General comments:

(Comment #1)
It is a strange terminology when MM5 in combination with RAQM is considered as "one" model. You may talk about a model system consisting of these two models and give this model system a specific name. This description of "the model" should be rephrased throughout the article.

(Answer #1)
Thank you for your comment. Instead of name the system, we rephrased the model as simply RAQM and then separately stated that RAQM was driven by MM5. We modified sentences in ln.3-4 of p.30090, ln.2 of p.30093 and ln.5 of p.30109.

(Comment #2)
It is worrying when the sulphur wet deposition is underestimated by 30 to 50% despite for a significant overestimation of precipitation amounts in the applied data from MM5. One would have expected more discussion in possible reasons for this underestimation.

(Answer #2)
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. "

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. Thus, 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 according to your advices and comments #3, #4 & #6. Please see the respective answers of us.

(Comment #3)
Apparently SO2 concentrations are well reproduced by the model, whereas SO4 2- contents in aerosol phase are underestimated by about 30%. I am not familiar with the applied emission inventories –the submitted paper provides references to Park et al. (2005) and Kim et al. (2010) for which the later is still in review (and therefore not available for the review). Both in Park et al. (2005) and in the current article biogenic VOC emissions are explicitly mentioned, but it is not clear whether the emission inventories includes biogenic sulphur compounds like DMS, DMDS, H2S, although the article describes the handling of volcanic sulphur emissions explicitly and these emissions seem to be well accounted for. Another issue may be the quality of ship emissions that often have larger uncertainties than land based emissions. Similarly it would be interesting to know whether inventories from e.g. China and India with strong growth are well updated with information about emissions from power plants and industries.
(Answer #3)
We inserted the explanation on the other S emissions at the end of section 2.2, ln.21 of p. 30095 as follows:
"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."

The base year of the emission inventory used for the study is the same as the simulation year 2002, but restricted for the three Northeast Asian countries. Therefore, the rapid growth of emissions from power plants and industries are well updated for China but not for India. We added the following sentences at ln.5 of p.30095:

"The base year of the emission inventory is same as the simulation period, the year 2002. LTP is one of the joint research projects under the Tripartite Enviironment Ministers Meeting among Korea, China and Japan (TEMM), which aims to understand the state of air quality in Northeast Asia, laying a foundation for research on long-range transports, to develop the scientific basis for environmental decision-making, and ultimately to improve air quality in Northeast Asia. In this context, we focused mainly on environmental impacts of anthropogenic emissions from the three huge-emitter countries over the Northeast Asian sub-region. The emissions from other Asian region such as South Asian and Southeast Asian countries are not included in the emission inventory."

(Comment #4)
Another issue very important issue and maybe the main reason for underestimations is how realistically the initial and the boundary conditions have been handled in the simulations. It is stated that these are obtained from lower end values of observations from "recent" studies in East Asia with reference to papers by Carmichael et al. (1998) and Luo et al. (2000). Again with the rapid development in power production and industry in this part of the world, it seems as these values may be completely outdated, which again may be the reason for underestimations?

(Answer #4)
As discussed later in Answer #6, the initialization problem is not very serious for the whole simulation as the integration period for the simulation is one year. As you mentioned later, at least 10 days are necessary for the spin-up period, whereas we used only 3 days. Thus, the first 7 days of the 365 days simulation can be underestimated due to the initialization problem. Therefore, as you pointed out, the boundary condition could be a reason for the underestimations. We inserted the following sentences at ln.17 of p.30099:

"As we focused on SRR of sulfur among the three Northeast Asian countries, the contributions of boundary conditions, which were not seriously taken into account, could be a reason for the underestimation of the modeled SO_4^{2-}. For the rapid growth in power production and industry in this

part of the world, our initial and boundary conditions obtained from Carmichael et al. (1998) and Luo et al. (2000) could be outdated for the simulation of the year 2002. Besides, emissions from South and Southeast Asian countries were not included in the simulation. As the plume from outside the domain was transported in longer distances, S(IV) could be sufficiently oxidized to S(VI). This is consistent with the fact that modeled S(VI) was underestimated whereas S(IV) was reasonable."

We newly added discussion in the revised manuscript that the uncertainty in measured nss-SO_4^{2-} at the Japanese monitoring stations could be one of the reasons for the discrepancies. The following paragraph is located at the end of section 3, ln.7 of p.30100:

"Recently, concerns are raised for uncertainties in observed nss-SO_4^{2-} at the Japanese EANET stations, located on small islands or isolated capes. Because the monitoring sites are located very close to the ocean (within 1km apart from coastlines), Na^+ concentrations in the air as well as rain water samples are high. For example, at Sado island, facing the Sea of Japan where surface wind is strong and ocean surface is wavy during winter monsoon, monthly mean nss-SO_4^{2-} in January 2009 in the air and in precipitation were calculated as only 68.7% and 13.9% of measured total SO_4^{2-}, respectively, using Eq.(2). This too simule form of Eq.(2) cannot perfectly subtract the seasalt contribution from measured SO_4^{2-}, because the weight fraction of SO_4^{2-} in sea-water should have natural variations. As the concentration of Na^+ increases, the uncertainty in observed nss-SO_4^{2-} values should be enhanced. Still, at the current stage, we cannot judge whether this uncertainty will result in overestimation or underestimation of simulated nss-SO_4^{2-}, but this could be one of the reasons for the discrepancies between modeled and observed nss-SO_4^{2-} in the air as well as in precipitation."

(Comment #5)
The formula on page 30095 is applied for determining the relative contribution from a specific source to a given receptor. One of the major difficulties is that emissions in one region affect the fate of emissions from another region. In the current paper, scenarios have been performed by switching out one source at a time. In recent works e.g. in EMEP modelling work it has been found to be more robust in case a 20% reduction is introduced rather that a full removal of the source input. Still the non-linearity means that a sort of double counting may easily take place, although this problem is usually of minor importance when sulphur compounds are considered.
(Answer #5)
Thank you for your useful comments. We inserted the following paragraph at the end of section 2.3, ln. 7 of p.30096:
"In recent works, to derive the SRR, emissions of precursors are reduced by 15% and the simulation results were scaled up to represent the entire emission from an emitter (Nyiri et al., 2010). The

method was found to be more robust as the full removal of the source input caused non-linearity effects on SRR for highly reactive nitrogen compounds and O₃ through the photo-chemical chain reactions. Still the problem is usually of minor importance when sulfur compounds are considered, and thus, we utilized Eq.(1)."

(Comment #6)

For the simulations a 3-day spin-up period has been applied, but this may not be sufficient to provide realistic aerosol phase concentrations in the model. One has to realise that aerosol phase compounds like sulphate may have an atmospheric lifetime of up to 10 days in case the air mass does not meet a precipitation event. The work should therefore include a sensitivity analysis of the importance of length of spin-up period and initialisation procedures. If possible it would have been useful for the simulations to be initialised with course scale model results from model calculations covering the entire Asia region.

(Answer #6)

Yes, as you pointed out, 3-day spin-up period is not enough for longer lived secondary components such as O₃ and aerosols. However, we set the short spin-up period for the beginning of the one-year simulation so the initialization problem is not very serious for the whole year simulation. We added the following sentence after the first sentence of section 2.2, ln.17 of p.30094:

"A 3-day spin-up period is usually insufficient for longer-lived secondary components such as O₃ and aerosols. However, as the integration period of the simulation was the one year, the initialization problem may not be serious for the whole simulation."

(Comment #7)

It seems that the simulations have been performed having no seasonal and no diurnal variation in the applied emissions, which is rather odd given that simple assumptions may very easily be applied. Such assumptions where applied already in the early EMEP model calculations in the 1980ties. Since this would improve the results, the simulations should be carried out again applying such assumptions. Higher emissions during winter would lead to slower conversion from sulphur dioxide to sulphate which would change the overall results.

 (Answer #7)

Thank you for your advice. We inserted the following paragraph at the end of section 4 at ln.1 of p.30109:

"Because seasonal variation of anthropogenic species was not provided in our emission inventory, we didn't use it for the simulation. Even though nonlinearity effect is less significant for sulfur chemistry as discussed in section 4.2, some nonlinearity effect may be possible, e.g.

higher emissions during winter lead to slower conversion rate from S(IV) to S(VI), resulting in non-linear changes in the SRR. Still, as the most contributing sectors for Asian SO_2 emission were industry and power generation, which accounted for about 80% of the total SO_2, the seasonal variation was not very substantial (SO_2 in winter is 25% larger than that in summer; Streets et al., 2003). Thus, the seasonal variation in SO_2 emission will not cause highly non-linear impacts on the SRR of sulfur in this simulation. Under the LTP project, we are now heading to investigate SRR of other reactive and highly nonlinear components such as nitrogen compounds, O_3 and PMs among the Northeast Asian countries, by following the EMEP modeling works (Nyiri et al., 2010). In this case, it is indispensible to implement temporal variations in emission fluxes and to utilize 15 or 20% reduction method instead of full reduction for the accurate assessment of SRR for those reactive components"

(Comment #8)

The authors state that most of this conversion takes place in cloud and rain droplets, but generally it has been found that this accounts for half of the conversion whereas the other half is through reaction with OH radical. In case this picture has changed or it looks different for Asia compared with Europe, this should be documented in the paper.

(Answer #8)

The word "most of" was not appropriate. There is no evidence that "most of" the conversion takes place in droplets. We modified the sentence at ln.11 of p.30102 to "because the conversion from SO_2 to sulfate takes place efficiently in cloud and rain droplets."

Specific comments:

(Comment #8)

Page 30090 line 13 – the term "domestic origin" is unclear and should be specified

(Answer #8)

Thank you for your comment. I rephrased "domestic origin" as "originated from the same region", or just deleted the term in lns.13 and 14 of p.30090, lns.2 and 3 of p.30110. Also rephrased "domestic contribution" as "self contribution" in section 4.3 with a wording of "(defined as contribution of one source region to the same receptor region) in lns.10-11, 22, and 29 of p.30104.

(Comment #9)

Page 30091 line 12 – the term "fair and accurate emissions inventories" is very strange. "Fair" in what respect? The inventories are hopefully derived using stringent procedures and guidelines

and using the best available information.

(Answer #9)

We meant "fair" as "e.g. not to intentionally underestimate the value to avoid public accusation or inter-governmental conflicts" or something like that. We simply deleted the word as it is just a scientific paper.

(Comment #10)

Page 30092 first block – it is questionable whether this section is necessary as it is talking about SRR for persistent compounds like PAHs when the current paper is on sulphur

(Answer #10)

We modified the block accordingly together with the next of next comments. The modification is described in detail later.

(Comment #11)

Page 30092 line 1 the term "rarely focused" is unclear – do the authors mean "poorly addressed"?

(Answer #11)

Yes, we rephrased the words.

(Comment #12)

Page 30092 line 2 – talking about sulphur as a potential hazardous compound for the oceans is a bit odd as this is a natural constituent of the ocean.

(Answer #12)

We intended that the emission source of sulfur and PAH are similar in China (coal) so SRR over ocean of S could be an indication for SRR of PAH. As you mentioned, this paper focused on S and not on PAH. So we modified the statement in the first block of p.30092 as follows:

- We showed SRR of S over not only land but also over ocean, as it is informative for the budget study

- And SRR over ocean is important for PAH.

Discussion of PAH was refrained later in some places in the manuscripts. As it is not a paper for PAH study, we excluded the corresponding sentences in the 1st paragraph of section 4.4 (ln.10-15, p.30105) and 3rd paragraph of section 5 (ln.21-22, p.30109).

(Comment #13)

Page 30092 line 3 /diasel exhaust/diesel exhaust/

(Answer #13)

We deleted the sentence including the word.

(Comment #14)

Page 30092 line 23 /amounts/loads/

(Answer #14)

The phrase "deposition amounts" are found to be used as frequently as "deposition loads". So, I didn't change the phrase. Thank you for your suggestions, anyway.