

Response to Reviewer #1 comments:

We thank Reviewer #1 for the valuable comments and suggestions. According to the comments, we have revised the manuscript. Below we provide a point-to-point response to address the reviewer's comments.

Review of

The NOAA Aerosol Reanalysis version 1.0 (NARA v1.0): Description of the Modeling System and its Evaluation

By Wei et al.

Overview:

The paper presents a global aerosol reanalysis data set for the year 2016 (NARA v1.0) produced by assimilating MODIS AOD in the GEFS aerosol model using the JEDI data assimilation framework. The reanalysis data and a control run (free) without AOD assimilation is evaluated with AOD and Angstrom exponent (AE) observations from Aeronet, surface PM observations derived from openAQ and speciated observations from the IMPROVE network. Further, it is intercompared against the MERRA-2 and CAMS aerosol reanalysis data sets. The authors find improved AOD compared to AERONET observations but no or only smaller improvements for AE and PM. Overall NARA v1.0 seems closer to the MERRA-2 re-analysis than to CAMS.

General remarks

The paper gives a reasonable description of the modelling system and the scientific accuracy of the NARA data set. However, there are further updates required before the paper can be recommended for publication.

- It is an omission of the paper that NARA, CAMS and MERRA-2 are only compared against each other for AOD and regional vertical profiles. Instead, the evaluation against independent observations should be carried for CAMS and MERRA-2 too. The accuracy measure of the three data should be compared in the paper. It is of great interest to the reader to get know which of these data sets is closer to observations.
- The evaluation results should be represented in a more quantitative manner and always for NARA and the FREE run to document the impact of the AOD assimilation. For example, the pdf plots do not have a colour scale legend and only some of them contain information about bias and correlation.

- The approach for the PM evaluation needs to be described in more detail, in particular the seemingly missing application of any quality control of the openAQ data should be justified. Further, the method to account for vertical stratification and spatial representativeness is not always clear.
- The abstract should be revised to contain more factual information from the paper. General remarks that are as such not direct conclusion from the paper should be avoided.
- The data assimilation procedure seems to include an optimisation of the emissions (see also Fig 1) Please provide more detail on this important aspect from a technical and a scientific perspective.

Response:

Thanks for the comments. We have revised the manuscript to address the comments. Please see the point-to-point response below.

Specific comments:

L 11: once published that is not a manuscript

Response: It has been revised to 'study' (L1).

L 11: "first ever" may be misleading because there are other aerosol re-analysis

Response: The "first ever" has been revised to "first prototype version of" (L1).

L 24: There is no clear evidence in the paper that single-wavelength assimilation is the main factor of the limited impact.

Response: The interpretation statement (next sentence) has been removed and the relevant discussion in section 5 has been revised accordingly (L209-213).

L 28: What is meant here by "climatologies"

Response: It has been modified to 'among datasets' (L27).

L 28: "In our opinion, such uncertainties may translate to inaccuracies in weather and climate modeling when impacts of aerosols on atmospheric radiation and/or cloud processes are considered." The paper does not deal in any way with the impact of aerosol and radiation and cloud processes and it is therefore not a conclusion of this paper. Please avoid statements that are not substantiated by the paper.

Response: This sentence has been removed in the revision and the relevant discussion in Section 6 is revised (L370-373).

L 71: Please check if SOA's and Nitrates are included in CAMSRA (I believe not)

Response: We confirmed that SOA is included in CAMSRA, which is detailed in section 2.1.1 of Inness et al. (2019). This is not considered in the previous version, CAMS interim reanalysis, which is the 'CIRA' in Inness et al. (2019) and 'CAMSiRA' in our study.

L 114: openAQ data are known for the lack of QC. Please expand on that and clarify the data sources that were compiled in the openAQ data set for 2016. A good solution would be to provide a global map to show the spatial distribution of the PM_{2.5} observations.

Response: To evaluate pNARA v1.0 against the OpenAQ dataset, we only utilized measurements from reference grade monitors (i.e. those maintained by government/research institutes) which are considered "the gold standard" (<https://openaq.org/developers/help/>). This detail has been added in the revision (L121-125).

L 123: Why are gases mentioned here? Did you use the data, for example SO₂?

Response: Additional information is helpful for future utilization of the dataset. The sentence has been revised accordingly (L124-125).

L 134: Comment on the importance of representing Nitrates and Secondary Organic Aerosol especially for the accuracy of PM.

Response: SOA and nitrates may constitute significant parts of fine PM depending on season and geographical location. Since the formation of SOA involves condensation of gases and complex chemical reactions including the organosulfates, organic nitrates, and interactions of ozone with free radicals (OH, NO₃) computational expense of including its parameterization in a global model like ours is beyond consideration. We expect that impact of neglecting SOA in PM would be largest over urban areas. Inorganic nitrates are indeed included in a recent extension of GOCART at NASA, GOCART2G, and will be accounted for in calculating PM in the future. We expect that the impact of neglecting nitrates in PM would be largest over agricultural areas. Rather than speculate we will not attempt to estimate by how much our results are influenced by lack of accounting for SOA and nitrates.

L 170-174: Please expand on these aspects (SPPT and emission updates) and provide more information about their usefulness for the realism of the NARA data set.

Response: The description has been expanded with how the scale factor and perturbation of emission can impact the analyses in pNARA v1.0 in the revision (L173-179).

L 174: Please show and discuss the modified emissions.

Response: The discussion of the impacts of scaling and perturbation to emissions on reanalyses is beyond the scope of the study. The influence of modified emissions induced by the scaling factor and perturbation are detailed in Huang et al. (2023).

L 205: Please provide more evidence for this statement “We hope”

Response: The statement has been revised and citations for the benefits/improvements induced by assimilating multiwavelength observations were added (L209-213).

L 232: Section 5.2 should also include a verification of CAMS-RA and MERRA-2 consistent with the verification of NARA and a comparison of the results

Response: We agree the intercomparison between reanalysis datasets may contain important information for users. However, it is beyond the scope of this study because we make no representation of other agencies. We believe that a fair intercomparison study would require a broad collaboration with others. Therefore, the citation of the evaluation papers of MERRA-2 and CAMSRA have been added for reader’s reference in Section 6 (L363).

L 304: Please, clarify how you account for the stratification. This would require making assumptions about the vertical aerosol profiles within the model grid box. Just using the diagnosed air density, will not achieve that.

Response:

We agree that deriving surface concentrations of species would require (numerous) assumptions. Below we elaborate why we chose a simpler approach.

As noted in the manuscript, the model mid-level where mixing ratios of species are calculated is at 20 m. This in most cases would fall within the atmospheric

surface layer. Vertical profiles of concentrations/mixing ratios of fine aerosols in the surface layer follow Monin-Obukhov theory and are expressed in the form

$c - c_0 = c^*/k [\ln(z/z_c) + \psi(z, z_c, L)]$, where

c is species concentration at layer depth z , c_0 is species concentration at the top of the interfacial layer, z_c is species dependent height scale, ψ is stability dependent function, L is Monin-Obukhov length, k is von Karman constant, and c^* is surface flux divided by friction velocity (e.g. The Atmospheric Boundary Layer, Garratt, J., 1992; Atmospheric Chemistry and Physics: From Air Pollution to Climate Change, Seinfeld, J.H, Pandis, S.N, 2016).

From this equation a formula for a concentration of species at measurement level can be derived by plugging the known concentration at model level and eliminating c^* . That leaves us with c at the measurement level as a function of stability dependent L and poorly characterized z_c and c_0 . We could simplify the equation by assuming neutral stratification (i.e. $\psi=0$) but z_c and crucially c_0 would need to be further parameterized. We don't believe that following this theory-based approach would provide a better approximation to the one we adopted. Our approach accounts solely for air compression i.e. by scaling model level concentration using densities at the lowest model level and at the measurement level assumed to be 2m (though it is also quite uncertain at what height the actual OpenAQ measurements are taken). We performed the evaluation of the modeled PM2.5 being fully aware of the shortcomings of such an effort.

L 307: Please provide information about the size distribution of the different aerosol component and motivate the PM formulae.

Response: The formula was provided by the lead author of the publication on the GEFS-Aerosol model (Geosci. Model Dev., 15, 5337–5369, <https://doi.org/10.5194/gmd-15-5337-2022>, 2022, Kate Zhang, personal communication, March 20, 2023). We believe that the formula is derived from the prescribed distributions of the mass of the aerosol component species given their mean radii and standard deviations.

L 358: Please add a comparison of the evaluation results from MERRA-2, CAMS and NARA here.

Response: Please see the response for L232.

L 362-365: "These observations ... " That is a very general statement and not really justified by the findings of the paper as you do not test the impact of NARA aerosol on

radiation and clouds. Please include multiple references if you want to make a point here. For example, you mention yourself that Bozzo et al. 2017 (there is also an ACP paper) successfully used CAMSRA to represent aerosol in the ECMWF model.

Response: Bozzo et al. (2017) and Mulcahy et al. (2014) have been cited in the revision to point out the importance of including the more representative aerosol states in the physical processes of aerosol-cloud-radiation interactions.

Figures:

Fig 1: Please here or in the text, provide more details in particular on the emission updates, the observation operators and the length of the forecast and assimilation window.

Response: The information regarding the system configuration has been revised in Section 4.

Fig 2: Please add colour legend and basic statistics (bias, R) . Please show AOD in linear scale and not in log scale. The pdf of AOD does not justify the use of the log scale.

Response: The color bars and statistics have been added to each sub-panel and thus Table 2 is removed in the revision. The logarithmic scale is used to increase the visibility of the agreement between model and baseline for small values of AOD, which was utilized in the evaluation of MERRA-2 (Buchard et al., 2017).

Fig 3: Please, clarify if you show a spatial or temporal Correlation coefficient R.

Response: This correlation coefficient is calculated based on the all paired data within each month. The figure caption has been revised accordingly.

Fig 12: Please add a colour legend, consider showing the plot for different regions and not just a global plot.

Response: The colorbar and statistics have been added. We also added the 2D map of temporal mean biases, RMSE, and correlation coefficient in Appendix A for reference. As shown in Figure A2 in the revision, the 'reference grade' stations in the OpenAQ dataset are mainly located over US and Europe. It does not show discernible improvements in pNARA v1.0. Compared to smaller biases over US and Europe, the substantial underestimates and large RMSE over India indicate the GOCART scheme cannot adequately capture the processes.