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Atmospheric Chemistry and Physics Discussions

Interactive comment on "A refinement of the emission data for Kola Peninsula based on inverse dispersion modelling" *by* M. Prank et al.

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Let us first thank the reviewer for comments and suggestions. Below is the first response to the criticism. The comments themselves are quoted.

"The general outcome of the exercise is rather moderate. Modelled SO2 levels still underestimate observations by a factor of 2 to 3. This is a major problem in the presented analysis of the signal from the sources on the Kola Peninsula. The paper does not present an analysis of the agreement between boundary SOx levels observed and modelled, and therefore it is not clear whether such discrepancies are due to the uncertainties in inventories for the Kola Peninsula."

Indeed, the remaining under-estimation is substantial. However, it has to be split to:

a missing background (DMS, possibly boundaries, Arctic ship traffic) and the major peaks. The background missing from the computations is a fraction of a ug S m-3 (fig.8). It is clearly distinguishable from the peaks, represent the small fraction of the load and thus left outside this paper. According to Fig.10 and related discussion, the large fraction of the missing peaks still originates from the Nikel plant area, i.e. the correction is indeed conservative.

The final agreement pattern is more homogeneous than before the correction: there is little dependence of the relative bias (see average values in table 5) on distance from the Nikel plant. The stations with the worst disagreement – a factor of 5-10 – were brought to within a factor of 2-3 as the others.

The next step would require review of the regional totals. Just source rearrangement would not help any more. Another approach is needed: bottom-up emission inventory based on correct activity data.

"The authors state that uncertainties in DMS emissions are minor sources of uncertainty in the analysis - but this is not shown in the analysis and the statement may be questioned. Another potential source of uncertainty may be related to ship emissions that is a major source of SOx."

The ship emission in Baltic Sea is included into the computations. The one in Arctic Ocean is not but the traffic there is low. The DMS emission in the Arctic is very low (e.g., Korhonen et al, 2008 and references therein, as well as the background level in the example of fig.8) and homogeneous, that is why their input was not considered. The reference will be added.

"The applied SO2 to SO42- conversion rate of 4 to 5% sounds very high. Usually values in the order of 2 to 3% are reported, and this is taking place far to the north where one would imagine even lower conversion rates. Such a high conversion rate therefore demands more argumentation."

The chemical mechanism and its evaluation have been published by Sofiev (2000) where the agreement of SO2 and SO4= predictions was estimated for the EMEP network over 6 years (albeit with different dynamic part of the model). The clarification will be added to the revised paper version.

"The applied 20% fraction of SOx emitted as SO42- is very high and the analysis points at even 30 to 70% direct emitted sulphate. This analysis may, however, be questioned as this may be due to problems with the initial concentrations on the boarder of the model domain. The paper does not present any data on how well the initial SOx value fits with observations."

Thanks for the correction. We will review this part of discussion to better reflect the sources and levels of the corresponding uncertainty.

"It is not clear what kind of data assimilation that has been performed in this study!?"

There was no data assimilation during the forward model runs. Technologically, the approach to emission correction is close to 4D-VAR, i.e., it uses adjoint dispersion simulations to obtain the sensitivity distribution for the cost function to the model-measurement disagreement. However, we did not follow the strict 4D-VAR procedure due to limited amount of observations.

"The authors put emphasis on the use of ensemble modelling, although the computations are performed using tools with differences regarding only transport description and meteorology. This is not the common use of the ensemble principle."

The applications of the ensemble idea are very diverse and depend on the problem. The ensembles comprised of different models or using different inputs are most frequently used in AQ, while the NWP applications usually rely on perturbations of initial fields. These differences refer to the specifics of the applications: the AQ computations are largely dependent on the meteo input data, which is thus an easy way to generate the ensemble.

- Style and specific suggestions regarding the formulations and individual paragraphs: Thank you, the text will be corrected in the revised paper version

Reference

Korhonen, H. Carslaw, K.C, Spracklen, D.V. Mann, G.W., Woodhouse, M.T. (2008) Influence of oceanic dimethyl sulfide emissions on cloud condensation nuclei concentrations and seasonality over the remote Southern Hemisphere oceans: A global model study. JGR, 113, D15204, doi:10.1029/2007JD009718.

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