Response to Reviewers' Comments: Global Nitrogen and Sulfur Deposition Mapping Using a Measurement-Model Fusion Approach

We thank all the reviewers for their insightful comments. In general, the three reviewers raised a number of methodological issues, all of which are valid in their own right, but also a few that are somewhat beyond the scope of the current manuscript. As now more explicitly mentioned in our introduction, the main purpose of our study is to demonstrate the viability of a straightforward but globally applicable MMF approach while remaining consistent with previous work that provides impact assessments for various communities. As also outlined in our previous paper, the introduction of measurement-model fusion approaches for deposition entails a host of issues and possible approaches, some of which have been demonstrated on regional scales in Europe and North America. However, MMF for deposition has thus far not been applied on global scale, for a variety of reasons, but the most important one is the absence and heterogeneity of available data sources. The World Meteorological Organization (WMO) has recently commenced an activity (MMF-GTAD) that sets the roadmap for improvement of deposition datasets. Our manuscript intends to use a relatively simple method that demonstrates the potential of MMF for Global Nitrogen and Sulfur deposition- a necessary building brick for further progressing the WMO activity. Keeping in mind the overall goal of our paper, we address suggestions individually below.

Reviewer #1:

The causes for major revision are: in this work, observed wet deposition is fused with modelled wet deposition. The more common method is to fuse the concentration in precipitation, not wet deposition in itself. Precipitation and wet deposition have a larger variation in space compared to concentration in precipitation. Fusing the latter would allow for a longer length-scale in the fusion. Precipitation should be applied after the fusion of concentration in precipitation.

Response: We thank the reviewer for the comment. While we agree with the reviewer that it in regional studies it is common to fuse the concentration in precipitation, for a consistent approach in our global study this would involve a resource intensive activity of merging precipitation fields and rain concentration fields in global models. Therefore, as a first step, and also to connect to previous global deposition studies, we opted to use a gridded ensemble mean wet deposition dataset created from 11 HTAP-II models. Unfortunately HTAP-II does not provide independent gridded datasets of precipitation and concentration. Within the scope of our current study (see above) we think this is a reasonable method, while the method described by the reviewer will be explored in future work under the WMO MFF-GTAD umbrella.

Here the grid resolution (1 degree) was used as maximum length-scale, which is very short (too short) and will cause for in principle only one or a few grid boxes to be influenced by the observation. There was no scientific explanation to the choice of this length-scale, which can be considered too short. The fusion method explained here, was rather used to estimate the observation error within the gridbox, but is that really how it should be done? Is the observation error dependent on the distance of the observation to the middle of the grid box?

(answer is NO). The grid centre in the model is not the actual centre but an average of the whole grid.

Response: The reviewer raises an important issue. The optimal length scale for inverse distance interpolation of deposition fluxes will depend on multiple factors, including the distribution of the emissions, transformation by atmospheric chemistry, the distribution and intensity of rainfall and the associated removal time scales. The corresponding spatial scales will therefore vary from <100 km to several 100s of kms. We have added a sensitivity analysis where we compare the results of changing the interpolation distance. See Tables S2 and S3 and Figures S2-S4 and 3 (shown below). We fully acknowledge that IDW is not the best measurement-model fusion method, but our aim is to apply globally with all available measurements a method that is already verified and in use officially in the National Atmospheric Deposition Program (NADP) of the US.

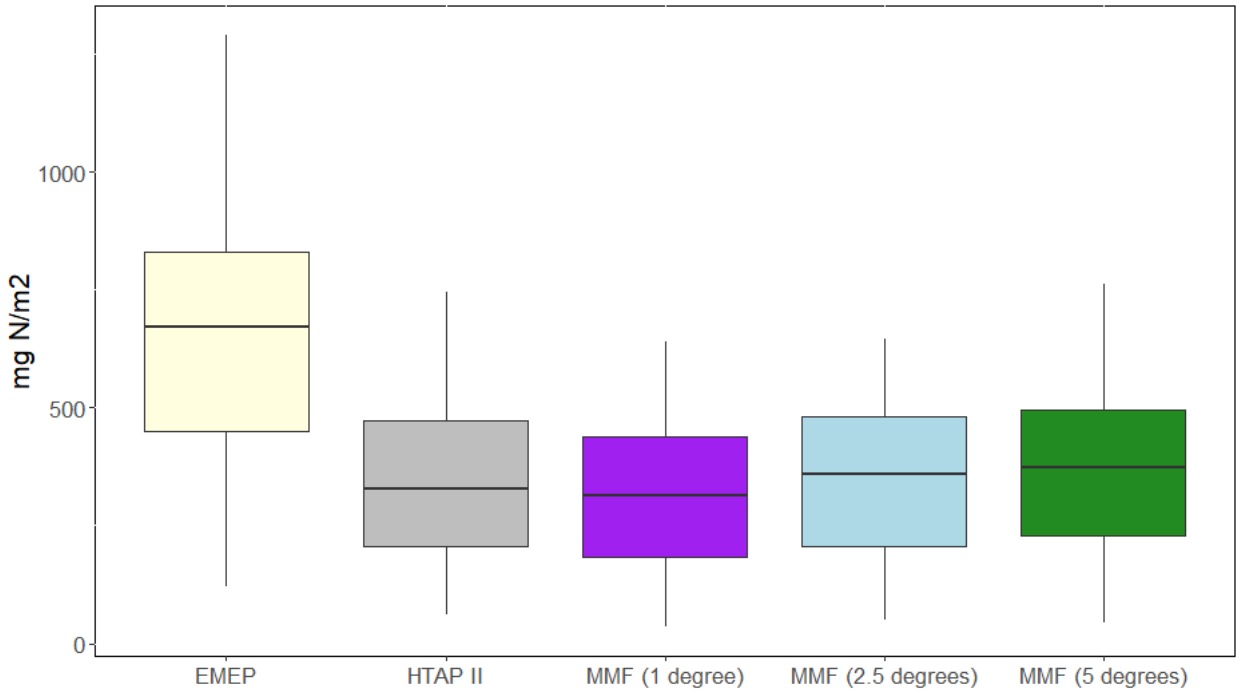

Figure 3: A comparison between HTAP II, MMF, and EMEP results at EMEP observation sites. A boxplot shows the distribution of EMEP, HTAP II, and MMF modeled wet reactive nitrogen deposition (NH_x and NO_y) results at each EMEP observation location. Three different interpolation distances are compared using MMF, 1 degree, 2.5 degrees, and 5 degrees.

The final product shows very little influence from the observations, which is not reasonable to my experience, and a result from the erroneous method. The figures show that the MMM (pure

model) and MMF are very similar also in places with dense observations, and the MMF has a large deviation to observations (see e.g. the bias in deposition in China and US).

The comparison to observations is not clear whether it is by independent or dependent observations, but in whichever case the comparison shows also too weak influence from the observations in the MMF product. It is strange that even in the specific grid box of the observations, the MMF deviate very much to the observed deposition.

For these reasons I recommend that the authors revisit their methods, to improve the MMF results before resubmitting.

Response: While we acknowledge that IDW has some drawbacks, our goal with this paper is to demonstrate that a global measurement-model fusion approach is feasible and can be performed using simple methods. Future work absolutely should investigate other MMF approaches if there is enough or large enough observation data available, but that is beyond the scope of this paper.

We have increased the interpolation distance to 2.5 degrees to address the lack of influence from measurements and added figures to the supplement to demonstrate the impact of either increasing or decreasing this distance. This is now shown in Figure 4, below. And a sensitivity analysis has been added (Figures S4, S5) with a 1 degree and 5 degree interpolation distance. This has changed our results and all totals, tables, and maps have been updated accordingly.

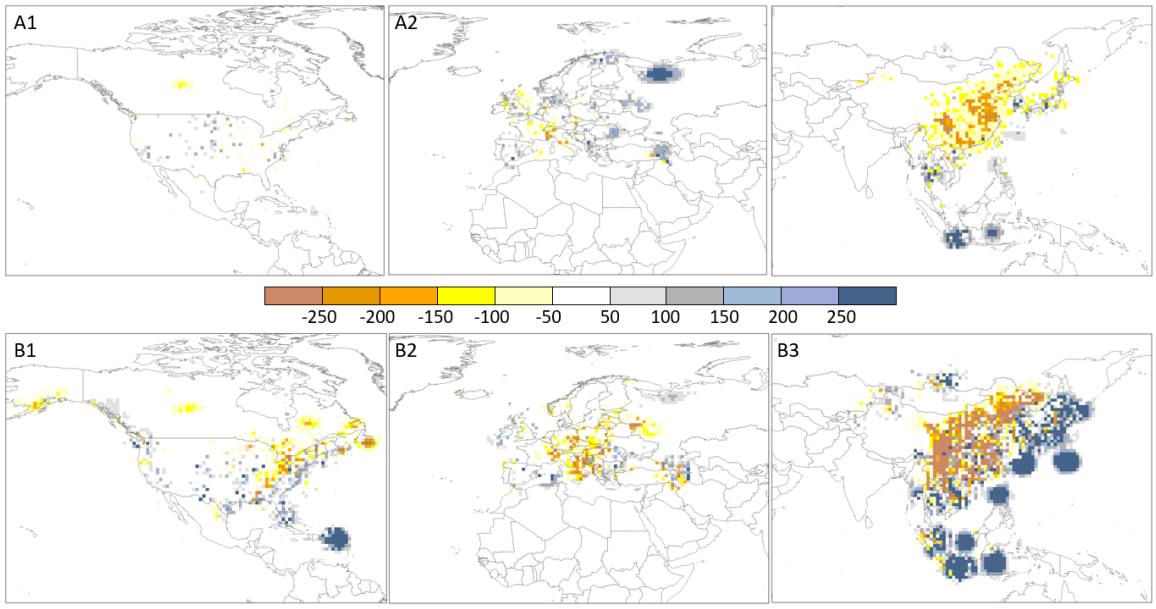

Figure 4. The difference between MMF and MMM deposition with a 5-degree interpolation distance. **A)** MMF minus MMM reactive nitrogen deposition in North America (**A1**) Europe (**A2**) and East Asia (**A3**) in mg/m^2. **B**) MMF minus MMM sulfate deposition in North America (**B1**) Europe (**B2**) and East Asia (**B3**) in mg/m^2.

Row 112-116. I suggest not to include datasets in the manuscript that are only promising. There are likely many promising national datasets in the global arena that potentially could be used, and to include all that are not used will be a paper in itself.

Response: We thank the reviewer for the suggestion. We understand the concern with a "promising" dataset and it was not included in the study because the data only include a few months of the year 2010, while in following years completely annual coverage is provided. It is now mentioned in the discussion to provide context for the regional dataset we did include and to suggest that future work could incorporate those measurements that are publicly available.

Row 136: title: please state MMF procedure

Response: We thank the review for the comment. We changed the title to "MMF Procedure".

Row 165-166: change to "… include measurements from Asia, Europe and North America, and the dry deposition MMF surface includes measurements from the USA and Asia, …". Explanation: many parts of the world are not covered for wet deposition either. The phrasing was now overenthusiastic about the coverage of wet deposition observations.

Response: We thank the reviewer for the suggestion. We have changed the phrase as suggested to reflect the lack of worldwide measurements.

Table 1. row open oceans has values in "non-coastal" but not in "coastal". This does not seem correct to me, it should be the other way around. Are the columns mixed up?

Response: The columns are not mixed up; open ocean values are not "coastal" in that they are not near land. A sentence was added to the caption for Table 1 to clarify:

"Open ocean does not include near-land "coastal" waters."

Figures: in general – please label panels a-f etc, it is easier to understand the description if all panels are referred to and labelled.

Response: We have added labels to all graph panels, as suggested.