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

Anonymous Referee #2

Received and published: 26 March 2017

Based on an extensive ship-based observations of aerosol NO3 and NH4 concentrations in globe, this manuscript assesses the performance of simulated N concentration and deposition fluxes over three remote oceans. This is a very impressive manuscript that reports on the model-observation comparisons and is generally well written. This manuscript is thus a significant contribution to understanding state-of-the-art model limitations for annual average, seasonality, and spatial patterns. The primary shortcomings in the manuscript include: clarification of the methodology of models used, uncertainties due to emissions and meteorological forcing data, sensitivity of the model-observation comparisons on the size of spatial window, rationality of model-observation comparison analysis on deposition fluxes, implications for atmospheric community in

improving models. I presented the review of the paper into separate main topics corresponding to different section of the manuscript.

Abstract: The first paragraph should be shortened as much as possible to indicate the importance of model-observation comparisons as well as the influence of mineral dust on N depositions. The methodology in the second paragraph is much hard to follow: why choose TM4? How about the two commonly applied methods to calculate N deposition fluxes for CalDep? Does CalDep have the results of deposition fluxes of NOy and NHx? Are NOy and NHx derived from wet and dry depositions? Is it possible to compare nitrate and ammonium with ACCMIP means? In addition, is it possible to separate the contributions of deposition velocities and N concentration to model-observation discrepancy? I believe that it is much importance for scientific community to improve the model-related works in the future.

Introduction: Line 71-73: A few of global atmos. Models has been applied for large scale assessment of N deposition over oceans, such as Dentener et al. (2006), Wang et al. (2015), etc. such works should be cited: Dentener, F., Drevet, J., Lamarque, J. F., Bey, I., Eickhout, B., & Fiore, A. M., et al. (2006). Nitrogen and sulfur deposition on regional and global scales: a multimodel evaluation. Global Biogeochemical Cycles, 20(4), 16615-16615; Wang, R., Balkanski, Y., Bopp, L., Aumont, O., Boucher, O., & Ciais, P., et al. (2015). Influence of anthropogenic aerosol deposition on the relationship between oceanic productivity and warming. Geophysical Research Letters, 42(24), 10745-10754. Lin 88-89: again, please explain why choose TM4 in this study Lin 105-108: Sampling biases due to multiple sources should be discussed in details. Is any type of samples excluded based on sampling regulation related to N deposition in this study? It is better to describe the sampling regulation (sampler, period, temporal resolution, size fractions, sampling method, analytical method, etc) in main text or SI file or the dataset SOLAS

Methods: Line 127: Explain the abbreviation ECMWF first, and justify why choose ECMWF. Actually a few of reanalysis datasets including surface wind speed could be

used for estimating Vd. Could you please discuss more on the uncertainties due to the choice of meteorology dataset? Line 128: what is the difference between variable Vd model and well-tested deposition velocity model in previous works? How to calculate aerodynamic resistance and quasi laminar boundary layer resistance? A detail of variable Vd model should be provided in SI file. 2.3 model products: I strongly suggest introducing the methodology of TM4 model in SI file, forcing data, emissions of N, simulation setup, etc Line 139: how to compare modeled N deposition at coarse scale with site-scale observations? Line 152-153: CalDep is based on the observations for the period of 1995 to 2012, but ModelDep is simulated for a specific year (2005 for TM4, 2000 for ACCMIP MMM). Line 172: Spatial window of 5ĚŽ 5ĚŽ is used for model-observation comparisons. Could you please check whether the result is independent of the size of spatial windows?

Results and Discussion Line 203: explain the abbreviations TEAtl, NInd and NWPac. I suggest to use the full name of study regions in main text, but abbreviations in Tables or Figures. Line 333: I am not sure if it is necessary to have subtitles in the main text according to ACP style. Line 340-349: the total columns of NH3 retrieved from IASI satellite observations would be an effective way to validate the spatio-temporal patterns of ammonia, referring to: Van Damme, M.; Clarisse, L.; Heald, C. L.; Hurtmans, D.; Ngadi, Y.; Clerbaux, C.; Dolman, A. J.; Erisman, J. W.; Coheur, P. F. Global distributions, time series and error characterization of atmospheric ammonia (NH3) from IASI satellite observations. Atmos. Chem. Phys. 2014, 14 (6), 2905–2922. Line 350-353: Besides of the effect of pH, the inconsistency of sampling regulation for ship-based observation would be another source of biases. Please discuss in details on it. Subsection 4.2: Model-observation comparisons should focus on the difference in aerosol concentrations, but not on that in deposition fluxes. Actually CalDep deposition fluxes are also derived based on two simple methods, where the uncertainty of dry deposition velocity cannot be rationally quantified in this study. Subsection 4.3: I guess that the discrepancy in seasonality between modeled and observed N depositions would be due to the uncertainties of emission source and meteorological data. It would be of use

C3

to discuss in details on their influences. Subsection 4.4: please extend the discussion on the role of mineral dust on N deposition. It would be better to specify the limitation of models, such as TM4 and ACCMIP multi-model ensemble?

Conclusions: it is too long to follow as conclusion of the manuscript. I suggest to shorten it and focus on the main findings on the limitation of current models in estimating particulate N depositions and the recommendations on improving the models for atmospheric modeling community.