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GLOBAL NITROGEN AND SULFUR BUDGETS USING A MEASUREMENT-MODEL FUSION APPROACH

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1. INTRODUCTION

Atmospheric nitrogen and sulfur deposition from human activities related to the use of fossils and land use have significant implications for ecosystem and human health (Bobbink et al., 2010). Oxidized nitrogen (NO_y) and reduced nitrogen (NH_x), together called reactive nitrogen (Nr), and oxidized sulfur (SO_x) deposition occur as wet and dry processes. The United Nations Economic Commission for Europe's Task Force on Hemispheric Transport of Air Pollution (HTAP) is an international effort to improve the understanding of air pollution transport science with emissions models. The second phase of HTAP was launched in 2012. Tan et al. (Tan et al., 2018) used the multi-model mean (MMM) of 11 HTAP II chemistry transport models to estimate the sulfur and nitrogen deposition budgets for 2010. Significant uncertainty remained due to a lack of station measurements, especially in East Asia, a large contributor to the overall budget. Combining measurements and model estimates in a "measurement-model fusion" (MMF) approach has the advantage of retaining the broad spatial coverage of models while accurately matching observations. Generally speaking, MMF takes model estimates for a region and modifies them based on in-situ point measurements of the phenomenon to "nudge" the model towards the observed values (Labrador et al., 2020).
More details of the MMF approach are described in Fu et al. (Fu et al., 2022) as they lay out a roadmap for future work, following the World Meteorological Organization's Global Atmosphere Watch Program (WMO GAW) and the intended role of the MMF Global Total Atmospheric Deposition (MMF-GTAD) project. This study updates Tan et al.'s (Tan et al., 2018) global S and N deposition budgets using a variation of the TDep methodology (Schwede & Lear, 2014) to merge NH_x, NO_y, and SO_x gridded surfaces from modeled results with observations of NO_3^-, NH_4^+, and SO_4^{2-} in precipitation and as an aerosol. We demonstrate the viability of a straightforward but

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globally applicable MMF approach while remaining consistent with previous work that provides impact assessments for various communities. This approach is an essential step towards the WMO's goal of reliable deposition products to aid decision-making. We update the 2010 deposition budgets using MMF to combine the broad spatial coverage of a model with accurate in-situ measurements.

2. DATA AVAILABILITY

All data are from 2010, collected monthly and all measurements were converted to mg-N (or S) /m^2. Wet deposition measurements (NO_3^-, NH_4^+, and SO_4^{2}) from the US's National Trends Network (NTN) and Atmospheric Integrated Research Monitoring Network (AIRMoN) are available through the National Atmospheric Deposition Program (NADP, http://nadp.slh.wisc.edu/NTN/). Dry deposition generated values are available from the Clean Air Status and Trends Network (CASTNET) at 84 locations. Nitrogen and sulfur wet deposition measurements and dry deposition estimates throughout Canada are recorded by the Canadian Air and Precipitation Monitoring Network (CAPMoN). The European Monitoring and Evaluation Programme (EMEP, http://ebas-data.nilu.no/) has records of precipitation chemistry (NO_3^-, NH_4^+, and SO_4^{2-}) for Europe. A multi-year nationwide field study was compiled by Li et al. (Li et al., 2019). EANET (https://www.eanet.asia/) wet and dry deposition and precipitation data are available at 47 sites.

3. METHODS

Global yearly wet and dry NO_3^-, NH_4^+, and SO_4^{2-} deposition observations (for wet deposition) or estimates (for dry deposition) were combined with the respective HTAP II model average grid cell estimates, using common 1 degree x 1 degree (1º x 1º) grid cells (Fig. 1). For example, wet NO_3^- deposition observations are combined with the wet NO_3^- modeled deposition in the nearest HTAP II MMM grid cell to the observation, where observations exist. Dry deposition values (NO_3^-, NH_4^+, and SO_4^{2-}) from CASTNET and n inverse-distance weighted 1º x 1º gridded dataset was created based on the distance from each observation to the center of the nearest HTAP II model grid cell. Inverse-distance weighting (IDW) was selected as the most implementable method to introduce MMF on a global scale while remaining consistent with previous work (Schwede & Lear, 2014).

The weighting function was calculated as

$$\left(1 - \frac{distance}{max\ distance}\right)^2 \qquad (1)$$

following Schwede and Lear's (Schwede & Lear, 2014) approach for the TDep product, where "distance" is the distance between the site location and the center of the HTAP II model grid cell nearest to that sampling site location, within a maximum distance of 1º (approximately 111 km at middle latitudes). This maximum distance was chosen because that is the resolution of the HTAP II grids, not because that is the distance a species might travel in the atmosphere before it is deposited. The output values of the weighting function at each observation location are then multiplied by the observed deposition. For the center of every HTAP II model grid cell near that site, the modeled deposition is multiplied by 1 minus the value of the weighting function. As a consequence, if there are no observations near a model grid cell, the cell value remains the same. The two grids ([weighting function times observed deposition] and [1-weighting function times modeled deposition]) are added together. This has the effect of modifying the HTAP II grid only in locations where there are observations nearby. The MMF gridded surfaces were then summed by species along with the remaining unchanged HTAP II gridded surfaces that lacked in-situ measurements to create total N and S deposition gridded surfaces (e.g., the MMF wet and dry SO_4^- gridded surfaces were added to the HTAP II wet and dry SO_2 gridded surfaces to get total S deposition).

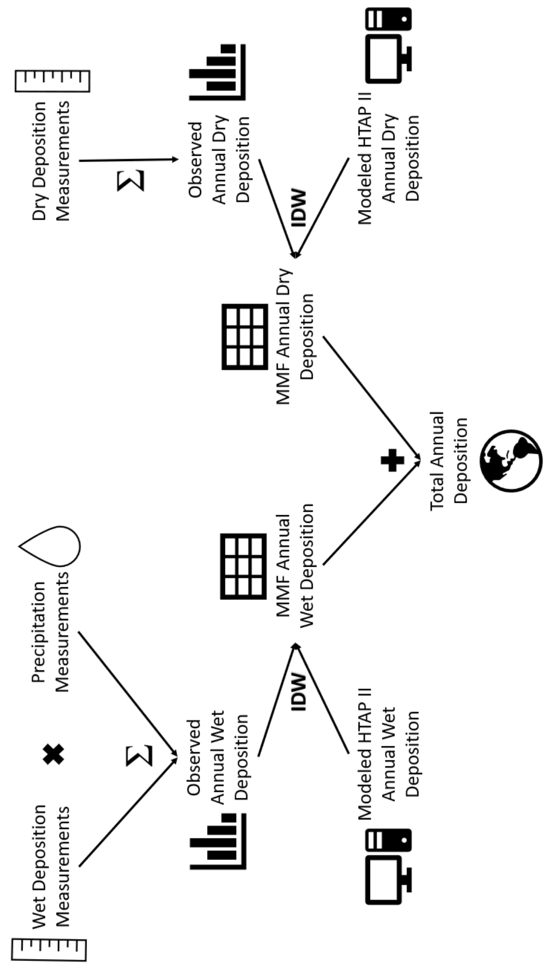


Fig. 1: A flowchart describes the MMF methodology implemented in this paper.

4. RESULTS

The total global NH_x deposition in 2010 is adjusted from 54 Tg-N (from HTAP II models) to 70.65 Tg-N. Combined with a NO_y deposition of 59.4 Tg-N (from a modeled HTAP II 59.3 Tg-N), the total global deposition is adjusted to 130 Tg-N (from 113 Tg-N). Most of this increase comes from a model underestimation in East Asia. Total S deposition is adjusted to 80 Tg-S, a slight decrease from the HTAP II model prediction of 83.5 Tg-S.

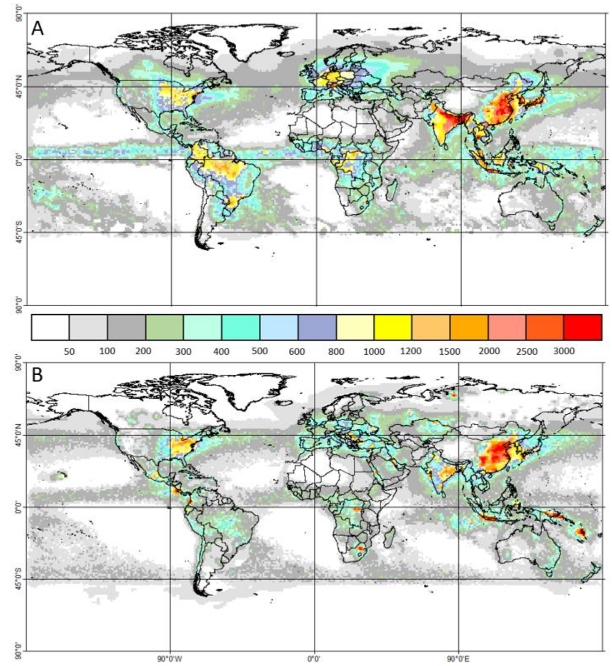
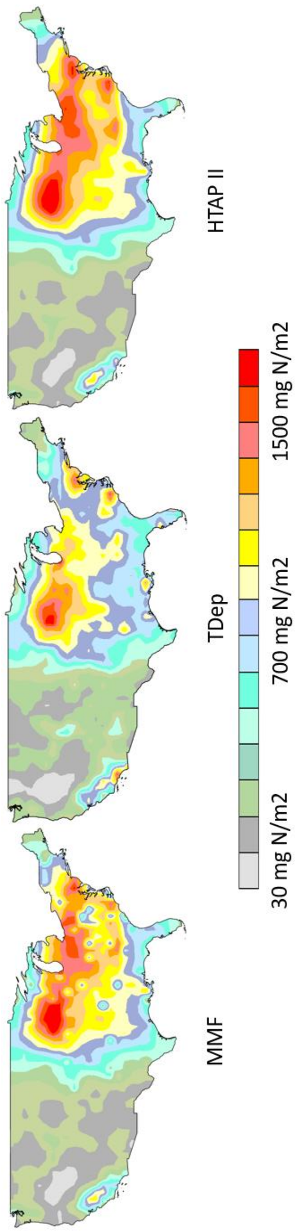


Fig. 2: A) Total N deposition (mg N/m^2), the sum of wet and dry NO_3^- and NH_4^+ after applying the MMF approach, as well as HTAP II gridded surfaces of wet and dry NH_3, HNO_3, and NO_2. B) Total S deposition (mg S /m^2), the sum of wet and dry MMF SO_4^{2-} and wet and dry HTAP II SO_2.

The spatial distribution is different, with more deposition in coastal areas in the MMF estimate Tan et al. (Tan et al., 2018) report that the HTAP II MMM overestimates NO_3^- wet deposition in North America, but underestimates NH_4^+ deposition. We find that the MMF interpolated deposition slightly improves these estimates, although the spatial distribution is very similar. The largest change for N deposition (comparing MMM and MMF) is in grid cells classified as ocean because of an increase in East and Southeast Asia deposition which mostly occurs in areas classified as ocean due to the small island size and low spatial resolution. Ocean cells were classified as such if they were located further than 1 degree from the mainland; therefore, any islands smaller than 1 degree were counted as ocean. The largest change for S deposition is in continental grid cells due to a decrease in East Asia.

There are spatial differences between an aggregated 1º x 1º the original TDep map of nitrogen deposition for the United States as available from the NADP (Fig. 3A), the HTAP II surface produced by Tan et al. (Tan et al., 2018) corresponding to the same area, and the deposition map produced in this work. A similar pattern is seen in the map of SO_4^{2-} (Fig. 3B)



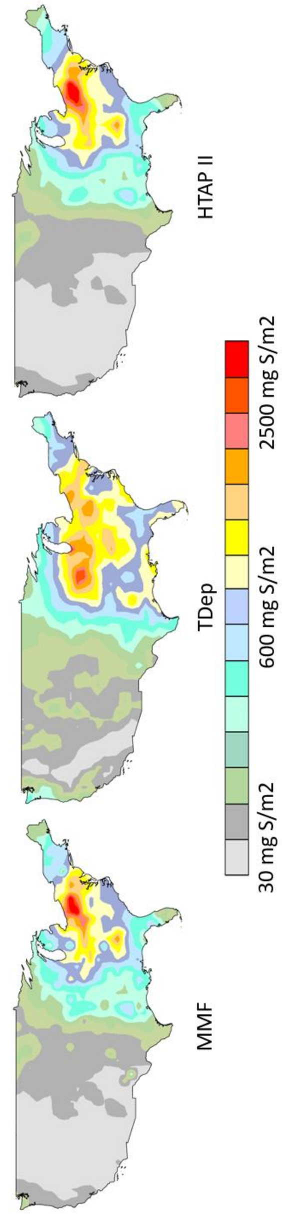


Fig. 3: 2010 Total N deposition in the US. A) Total N is modeled with 1) MMF (this work), 2) TDep annual map available from the NADP and 3) Tan et al.'s 2018 MMM. B) 2010 SO_x wet deposition in the US as modeled with 1) MMF (this work), 2) TDep annual map available from the NADP, and 3) Tan et al.'s 2018 multi-model mean HTAP II output.

The R^2 value for the linear regression between MMF wet NH_4 and observed wet NH_4 in the US is 0.76 (Fig. 4). The R^2 value for the linear regression between the HTAP II wet NH_4 and observed NH_4 is 0.66, and 0.92 for the linear regression between the TDep wet NH_4 and observed NH_4 (Fig. 4). All three datasets produce similar values to the measured wet NH_x deposition at the NADP/NTN sites (Fig. 4).

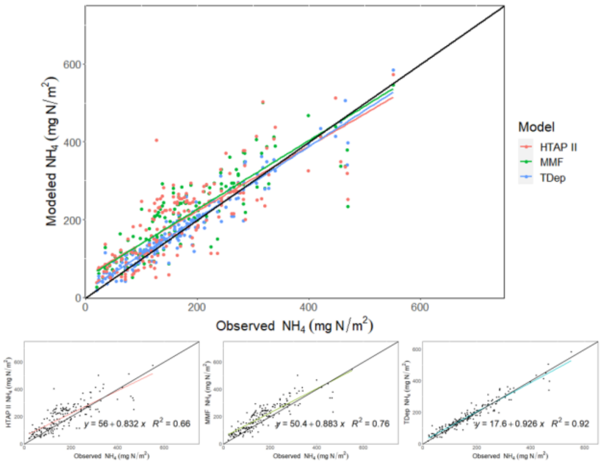


Fig. 4. Observed and modeled wet NH_4 deposition in the US in 2010. The black line is the 1:1 line.

5. DISCUSSION

While MMF does give better deposition estimates by incorporating in-situ measurements, it is worth considering the scale of the model. Observations of deposition are probably not representative for a 1º resolution and observations of precipitation may not be homogenous in all directions at that scale, especially over varying terrain. So, for example, the coarse resolution of the model, even with added measurements is likely not accurately capturing the gradient between coastal and inland deposition. The differences between the TDep, MMM, and MMF gridded deposition in the US (Fig. 3) are clear on the coasts. While the general patterns of deposition are similar for the three products, the magnitude of deposition in the aggregated TDep dataset (1º x 1º) is higher in the eastern US and lower in the western US than either of the other two deposition fields. This difference is likely due to the precipitation dataset used to calculate wet deposition. MMF deposition is based on the MMM dataset; therefore, both utilize the same precipitation dataset, from a combination of 11 global models. However, TDep wet deposition is produced by multiplying PRISM precipitation data and an interpolated gridded surface dataset of wet NH_4^+ concentrations. PRISM is a reanalysis product designed to interpolate precipitation in particularly complex landscapes using weather radar and rainfall gauge observations, though it is not identical to observations because it used long-term averages as predictor grids (Zhang et al., 2018). It captures much more localized variation in precipitation due to geographical variations which are not captured in the lower resolution global precipitation models used in the HTAP II MMM (Tan et al., 2018). The total deposition within the US borders is similar for the MMF, HTAP II, and aggregated TDep gridded surfaces; however, the spatial distribution is different.

TDep maps of North American nitrogen deposition created with Schwede and Lear's methodology (2014), using IDW, are widely in use and freely available from the NADP. However, there are limitations associated with IDW (Sahu et al., 2010), and other interpolation methods such as kriging or geographically weighted regression could provide smoother surfaces with fewer artifacts. IDW is a fast and flexible interpolation method, but it does not minimize error and can produce inaccurate results in regions with sparse measurements and large sub-grid variability. This problem is relevant to much of the world. The lack of measurement sites globally is a hindrance that can be alleviated by including remotely sensed observations. Future work should also investigate methods such as machine learning techniques with spatial information to avoid these limitations.

These results from measurement-model fusion are important because previous methods on a global scale have relied primarily on models (Vet et al., 2014). They compare their results with measurements, of course, in order to demonstrate the model capabilities but they do not explicitly incorporate point measurements into the final product. Our results serve to emphasize that the models are adequately simulating deposition (in terms of total deposition budgets) but that the regional discrepancies between models and measurements can still be quite large; and measurement-model fusion helps to ameliorate this without changing the fundamental model parameters and processes that capture the overall deposition reasonably well.

5.4 References

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