source regions. I support publication in ACP with only few comments to be taken into account. Particularly the seasonality of the trend analysis could be discussed in more detail.

Response: We added additional discussion and a description of our analysis of the seasonality of trends to the revised manuscript using the new Fig. 4 and Tables 1 and 2.

Comment: Emission inventories: the use of climatological values for the biomass burning emissions and the only annual resolution of the REAS database may limit the significance of the simulation results but I'm not sure whether the use of novel products like e.g. GFEDv2(http://ess1.ess.uci.edu/∼jranders/data/GFED2/readme.pdf), GEIA for SO2 could yield a closer agreement with the stations observations. Admittedly, the effort of implementing new databases into the model system would be beyond the scope of this paper. Mayby the authors could comment on respective future plans.

Response: The largest spikes (May–June 2003) detected in Fig. 2 were not reproduced in Fig. 7. Large variations in seasonal and year-to-year emissions from open burning are recognized along with boreal forest fires (Streets et al., 2003; van der Werf et al., 2006). Our modeling system does not incorporate these temporal variations. Therefore, simulated EC and OC concentrations might not reproduce such temporal variations. A further direction of this study will be to include various temporal variations in greater detail. We added this to the revised manuscript.

Comment: p 16532, line 6ff Can you estimate how the SO42- immision (and it's seasonal contribution) at Mt. Tateyama in the CTM would change if increasing Chinese SO2 emission are anticipated instead of keeping them at the 2005 level? Is it likely that Chinese emission control is that effective during the recent years to suggest your approach?

Response: As discussed on L18–21 in P16534 of the original manuscript, recent SO2 emissions in China became difficult to estimate after 2005. Our intention with this paper is to demonstrate the impact of anthropogenic emissions in China on aerosol concentration measured at Mt. Tateyama. For that reason, we decided not to use projected values after 2005.

Although our data show that volume concentrations in winter seem to be decreasing slightly, more long-term data are necessary to confirm the effectiveness of Chinese emission control on SO2.

Comment: P16533, line 5ff: How stable are the general weather situations during the flow regimes in early summer and in winter. What could be the relative contribution of trajectories in summer reaching back to the fire regions?

Response: We added a new Fig. 9 to the revised manuscript to portray the variability of trajectories during June and December for the three years from 2006 as examples for this topic. In summer, air trajectories were derived from various locations in eastern Asia with large year-to-year variation. The relative contribution of the Siberian air mass also varies according to the season and year.

Comment: P16534, l 677: Can it be excluded that the percipitation rate and its seasonality at Mt. Tateyama deviates significantly from that the Kamiichi station, e.g. due to different orograpic flow conditions? Can the model uncertainty due to sub-grid scale scavenging be estimated?

Response: We added additional discussion and analyses to the revised manuscript, including that of the local meteorological situation. Local meteorological conditions might be different in summer because of variable airflow patterns to Mt. Tateyama, but we expect mostly similar conditions in winter as a result of nearly constant northwesterly winds. In our model system, wet and dry depositions are included but limited for resolution of the 80 $\times$ 80 km grid.

Comment: Fig 4. and correcponding discussion of trends: Add vertical axis annotation

ug/m3. The linear trend analysis does not take into account the seemly levelling-off of the observed particle volume in the recent years in spite of the presumably increased emissions. Possible contributions from sources other than China. As the model does not know emission seasonality: what trend does it produce for the other periods selected in Fig 4?

Response: We revised Fig. 4 including units of the vertical axis and data for the other months. We added discussion of the new Fig. 4 to the revised manuscript. As you pointed out, Chinese emissions do not increase linearly over the years. To discuss the direct relation between the Chinese contribution of SO42- simulated and aerosol volume observed, we added a new Fig. 8 (lower panel). We also added simulation results of total anthropogenic aerosols to the new Fig. 7. Seasonal variation was not given in the emission data. Therefore, seasonal variations simulated in new Fig. 7 (old Fig. 6) are attributed to the seasonality of transport. We added this explanation to the revised manuscript.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 16527, 2009.