

Interactive comment on “Observation- and Model-Based Estimates of Particulate Dry Nitrogen Deposition to the Oceans” by Alex R. Baker et al.

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Received and published: 16 May 2017

Responses to Comments by Reviewer 1

This manuscript is relevant for publication in Atmospheric Chemistry and Physics. The authors compile extensive datasets of ocean based observations of nitrate and ammonium concentrations from aerosols. They then compare and contrast observed aerosol concentrations and calculated dry deposition fluxes to a global model (TM4) and the mean product from a global model intercomparison project (ACCMIP). There are limitations, both in how the data are treated and in the model–observation comparisons, making it difficult to draw new conclusions beyond what is essentially already known. Still this represents an important compilation of data, an important recognition of the

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state of the field and recommendations on needs for significant improvements to advance this science. The work is well cited and includes a table in the supplement of all data used in the compilation, which is also available on a public database via the SOLAS website. Below I outline areas of the manuscript that could be improved, however, overall I recommend publication of the manuscript with minor revisions. The primary weaknesses include: a lack of discussion of the influence of meteorology on results; a need for a more representative abstract; and a need for clearer recommendations to the community to improve and make advancements. Finally, this manuscript is an important review of the state of this field, and as such should address a few basics that will be important to the broad readership of AC&P.

Response: We thank the reviewer for their thoughtful and constructive comments on our manuscript. We set out our responses to these in detail below, together with proposed changes to the manuscript.

Abstract: The abstract could better represent the findings in the work. The last sentence of paragraph 1 (line 37-38) should mention how/why mineral dust alters deposition of N (thinking of a broader audience). The second paragraph should include some type of quantitative summary of the results.

Response: We have modified the Abstract to address the comments of both reviewers. For the point on dust see response to Reviewer 2.

It should be addressed here why the focus here is on comparison with TM4 (which is justifiable but why mention ACCMIP if not actually discussing the comparison here?).

Response: We focus the discussion on TM4 because of the availability of the calculated individual aerosol component concentrations and deposition fluxes (speciation), which is not the case for the ACCMIP data. In addition, TM4 model has a comprehensive representation of the N atmospheric cycle, including Fe redox reactions and organic nitrogen sources and fate (Kanakidou et al., 2012; 2016). On the other hand, ACCMIP deposition fluxes are the ensemble of several models and as such are a more robust

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model product than one model output. Therefore comparison of ACCMIP and TM4 results puts TM4 results in context. (For changes to manuscript, see revised paragraph of the Abstract below).

Indeed, the TM4 model over-estimates NO₃⁻ and underestimates NH₄⁺ - however, the comparison with NH₄⁺ is much better in all of the ocean basins. Can this be quantified/summarized more concretely here?

Response: In response to these comments we have added a more quantitative description of the model - observation comparison to the Abstract (see revised paragraph below) and further discussion has been added at the end of Section 3.3.

Revised text, Section 3.3: Note that over land, NO₃⁻ and NH₄⁺ levels are affected by the vicinity of the sources. In particular, biomass burning and dust emissions affect the partitioning of NO₃⁻ and NH₄⁺ to the aerosol phase. Even small inaccuracies in the model simulations of this partitioning can lead to higher discrepancies between model results and observations over land than over the ocean. Indeed, Kanakidou et al. (2016) have compared NO₃⁻ and NH₄⁺ concentrations in PM₁₀ over Europe and found an overestimate in NO₃⁻ PM₁₀ content of about 115% and an underestimate in NH₄⁺ in PM₁₀ of about 55% (Figure S4 in the Kanakidou et al. (2016) supplementary material), results that are consistent with, but larger than the 70% and 44% respectively reported here for oceanic regions (Fig. 4 of the present paper).

Also missing from the abstract is concrete suggestions or recommendations. The abstract makes it appear that little is concluded in this study beyond the clear limitations of our understanding of dry deposition velocities. What's needed to really address this or are there specific things the community should at least be worried about addressing in the near future? In other words, the abstract should address this a bit more to be more representative and garner community attention. At the very least, the clear recommendation that measured aerosol concentrations be reported for observation and models and that this be the key comparison that is made rather than dry deposition

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fluxes is an important point that belongs in the abstract.

Response: We have re-written the second paragraph of the Abstract to address the reviewer's comments (and those of Reviewer 2). In order to avoid overly increasing the size of an already long Abstract, we have chosen to draw the reader's attention to the existence of recommendations for improvements, but have only explicitly stated the conclusion regarding comparisons to modelled surface concentrations (as suggested by the reviewer).

Revised text, Abstract: Assessment of the impacts of atmospheric N deposition on the ocean requires atmospheric chemical transport models to report deposition fluxes, however these fluxes cannot be measured over the ocean. Modelling studies such as the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP), which only report deposition flux are therefore very difficult to validate for dry deposition. Here the available observational data were averaged over a 5° x 5° grid and compared to ACCMIP dry deposition fluxes (ModDep) of oxidised N (NO_y) and reduced N (NH_x) and to the following parameters from the TM4-ECPL (TM4) model: ModDep for NO_y, NH_x and particulate NO₃⁻ and NH₄⁺, and surface-level particulate NO₃⁻ and NH₄⁺ concentrations. As a model ensemble, ACCMIP can be expected to be more robust than TM4, while TM4 gives access to speciated parameters (NO₃⁻ and NH₄⁺) that are more relevant to the observed parameters and which are not available in ACCMIP. Dry deposition fluxes (CalDep) were calculated from the observed concentrations using estimates of dry deposition velocities. Model – observation ratios, weighted by grid-cell area and numbers of observations, (RA,_n) were used to assess the performance of the models. Comparison in the three study regions suggests that TM4 over-estimates NO₃⁻ concentrations (RA,_n = 1.4 – 2.9) and under-estimates NH₄⁺ concentrations (RA,_n = 0.5 – 0.7), with spatial distributions in the tropical Atlantic and northern Indian Ocean not being reproduced by the model. In the case of NH₄⁺ in the Indian Ocean, this discrepancy was probably due to seasonal biases in the sampling. Similar patterns were observed in the various comparisons of CalDep

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to ModDep ($RA_n = 0.6 - 2.6$ for NO_3^- , $0.6 - 3.1$ for NH_4^+). Values of RA_n for NH_x CalDep - ModDep comparisons were approximately double the corresponding values for NH_4^+ CalDep - ModDep comparisons due to the significant fraction of gas-phase NH_3 deposition incorporated in the TM4 and ACCMIP NH_x model products. All of the comparisons suffered due to the scarcity of observational data and the large uncertainty in dry deposition velocities used to derive deposition fluxes from concentrations. These uncertainties have been a major limitation on estimates of the flux of material to the oceans for several decades. Recommendations are made for improvements in N deposition estimation through changes in observations, modelling and model - observation comparison procedures. Validation of modelled dry deposition requires effective comparisons to observable aerosol-phase species concentrations and this cannot be achieved if model products only report dry deposition flux over the ocean.

Section 2.2: Include units in defining the variables for Equation (1).

Response: We have added units for these variables, as requested by the reviewer.

Revised text, Section 2.2: Where possible, the observed aerosol concentrations (C : $nmol\ m^{-3}$) for NO_3^- and NH_4^+ were compared directly with corresponding particulate concentrations simulated by the models (i.e. for the TM4 model, see below). Dry deposition fluxes from the models were also compared to the observational database. In order to do so, dry deposition fluxes (F : $mg\ N\ m^{-2}\ d^{-1}$) were calculated from the observed concentrations of the two species using dry deposition velocities (vd : $m\ d^{-1}$) (Eq. 1), with appropriate correction for the relative atomic mass of N. (Note that hereafter we quote vd in units of $cm\ s^{-1}$).

Section 2.3 For each model description, it should be mentioned what is considered surface level in each model - i.e. what vertical resolution is the model output averaged over?

Response: It is not possible to give a unique surface level for the ACCMIP multi-model product, as the underlying models each have their specific coordinate systems. In

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general, the mid-level of the lowest layer in the models varies between 20-40 m. For TM4, the mid-level of the lowest layer is at 40 m.

Revised text, Section 2.3: TM4: The model's lowest level has a mid-level height of 40 m and its native resolution is 2° (lat.) \times 3° (lon.), but for this study the model output was interpolated to a grid scale of $1^\circ \times 1^\circ$. ACCMIP: The fields used were MMM dry deposition from 10 (for NO_y) or 5 (for NH_x) individual atmospheric chemical-transport models, generally with surface mid-level heights of 20 - 40 m, and were reported by ACCMIP on a grid scale of $0.5^\circ \times 0.5^\circ$, although the resolution of individual models was coarser.

Lines 141-142: replace "sigma" with 1 sigma or 1 std dev or similar

Response: These values were 1 sigma. The text has been updated accordingly.

Line 152+ : It is not discussed whether ACCMIP includes a flux of NH_3 from the ocean similar to TM4 and how much is emitted on an annual basis.

Response: Most of the ACCMIP models include a prescribed NH_3 surface flux following very old recommendations by the GEIA community from 1996 (Bouwman et al., 1997). The emissions amount to some 8 Tg NH_3 /yr. Much of these ocean emissions are recycled close to the sources, if no uptake by acidic aerosol is considered. This has been clarified in the text.

Revised text, Section 2.3: The ACCMIP products used in this comparison were based on emissions for the year 2000 and average meteorology for the decade 2000 - 2009 (Lamarque et al., 2013a). The fields used were MMM dry deposition from 10 (for NO_y) or 5 (for NH_x) individual atmospheric chemical-transport models, generally with surface mid-level heights of 20 - 40 m, and were reported by ACCMIP on a grid scale of $0.5^\circ \times 0.5^\circ$, although the resolution of individual models was coarser. NO_y and NH_x dry deposition estimates were also available for TM4. (Neither particulate concentration nor dry deposition fields were available for NO_3^- or NH_4^+ from ACCMIP). For both

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ACCMIP and TM4 model results, NH_x corresponds to the sum of NH₃ and NH₄⁺. Most models in the ACCMIP product included marine emissions of NH₃ based on Bouwman et al. (1997).

Is it possible to report what percentage of NO_y is NO₃⁻ in the ACCMIP products? In the comparisons this is brought up as a reason for disagreement between model and obs, but it would be interesting to note whether NO₃⁻ is a majority of the NO_y deposition or not.

Response: The percentage of NO_y in the ACCMIP product that is due to particulate NO₃⁻ deposition was not reported in the original publication of this product (Lamarque et al., 2013a). This was because not all of the models used to produce the ACCMIP multi-model mean specifically simulated particulate NO₃⁻ deposition (Lamarque, personal communication, 2017). The text has been amended to state this explicitly, and the implications of this for the ACCMIP NO_y deposition flux estimates are also discussed.

Revised text, Section 2.3: Particulate NO₃⁻ was not simulated by all of the models contributing to ACCMIP, and hence the fractional contribution of NO₃⁻ to NO_y deposition was not reported by Lamarque et al. (2013a). In models without a specific simulation of particulate NO₃⁻, this species is likely to have been simulated as gas-phase HNO₃, whose dry deposition velocity is similar to that of particulate NO₃⁻ (Pryor and Sorensen, 2002). Thus, the dry deposition flux of NO_y in the multi-model mean was not greatly affected by this factor. The ACCMIP NO_y dry flux was not substantially different from that computed in the TM4 model, which does specifically simulate dry particulate NO₃⁻ deposition (Kanakidou et al., 2016). Therefore, in the present study, TM4 speciated results are more appropriate for comparison to the observations and are put in context when used jointly with the more robust, but less speciated, ensemble model results of ACCMIP.

Discussion: It would be useful to include some discussion on the influence of mete-

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orology. ACCMIP output is based on average met fields for 2000-2009. TM4 uses ECMWF. The calculated deposition for variable vd uses ECMWF wind speeds. But clearly, amongst all products and calculations, there is an important role of meteorology in determining deposition. Perhaps the variable vd could be calculated with another, different year of meteorology to give an estimate of how uncertain the influence is of a model-data met product on the calculations? While analyzed met fields are useful, their utility over the open ocean where direct measurements are limited may be an issue. This should be better addressed here.

Response: In fact, deposition velocities were calculated using mean ECMWF wind fields for all the years that observational data were available for (1995-2012). We have clarified this in the text (see response to Reviewer 2). We have added text to Section 4.1 to discuss the uncertainty introduced by the choice of year of ECMWF wind speeds and the uncertainty in ECMWF wind fields themselves, including a new figure (S8 in the revised Supplementary material) of deposition velocity relative standard deviation derived from the individual ECMWF wind fields for the years studied here (Fig. 1).

Revised text, Section 4.1: Uncertainty in analysed meteorology introduces uncertainty into deposition velocities derived for the variable vd CalDep calculation. This uncertainty was assessed by calculating vd from mean ECMWF wind speeds for each of the individual years (1995 – 2012) and the relative standard deviations of these annual vd values. Standard deviations were relatively high over the tropical oceans (up to ~25%) and lower elsewhere (<10% for coarse particles and <5% for fine particles) – see Fig. S8. While ECMWF wind fields are themselves subject to uncertainty, weather product skill continues to improve as a result of extensive use of global coverage satellite observations (Bauer et al., 2015).

Section 4.4: It is mentioned that the representation of mineral dust in models is limited. Can this be discussed specifically in the context of TM4 and ACCMIP? Are there direct model-obs comparison related to this from other studies? Is there some estimation of how poorly this might be represented in these models specifically?

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Response: Indeed, dust simulations are rather uncertain. We added text to illustrate the variations in simulated dust fluxes for TM4, ACCMIP and in mineral dust atmospheric transport models in general.

Revised text, Section 4.4: This is itself a considerable challenge. Dust emissions in TM4, simulated for the year 2008 using ECMWF meteorology, were 1181 Tg yr⁻¹ (Myriokefalitakis et al., 2016), while Kanakidou et al. (2016) simulated emissions almost 30% higher for the year 2005. In the case of ACCMIP, not all of the models involved included simulations of mineral dust aerosols (Lamarque et al., 2013b). In general, modelled dust deposition fluxes to remote ocean regions have been shown to vary by factors of 10 or more (Huneeus et al., 2011; Schulz et al., 2012) and to not reproduce key aspects of the dust cycle even in well-characterised regions (Prospero et al., 2010).

Section 4.5: There is great emphasis placed here and in the abstract and conclusions on the uncertainty associated with dry deposition velocities. How can new progress be made on this issue? Some type of recommendation, from the clear range of experts who appear here as co-authors, should be made. Also, it would be helpful to list the vd in Table 2 for both model products and that used to calculate the observed dry deposition here. From the text, it appears that modeled vd and assumed/calculated vd are not so vastly different, yet the discussion here (and in the abstract and conclusions) makes this appear as a vital issue. Does this suggest that we need to constrain velocities to within +/- 25% or more (or less)? More can be spoken to/digested here from this model-calculated data comparison.

Response: With regard to model product deposition velocities, TM4 calculates deposition velocities using the Ganzeveld et al. (1998) parameterisation (the same parameterisation used in the variable vd method for calculated deposition fluxes) every time that meteorology is changing (3-hours in TM4) and applies these to the concentrations of NH₄⁺ and NO₃⁻ every model time-step. Therefore, it is unrealistic to calculate a 'mean' deposition velocity. Instead we have calculated the 2-D field of the 'effective

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mean deposition velocity' as the ratio of the annual deposition fluxes to the respective annual mean concentration, both for NH₄⁺ and NO₃⁻. These fields (Fig. 2) have been added to the manuscript's supplementary material (as part of a revised Fig. S1) and additional text on this issue added to Section 4.2. Information on effective mean deposition velocity is not available for the ACCMIP product. Due to the limited number of cases for which effective mean vd is available, and because these values are not representative of the model function, we have added the available areal average vd values to a new table (Table S3) in the supplementary material, rather than in Table 2 as the reviewer requested. Overall though, agreement or disagreement between assumed / calculated deposition velocity fields and the effective mean deposition velocity fields of TM4 does not alter the fact that these deposition velocities are inherently uncertain. The reviewer's other comments here are addressed in our responses to comments on Section 5 of the manuscript.

Revised text, Section 4.5: Differences in the temporal scales of observations and model time-steps can also lead to biases. For instance, the variable vd method for CalDep and dry particulate deposition in TM4 are both based on the parameterisation of Ganzeveld et al. (1998). The CalDep calculation involves the use of mean observed aerosol concentration and ECMWF wind speeds averaged over the period 1995 – 2012. In TM4, wind fields (also based on ECMWF meteorology) are updated every 3 hours in order to calculate vd for each time step. In order to compare deposition velocities over similar time-scales, it is possible to calculate "effective mean deposition velocity" for NO₃⁻ and NH₄⁺ in TM4 (the ratio of the annual deposition fluxes to the respective annual mean concentrations), but these values are not representative of the deposition velocities used at the model time-step. Maps of variable vd used in CalDep calculations and effective deposition velocity for TM4 and areal average values of these for the study regions can be found in the Supplementary Information (Fig. S1 and Table S3).

Conclusions: Suggest renaming this to "Summary and Conclusions" Line 469: See also above – suggest making some type of recommendations on actions that can be

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taken here to improve the state of the field. More studies in different places? Specific types of studies – i.e. laboratory vs field measurements? More passive sampling?

Response: The section has been renamed as the reviewer suggests. We have added a new paragraph to Section 5 in which we discuss measures that might be used to improve models of atmospheric N input to the ocean. These include recommendations on observations, improvements to models and to model – observation comparisons.

Revised text, Section 5: There are a number of steps that can be taken to improve model predictions of atmospheric N inputs to the ocean. Observations of N deposition that target key areas of uncertainty (such as regions with strong seasonal cycles; intense gradients in N concentrations / deposition; and with contrasting mineral dust regimes) are required and these field campaigns should include measurements that address the needs of the modelling community. Examples of such measurements include: gas-phase N speciation and deposition flux, in addition to particulate N speciation (in order to better constrain modelled N simulations); more detailed measurement of N species aerosol particle size distributions and measurement of aerosol particle deposition fluxes over the ocean (to help improve estimates of particulate N dry deposition over the ocean); long-term measurement of dry particulate deposition N species fluxes, concurrently with N species wet deposition measurements, at suitable remote island locations. In the future, reducing uncertainties in v_d from small-scale wind and aerosol property heterogeneity may help provide more certain v_d estimates. One way to do so might be to estimate larger-scale v_d from remote sensing observations, based on relationships between N concentrations and surface and remotely-sensed aerosol properties. To date, these relationships are still poorly constrained. Improvements in emissions estimates, such as through the use of satellite-derived fire radiative power to assess biomass burning emissions (Freeborn et al., 2014), are key to improvements in the performance of models. Most model simulations of marine NH_3 emissions are based on the very old inventory of Bouwman et al. (1997). Both observations and models of air – sea NH_3 exchange have progressed since that study (e.g. Johnson

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et al., 2008; Paulot et al., 2015) and these advances should be incorporated into N atmospheric chemistry transport models more widely. Organic N species have been shown to comprise a significant fraction of atmospheric N (Jickells et al., 2013). Explicit inclusion of organic N into models (e.g. Kanakidou et al., 2012) should therefore result in more effective simulations of the atmospheric N cycle. Future model – observation comparisons would be more effective were the observations compared directly to the corresponding absolute time in the model, rather than over time-averaged periods as done here. Ideally, sampling of comparative values from the models should be done over time intervals matched to the collection period of the observations.

Table 2: Suggest reporting dry deposition velocities (v_d) for the models and the calculations for each ocean basin in this Table.

Response: See above.

Typos/minor edits: Line 121: “to” should be “for” Line 123: “associated” should be “association” Line 124: missing ‘with’ prior to “final aerosol fractions”

Response: These corrections have been made.

References Bauer, P., Thorpe, A., and Brunet, G.: The quiet revolution of numerical weather prediction, *Nature*, 525, 47-55, 10.1038/nature14956, 2015. Bouwman, A. F., Lee, D. S., Asman, W. A. H., Dentener, F. J., VanderHoek, K. W., and Olivier, J. G. J.: A global high-resolution emission inventory for ammonia, *Global Biogeochemical Cycles*, 11, 561-587, 1997. Freeborn, P. H., Wooster, M. J., Roy, D. P., and Cochrane, M. A.: Quantification of MODIS fire radiative power (FRP) measurement uncertainty for use in satellite-based active fire characterization and biomass burning estimation, *Geophysical Research Letters*, 41, 1988-1994, 10.1002/2013GL059086, 2014. Ganzeveld, L., Lelieveld, J., and Roelofs, G.-J.: A dry deposition parameterization of sulfur oxides in a chemistry and general circulation, *Journal of Geophysical Research-Atmospheres*, 103, 5679-5694, 1998. Huneus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R.,

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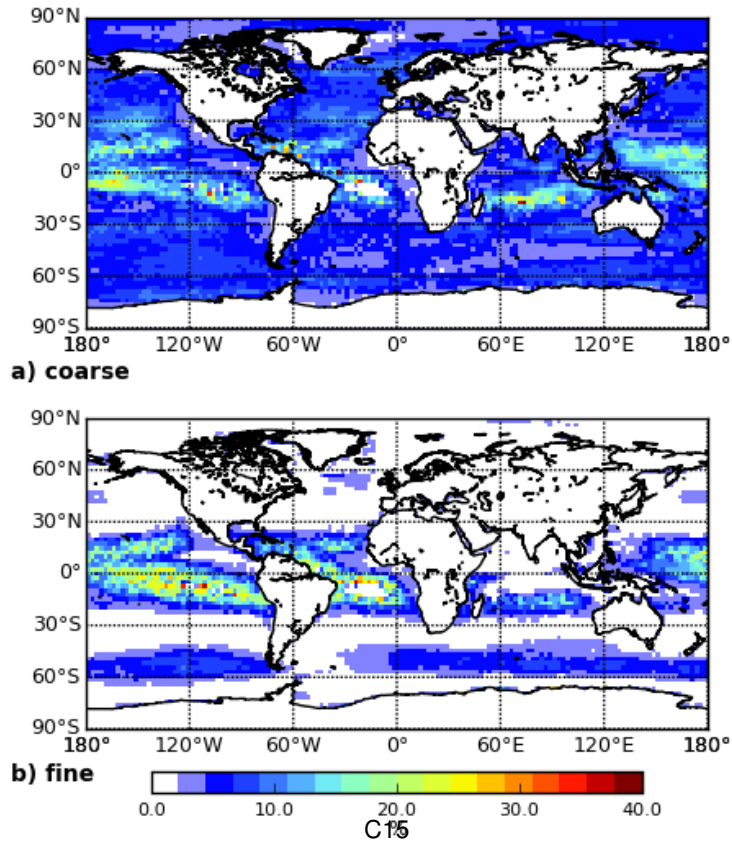
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Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-1123, 2017.

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Deposition velocity v_{sd}



TM4 Effective deposition velocity

