Review of ACPD article: “Spatial distribution of source-receptor relationship of sulphur in Northeast Asia” by Kajino et al.

1) Yes, the paper addresses a very relevant scientific question for ACPD
2) The concepts and tools are generally fitting the purpose, but there are potential problems in how the authors handle initial and boundary conditions for their model simulations
3) Conclusions are fine
4) There are some problems with the missing parameterisation of seasonal and diurnal variations in emission inventories
5) Generally the interpretations of results are fine, but see (2)
6) Description of setup is very well outlined
7) Credit is given to relevant literature
8) Title is fine
9) Abstract is fine
10) Overall structure is fine
11) Language generally fine, but some wording should be rephrased
12) Formulas are fine
13) Some clarification is needed see below
14) References are fine
15) The authors should have attached paper under review regarding emission inventories for the review of the present paper

General comments:

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.

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.

Apparently SO_2 concentrations are well reproduced by the model, whereas SO_4^{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, H_2S, 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.

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?

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.

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.

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. 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 documented in the paper.

Specific comments:

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

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.

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

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

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.

Page 30092 line 3 /diesel exhaust/diesel exhaust/

Page 30092 line 23 /amounts/loads/