Response to Referees Comments

AR: Authors' response

Anonymous Referee #2

The manuscript presents the comparison of the trends of sulphur and nitrogen wet deposition from 4 CTMs to the observations of EMEP Network. The results of the study are of interest because such models are commonly applied to model the impacts of future emission scenarios, creating a need for the knowledge of their reliability at reproducing the trends observed due to past emission changes. The paper is written in good English. The methods are well described and seem sound. However, the major shortcoming of the paper is the tendency to flood the reader with too much minor detail, making the reading tedious. I think the main messages could be delivered better by substantially cutting the length of the paper and leaving majority of the specific details about the skill of the individual models in tables instead of the main text, especially as the authors state that providing in depth analysis of the models' performance or inter-model differences it is out of the scope of their study.

AR: We thank the referee for their constructive comments. In the revised manuscript we have shortened the analysis by removing references to the performance metrics of individual models except where we want to highlight the performance of a particular model, for example a model that gives a large bias.

Specific comments:

1. Page 2, line 23. Why specifically semi-natural vegetation?

AR: This term is frequently used to refer to non-managed or extensively-managed ecosystems (e.g. woodland, moorland, meadows, mountain habitats etc.) in recognition that there are very few ecosystems in Europe that have not been directly influenced by human activity. To avoid confusion, we have changed this to "natural and semi-natural" [Page 2, line 22]

2. The analysis of previously published trends in observational data is currently cut to two parts (before and after the CTM results), making the structure of the introduction confusing and prone to repetition. This text includes too many details and all the specific numbers from all these studies would be far better visible and understandable if presented as a table.

AR: In the revised manuscript we have unified the two parts and reduced the size of this section, including only a summary of the (range of) trends estimated from observations without listing the trends from each study.

3. The overview of previous model-measurement comparisons could also be substantially shortened, as naming the specific models participating in those studies does not provide extra information, with the possible exception of if these are the same models as used in this study and this information is later used for discussion. I would suggest to try to compress this information into a few sentences per species, giving the general view whether the previous studies have shown any consistent under- or overestimation of its wet deposition. Or, if needed, including a supplementary table with the detailed numbers from these studies.

AR: The results of the previous model-measurement comparisons have been summarised to remove the detail and highlight the variability of model performance for wet deposition estimates as well as give an indication of tendencies of models to under- or overestimate wet deposition (median values of normalised bias for each species)

4. Please provide the reason why the 21-year period was divided to two 11-year subperiods.

AR: This was done to be able to calculate trends for two ten-year trends (1990-2000 and 2000-2010). This has been clarified in the revised manuscript. In addition, the emission trends over the 1990s are larger than over the 2000s and so deposition trends may be non-linear for over the full simulation period.

5. Were the NOx and NH3 emissions from wildfires included? How about SO2 from volcanoes?

AR: Emissions from wildfires were not included in any model and volcanic emissions of SO_2 were only included in the simulations by EMEP and MATCH. The following sentence has been added to clarify this "Emission from wildfires were not included and SO2 emissions from volcanoes were only included in the EMEP (Etna and Stromboli) and MATCH models." [Page 5, lines 29-31]

6. Could the CMAQ results be corrected for sea-salt sulphate (for instance using Na concentration in similar manner to how observations are corrected)?

AR: In CMAQ, marine sulfate is emitted directly in the coarse fraction, so considering only $PM_{2.5}$ sulfate will give the total anthropogenic sulfate. The evaluation of atmospheric concentrations has been modified so that total sulfate concentrations for CMAQ are calculated using the $PM_{2.5}$ sulphate concentrations, not the PM_{10} concentrations as done for the other models. Unfortunately it is not possible to separate out the contribution of sea-salt sulfate to WSOx in a similar way because the modelled contributions from $PM_{2.5}$ and PM_{10} are not provided separately. However, the non-corrected observations are available and so now the WSOx estimated by CMAQ are evaluated using the non-corrected data. The methods and results have been updated accordingly.

7. The description of emission changes could be shortened, for instance combining what happened to shipping emissions of both NOx and SOx into a single sentence and reducing the listings of specific countries and values.

AR: This section has been shortened by removing specific details of the emission trends and combining the description of shipping emission trends for NOx and SOx, as suggested by the referee

8. Spatial distributions are compared for 3 years (1990, 2000 and 2010) - are the differences in the patterns between these specific years representative of the overall trends?

AR: These years were not chosen to be representative although they do show fairly representative changes for situations with large trends (e.g. estimates of WSOx). These three years were chosen simply because they were the years that were simulated by all models

9. The paper could be shortened by skipping naming the models which simulated the largest and smallest results in majority of occasions apart from those few where the reason for the outlying model result is given.

AR: As mentioned above, in the revised manuscript we have shortened the analysis by removing references to the performance metrics of individual models except where we want to highlight the performance of a particular model, for example a model that gives a large bias.

10. Page 11, lines 28-31: If the emission data was given at 5-year interval and interpolated between the given years, the models cannot be expected to perfectly reproduce year-to-year variability which might result from instant changes in some emission sources due to closing of some facilities or implementation of emission control measures.

AR: This is true and it is one of the limitations of this emission dataset. However, with regards to the modelled deposition estimates, the inter-annual variability due to the meteorology is expected to be larger than that due to the emissions although, this may not be the case at certain locations due to the issues mentioned by the referee

11. Page 19, lines 27 – page 20, line 1 - "the net effect of these uncertainties is not expected to be a large systematic under- or overestimation of wet deposition." Due to the highly soluble nature of the compounds discussed here relatively little precipitation is needed for almost complete removal of them from the below-cloud column, leading to strong non-linearity of the wet deposition process. Thus, errors in modelled rain frequency might be more relevant for modelling the wet deposition than the annual precipitation amount and too frequent light rains instead of a few strong ones can for instance easily lead to positive bias in wet deposition.

AR: This is a good point and one that we have identified during our analysis as a subject of future evaluation studies. Doing this properly would require an analysis of the hourly measured and modelled precipitation and wet deposition (where available) for each model. This analysis is out of the scope of the current evaluation, which focuses on accumulated annual wet deposition and its trends over a twenty year period. We have highlighted this subject in the revised manuscript as a focus for future studies with the following sentences "In addition to the uncertainties in annual accumulated precipitation, the departure of the hourly, daily and monthly modelled precipitation from the observed values could lead to large errors in the modelled wet deposition for some models in some locations. The assessment of this effect would require an analysis of the hourly observed and modelled precipitation, atmospheric concentrations and wet deposition and should be considered for future analyses" [Page 17, lines 4-8]

Technical corrections:

1. Table 1. The optimal value of the geometric variance should be 1.

AR: This has been corrected in the revised manuscript

2. Figure 4 – Please correct the caption - the three time periods 1990–2000, 2000–2010 and 1990–2010 (left, middle and right columns) seem to be actually 1990–2000, 1990–2010, and 2000–2010

AR: The columns in the revised figure are now in the same order as the labels in the caption with the two 11 year time periods followed by the 21 year time period