Dear anonymous referee #2,

We very much appreciate your constructive comments, useful information and your time. Thanks to your review, our manuscript was substantially improved. Point-by-point responses to your comments are written in blue in this letter.

Sincerely yours,
Mizuo Kajino

General comments:

This is a useful paper describing a detailed model-analysis of source-receptor relations in East-Asia. The approach is not new, but the results are of interest, also in view of possible abatement strategies and policy
Thank you for your evaluation.

Specific comments:
(Comment #1)
On page 30093 a short description is given of the one-dimensional cloud model " based on RADM", and the box submodel. Based on the results discussed further of the wet fluxes of sulphate, it is suggested to present a more detailed description of this part of the model, and the possible sensitivity of the model results to the approach taken.
(Answer #1)
Thank you for your suggestion. We inserted a new paragraph describing a little more details of the cloud process submodel after the first paragraph of section 2.1, ln.27 of p.30093:
"Clouds play an important role in chemical transformation and removal of trace species, especially for sulfur, and thus the cloud process modeling of RAQM is described a little more in detail. The cloud process submodel used in RAQM is based on RADM (Chang et al., 1987) so one could find a full description in their paper. Each representative cloud in a grid has a depth and fractional area of coverage determined by the grid-scale environmental parameters. A one-dimensional diagnostic cloud model is used to specify vertical distributions of several cloud dynamical and microphysical properties such as cloud fraction, cloud base, cloud top, and condensed water content. To determine the properties, three sets of diagnostic equations are applied for the three respective cloud types, precipitating cumuliform clouds, precipitation stratiform clouds, and fair weather cloudiness. Rainout of aerosols, dissolutions of soluble and

reactive gases, and chemical reactions in a cloud are computed using a box aqueous chemical and scavenging submodel. SO_2 dissolved into water droplets is oxidized by H_2O_2, O_3, methyl hydrogen peroxide, peroxyacetic acid, and trace metals (Fe^{3+} and Mn^{2+}). Accumulated wet deposition from precipitating clouds is computed by integrating the product of the grid-averaged precipitation rate and mean cloud water composition during each cloud lifetime. It should be noted here that the precipitation rate predicted by MM5 and by this submodel are independent. "

We couldn't show the sensitivity analysis for wet deposition process within the RAQM model, because it is not a simple model like the one in which applying wet scavenging ratio as a function of surface precipitation to the air concentrations to get wet deposition rate. It is rather complex combinations of modules, such as cloud dynamical, microphysical and chemical schemes. Instead of showing the intra-model sensitivity, uncertainties deviated from the inter-model comparison study are newly discussed in terms of sulfur in precipitation. We inserted the following sentences regarding model uncertainties for the prediction of wet deposition at ln.3 of p.30100:
"Wang et al. (2008) showed the variations in prediction of wet deposition amounts of sulfate, nitrate and ammonium by the 8 different models participated to the MICS-Asia Phase II study. They showed those varied substantially even for sulfate by factor of >10. Even though the emission flux of sulfur could be accurately derived, wet deposition of sulfur would be varied substantially among the current chemical transport models"

(Comment #2)
On page 30095 it is stated that monthly values of biogenic VOC-emissions are used. When this is the case, the rather poor model performance for ozone are not a surprise. Could the authors comment on this, and explain why not the biogenic emissions are calculated on an hourly basis.
(Answer #2)
Because hourly variation of biogenic VOC emission is not provided, we didn't use it for the simulation. As you pointed out, it might cause poor model performance for ozone. We inserted the following sentences at ln.6 of p.30099.
"Since hourly variations for NO_x and VOC emissions were not provided in the inventories we used, the variations were not considered for the simulation. This might cause the poor model performance for ozone. Still as long as sulfur compounds are concerned, this level of deviations in O_3 (R~0.5 & FAC2>0.9) will not affect substantially on predicting conversion rates of S(IV) to S(VI) in the air as well as in water droplets. To predict highly reactive O_3 and nitrogen compounds, hourly variations in emissions should be carefully taken into account."

(Comment #3)
On page 30098 the grid resolution is given as 60 km, the resolution of MM5 is 45 km (page 30094). Why is there a difference in grid resolution?
(Answer #3)
I am sorry that I was confused with another setting of simulation. As we stated in section 2.2 at ln.20 of p.30094, the horizontal grid resolution of RAQM used in this study was 0.5deg. We changed the line as follows:
"It is coarser than other inventories and the model grid resolution (0.5 degrees)."

(Comment #4)
On page 30099 the overestimation of the precipitation, and the underestimation of the wet deposition flux of S is presented. This would mean that the conversion to sulphate is much too low. Could this be caused by the approach described on page 30093, see my first remark
(Answer #4)
According to your advice in comment #1, we added a little bit more detailed description of our cloud model. After that, we realized that precipitation amount we showed in Table 2 was the one predicted by MM5, and thus it was not consistent with the one used for calculating wet deposition amount of RAQM. The precipitation amount of RAQM is a diagnostic variable and not physically predicted like that of MM5. Therefore, we removed the precipitation amount from Table 2 and discussion on it in section 3.

The overall discussion for the model predictability and discrepancies in section 3 was thoroughly modified.

(Comment #5)
On page 30104 the deposition is given in kTS/yr, per region. Because the size of the regions differ, it would be informative to give the results also in ktS/ha/yr.
(Answer #5)
Thank you for your constructive suggestion. We listed the values in $mS/m^2/yr$ as well in Table 4 and inserted the following sentence to ln.27 of p.30103.
"Because the sizes of the regions differ, the deposition is given in $mgS/m^2/yr$, too. The unit is consistent with that in Figs.3 and 4."

(Comment #6)
The S-deposition to the ocean is given on page 30105. It is unclear whether ship emissions are included in the model calculations. Considering all percentages given, could something be said

about the overall uncertainty of the results.

(Answer #6)

SO_2 from shipping was not considered in the study. We inserted the following sentences at ln.21 of p.30095:

"SO_2 from shipping and biogenic sulfur compounds such as dimethyl sulfide (DMS), dimethyl disulfide (DMDS), OCS, H_2S, CS_2 and CH_3SH from ocean surfaces were not considered in the study. The contribution of emission fluxes of those species to concentrations and depositions of SO_2 and sulfate over the region could be much smaller compared to anthropogenic SO_2 from China and that from Miyakejima volcano (Streets et al., 2003; Kajino et al., 2004). However, those can be a reason for the discrepancy between the simulation and the observation because the observation sites are mostly located at isolated islands or capes, surrounded by ocean."

It was found difficult to evaluate the contributions of sulfur which is not calculated in the model. SO_2 from international ship emissions accounted for only 5% of that from the three Northeast Asian countries (Streets et al., 2003), but as you pointed out, the contributions on deposition over ocean should be substantial. We inserted the following sentences at the end of section 4.4, ln.27 of p.30105:

"It is difficult to evaluate the contributions of sulfur, which is not calculated in the model, such as SO_2 from shipping and biogenic S from ocean surfaces. SO_2 from international shipping accounted for only 5% of that from the three Northeast Asian countries (Streets et al., 2003) and that from biogenic dimethyl sulfide (DMS) accounted for less than 5% of that from the countries and the Miyakejima volcano (Kajino et al., 2004). However, as far as deposition over ocean is concerned, contributions of those S could be substantial."

Technical corrections

(Comment #7)

-page 30092, line 3: diesel instead of diasel

(Answer #7)

We deleted the phrase included the word after the revision.

(Comment #8)

-page 30094 line 10, were chosen for the simulations, so add " the"

(Answer #8)

We added "the".

(Comment #9)

-table 3. The subheader Annula deposition does not belong here

(Answer #9)

Thank you. We deleted the subheader.