Quantifying Dust Deposition over the Atlantic Ocean

High IF journals deserve very high-quality papers and in this case dataset. This is definitively the case and I strongly support its publication and I suggest it as a highlight paper.

The authors would like to thank the reviewer for his time, comments and suggestions. We did our best to incorporate the proposed changes and corrections in the revised manuscript, aiming at improving the presented paper. Following, you will find our responses, one by one to the comments addressed.

Kind regards, Emmanouil Proestakis et al.

Reviewer's Comments

Section 2.2.1: the error on dust concentration is not discussed at all here. The methodology is widely described in previous paper, but here it should be reported together with dust concentration profile method also an estimation of the error on it. This is crucial for all the following analysis within the paper.

Following the reviewer's comment and recommendation, the following paragraph was inserted in the manuscript, in the "Section 2.2.1 - Decoupling the atmospheric pure-dust component from the total aerosol load":

"Uncertainties in the retrieval of atmospheric dust properties (i.e., backscatter coefficient, extinction coefficient, and mass concentration) originate from multiple sources, primarily (a) the uncertainties in the CALIPSO L2 optical products and aerosol classification flags, (b) the methodology applied towards atmospheric dust decoupling from the total aerosol load, and (c) the assumed constants and conversion factors (i.e., lidar ratios (LRs) and extinction-to-mass conversion parameters) (Marinou et al., 2017; Proestakis et al., 2018; 2024). CALIPSO L2 retrieval uncertainties, particularly in backscatter coefficient and particulate depolarization ratio at 532 nm, are assumed to be random and uncorrelated (Vaughan et al., 2009; Winker et al., 2009), yet can be significant; particulate depolarization ratio uncertainties often exceed 100%, and the limitations in aerosol subtype classification introduce further biases, especially for mixed or tenuous layers. The aerosol subtype classification algorithm (Omar et al., 2009; Kim et al., 2018) may result in positive or negative biases depending on feature type misclassification, with particularly low accuracy (~35%) in identifying polluted dust layers (Burton et al., 2013). Moreover, CALIOP's limited sensitivity to optically thin layers (Kacenelenbogen et al., 2011; Rogers et al., 2014) leads to systematic underestimation of AOD, with negative biases of ~ 0.02 (nighttime) and < 0.1 (daytime), primarily due to its detection sensitivity (Toth et al., 2018). The application of dust decoupling methodologies (Shimizu et al., 2004; Tesche et al., 2009; 2011; Ansmann et al., 2019) introduces additional uncertainties, with the depolarization-based separation approach contributing 5%-10% uncertainty during strong dust events and up to 20%–30% in less pronounced cases, primarily linked to variability in assumed dust depolarization ratios (Tesche et al., 2009, 2011; Ansmann et al., 2012; Mamouri et al., 2013). The conversion of decoupled backscatter profiles to extinction coefficients using regional LRs introduces relative uncertainties on the order of 15%-25%, while the final conversion to dust mass concentration profiles introduces further uncertainties of approximately 10%-15% (Tesche et al., 2009; Amiridis et al., 2013; Marinou et al., 2017; Proestakis et al., 2024). Cumulatively, uncertainties propagate and increase with each processing step, resulting in total uncertainties that can reach 10%-30% in backscatter coefficient, 15%-50% in extinction coefficient, and 20%-60% in mass concentration for ground-based lidar observations (Mamouri and Ansmann, 2017; Ansmann et al., 2019). However, in the case of CALIPSO-based retrievals, where the dominant uncertainty sources are the backscatter and depolarization ratio inputs, frequently of the same order of magnitude or even higher than the optical products, total uncertainties in mass concentration retrievals can escalate to 100%-150%, underscoring the challenge of achieving high precision in satellite-based dust mass estimates (Marinou et al., 2017; Proestakis et al., 2018; 2024).".

Section2.2.2: the authors report: "As a next step, a three-dimensional (3D) closed cuboid surface is assumed, of 5° length (zonal), 2° width (meridional), and 10km height (vertical), with the base surface at 0 km a.m.s.l." Could authors explain the choice of such dimension of the cuboid?

Towards establishing a 4-D reconstruction of the atmospheric dust aerosol component, in terms of mass concentration, and accordingly quantifying the deposited component over the surface of the broader Atlantic Ocean Atlantic, different spatial and temporal resolutions were tested.

The different spatiotemporal resolutions were applied in the framework of the developments made, with the objective to establish the 4-D atmospheric dust climate data record, as representative for the climatological characteristics of the dust transport events and pathways over the Ocean as possible, reducing at the same time CALIOP L2 5km noiseeffects propagating and contaminating eventually the output datasets and information. Aiming to provide an indicative example of CALIOP characteristics and CALIPSO limitations that had to be considered driving the final choice on the spatial and temporal resolutions is provided in the following figure. More specifically, the following figure provides the number of CALIPSO L2 5km quality assured profiles per $1^{\circ}x1^{\circ}$ deg² grid for 06/2020 (left) and the number of CALIPSO overpasses per $1^{\circ}x1^{\circ}$ deg² grid for 06/2020 (right), both for the region of interest of the study. It is evident that (i) the number of compasses varies highly from grid-to-grid, extending between no overpasses (0) and eight (8), a number of observational overpasses significantly lower than the equivalent from passive sensors, (ii) frequently "neighboring" $1x1 deg^2$ grids are observed differently, with temporal intervals at least of a few days, thus capturing in some cases different dust transport events, and (iii) even a high number of CALIPSO overpasses may not provide a significant number of CALIPSO L2 5km quality assured aerosol profiles, for instance due to cloud contamination.



Figure: (left) the number of CALIPSO L2 5km quality assured profiles per $1^{\circ}x1^{\circ}$ deg² grid for 06/2020, and (right) the number of CALIPSO overpasses per $1^{\circ}x1^{\circ}$ deg² grid for 06/2020, for the region of interest.

Taking into consideration the conclusions of the above-provided figure, it concurs that specific grid areas, characterized by a low number either of CALIPSO overpasses or of quality assured profiles, may not provide representative information of atmospheric dust transport, thus not accurate information of the deposited dust component. In the framework of the study, and following the need to balance an approach aiming to ensure as possible representativeness of the CALIPSO-based atmospheric dust products and at the same time maintain a number of grids high enough to allow for dust deposition retrievals, different spatial and temporal resolutions were tested. More specifically, the pre-processing resolution tests included the following:

- Spatial resolution: 1°x1° deg² grids / Temporal resolution: monthly mean / 10 km upper boundary (vertical).
- Spatial resolution: 2°x2° deg² grids / Temporal resolution: monthly mean / 10 km upper boundary (vertical).
- Spatial resolution: 5°x2° deg² grids / Temporal resolution: monthly mean / 10 km upper boundary (vertical).
- Spatial resolution: 1°x1° deg² grids / Temporal resolution: seasonal mean / 10 km upper boundary (vertical).
- Spatial resolution: 2°x2° deg² grids / Temporal resolution: seasonal mean / 10 km upper boundary (vertical).
- Spatial resolution: 5°x2° deg² grids / Temporal resolution: seasonal mean / 10 km upper boundary (vertical).

With respect to the "10 km upper boundary (vertical)" threshold, this was selected on the basis of CALIOP observations over the North Atlantic Ocean – Saharan Aerosol Layer (SAL). In should be noted that the approach assumes no leak at the top of the atmospheric columns, and no dust sources over the Atlantic Ocean. The net input and output fluxes over closed hypothetical cuboid surfaces should equal zero, assuming no sources/sinks inside the conceptual column. With respect to the vertical extend of the transported pure-dust over the Atlantic Ocean, we implement one year (2010) of CALIPSO overpasses to investigate the climatological maximum altitude in the SAL, for the December-January-February (DJF), March-April-May (MAM), June-July-August (JJA), and September-October-November (SON). According to the atmospheric dust climatology, the vertical extend is not expended -on a



climatological basis- beyond 7 km height. We have selected though the upper boundary of 10 km to account for rare events.

Figure: Upper figure: CALIPSO daytime (red) and nighttime (blue color) for the North Atlantic Ocean region of interest subdomain and for the year 2010, and atmospheric dust and non-dust mean backscatter coefficient profiles at 532 nm for DJF, MAM, JJA, and SON, for 2010.

With respect to the spatial and temporal resolutions, an indicative example of the impact of different selected resolutions is provided in the following figure. More specifically, the following figure provides the mean mass concentration profile of dust and the corresponding standard deviation of the mean mass concentration profile of dust in the atmosphere (Fig.b-e), for domain I, domain II, and domain II + domain III extension (Fig.a), for different temporal resolutions of monthly-mean (Fig.b-d) and seasonal mean (Fig.e). The extraction of the dust deposited

component is extremely sensitive to fluctuations of the mass concentration profiles, since they towards computation of the dust fluxes, are enhanced through the implementation of the wind speed. As can be seen, moving from finer $(1^{\circ}x1^{\circ} deg^2 grid)$ to coarser $(5^{\circ}x2^{\circ} deg^2 grid)$ spatial resolution and from finer (monthly-mean) to coarser (seasonal-mean) temporal resolution, the profiles become less noisy and more representative for the atmospheric dust transport, and as such suitable for extracting the deposition rate of dust over the Atlantic Ocean. The case of Spatial resolution: $5^{\circ}x2^{\circ} deg^2$ grid / Temporal resolution: seasonal mean / 10 km upper boundary (vertical), was selected.



Figure: Illustration of different spatial resolution domains considered towards the development of the dust deposition product: Domain I (1°x1° deg² grid), Domain II (2°x2° deg² grid), and Domain II + Domain III (5°x2° deg² grid) (a) and mean dust mass concentration profiles for Domain I and 1°x1° deg² grid and monthly-mean temporal resolution (b), Domain II and 2°x2° deg² grid and monthly-mean temporal resolution (c), Domain II + Domain III and 5°x2° deg² grid and monthly-mean temporal resolution (d), and Domain II + Domain III and 5°x2° deg² grid and seasonal-mean temporal resolution (e).

Section 2.2.2: nice this approach to consider that deposition is in-out dust, but what about dust trapped into clouds? I think this could be a reason of overestimation of deposition in the cells. Could the authors say something about this possibility? At least they should mention this aspect.

In the aggregation approach the official CALIPSO Level 2 (L2) to Level 3 (L3) method is followed. As described by Tackett et al. (2018), the L2 algorithms, among others, initially detect and classify atmospheric features (aerosol, cloud, surface) and accordingly retrieve extinction coefficients from the attenuated backscatter signals, derived from the L2 aerosol profile product. The CALIOP L3 aerosol profile product reports monthly statistics based on quality-screened level 2 aerosol extinction profiles at 532 nm, near-globally (85°S to 85°N) on a uniform 2° latitude by 5° longitude grid with a vertical resolution of 60 m. Files are generated for each month, for day and night files for each of the four following different sky conditions: all-sky, cloud-free, cloudy-sky transparent, and cloudy-sky opaque. It is emphasized that the "cloud-free" conditions are constructed from columns where no clouds are detected at 5 km or coarser horizontal resolution (L2). Moreover, it is emphasized that the applied quality assurance and cloud-clearing algorithm prior to averaging the attenuated backscatter and retrieving extinction, results in "cloud-free" conditions to represent the highest quality level 3 data as extinction retrievals are minimally affected by errors in retrieving the attenuation of overlying cloud cover.

Following the CALIPSO L3 official recommendation, the present study applies also the "cloud-free" conditions, as it is considered of the highest quality possible. With respect to the presence of dust in the aerosol profiles of L2 5km characterized by presence of clouds and thus removed from the analysis we cannot say whether or not they would result in overestimation of the mean profile of underestimation of the mean profile, since frequently they are accompanied by no measurements (i.e. total attenuation of CALIOP lidar beam) and thus no provision of information on the presence of aerosols. The L3 algorithm iterates through all L2 files within each season. As such, here dust backscatter, extinction, and mass concentration samples are quality-screened and accordingly aggregated into the appropriate latitude-longitude grid cells, and accordingly averaged for each grid cell. Upon L2-to-L3 aggregation though, the dust aerosol profile products (i.e., mean backscatter coefficient, extinction coefficient, and mass concentration) are produced. It should be noted that the approach applied requires sufficiently spatial and temporal averaging to obtain meaningful dust spatial distributions, both in terms of atmospheric dust and dust deposition. Moreover, the dust deposition corresponds to the net loss of dust by all removal processes; it is not feasible to separate dry from wet deposition. Overall, upon the aforementioned described "cloud-free" L2-to-L3 averaging procedure the $5^{\circ}x2^{\circ}$ deg² grid provides representative reconstruction of the atmospheric dust transport, with the highest quality level 3 data as extinction retrievals are minimally affected by errors in retrieving the attenuation of overlying cloud cover (Tackett et al., 2018).

In order to demonstrate this even better, we provide a methodological illustration of the impact of averaging, in $1^{\circ}x1^{\circ}$ deg² grids, for an indicative CALIPSO nighttime orbit on the 13^{th} of May 2020 - 00:49:42UTC (a). The figure includes the CALIPSO feature type (b) and aerosol subtype (c) classifications to provide an overview of the presence of clouds-aerosols and on the classified as aerosol layers the cases of dusty layers. Figure d provides the quality-screened dust extinction coefficient at 532 nm profiles along the CALIPSO overpass, the "cloud-free" condition as described above. Finally, figure (e) shows the impact of the $1^{\circ}x1^{\circ}$ deg² grids averaging of the dust extinction coefficient at 532 nm profiles to a degree the effect of cloud contaminated removed aerosol profiles. The impact is quite more pronounced in case of larger spatial averaging as in the present study, significantly reducing the impact of cloud-presence.



Figure: Figure: Indicative CALIPSO nighttime orbit on the 13th of May 2020 - 00:49:42UTC (a), CALIPSO feature type classification (b), CALIPSO aerosol subtype classification (c), quality-screened dust extinction coefficient at 532 nm profiles along the CALIPSO overpass (d), and $1^{\circ}x1^{\circ} deg^{2}$ grids averaging of the dust extinction coefficient at 532 nm profiles (e).

Figure 6: maybe a better scale could be adopted for DOD, the pail blue is almost not visible and a reader could wonder about the highly visible deposition for a not-visible DOD.

According to the reviewer's comment, the DOD at 532 nm figures have been recreated and adapted, as follows:





Figure: LIVAS Dust Optical Depth at 532 nm (DOD), provided in annual-mean, DJF, MAM, JJA, and SON, estimated for the period 12/2006-11/2022. Left column: Initial version of figures. Right column: updated version of figures.

Figure 14: in the legend there are lines overwritten related to not plotted datasets. Please remove them and check all the other figures.



The figures were substituted with better-looking ones, as shown below:

Figure 14: Monthly series of EO-based DOD and dust total deposition (Obs, black line), EC-Earth3, MONARCH, and EMEP (models' results are collocated with LIVAS data at grid cells of 2°x2°).

It should be explained why the AeroVal comparison is done only for 2020.

We acknowledge that multi-year comparison of LIVAS dust with model simulations would provide more robust results. However, the manuscript to a large degree relies on the results obtained in the framework of ESA DOMOS project, where the year 2020 was selected for AeroVal comparison, for which LIVAS and modelled data were available at that time (as both the development of the AeroVal tool to implement DOMOS products/model results and model simulations are rather time demanding, in particular comparison modelled dust AOD and deposition with gridded

LIVAS data was implemented in AeroVal and applied for the first time). Despite an undoubtful benefit of multi-year evaluation, we believe that our consistency check between LIVAS and modelled dust for 2020 provides a good indication of fair data correspondence.

The following explanation has been included in the manuscript:

"Due to data availability at the time of preparing the manuscript, the comparison was made for only one year of 2020, however we believe that the presented consistency check between EO-based and model simulated dust AOD and deposition rates for 2020 provides a good indication of reasonable data correspondence."