Reply to Review of **Large synthesis of in 1 situ field measurements of the size distribution of mineral dust aerosols across their lifecycle**

by Paola Formenti and Claudia Di Biagio

This article compiles and unifies in situ dust size measurements from a large number of previous studies to form normalized distributions of particle volume versus diameter at three stages in the lifetime of a dust plume: near the source, a few days after uplift, and far downwind. The authors form averages across the studies for each stage, but to this reviewer (a modeler), what seems most valuable is the harmonization of the individual data sets to a physically consistent size variable: the geometric diameter. The authors additionally provide harmonized versions of the data that treat the particles either as spheres or make further assumptions to account for aspherical particles. What is particularly welcome, along with the accompanying files containing the harmonized data from each individual study, is the description of each study in the Supplement that lists the instruments and size descriptor that was measured, and most importantly, locates the measurements in space and time. Modelers can use these harmonized measurements from individual studies and information from the Supplement to compare their models consistently.

I think this paper is thorough, well-written and significant. My comments are only to help improve clarity.

We would like to thank Referee # 3 for his thoughtful review of our manuscript. Our answers are in indicated in blue.

line 10: are -> that are

Done

line 11: proper -> properly

Done

line 18: maybe use 'grid' for 'path'?

Done

line 25: "decreasing to ~5 μm and ~2 μm for MRT and LRT conditions" This seems excessively precise, even with the approximation symbols, given the uncertainty and fluctuations of each of the three distribution curves in Figure 3. Instead, I suggest describing only the shift of the mode from order 10 um to order 1 um as the dust moves downwind.

Following the reviewer's suggestion the abstract text has been revised as (lines 22-25): "The harmonized dataset shows consistent features suggesting the conservation of airborne particles with time and a decrease of the main coarse mode diameter from a value of the order of 10 μm (in volume) for SOURCE dust to a value of the order of 1-2 μm for LRT conditions. An additional mode becomes evident below 0.4 μm for MRT and LRT dust."

line 40: "started in the last decade" Dust models go back at least three decades to Tegen and Fung (1994) if not earlier (S. Joussaume, 1990; C. Genthon 1992).

Thanks for the comment. We have replaced with “started in the last decades”.

line 55: being -> been

Done

line 62: tenths -> tens?

The reviewer is correct and we have replaced “tenths” with “tens”.

line 67: "spurious": This variability is not necessarily spurious. It may be instead just a reflection of local conditions that may not apply elsewhere.

The word “spurious” has been replaced with “local”.

line 79: by "ensembles" do you mean collections or compilations of measurements?

The word “ensembles” has been replaced with “compilation of measurements”

line 88: why exclude deposition samples? Is this because of the additional assumptions that must be made to convert a flux into the concentration measurements compiled here?

Deposition samples were excluded because the main aim was to synthetize observations for airborne dust, therefore we considered more consistent to use measurements from both ground based and aircraft observations only.

line 106: How confident is this assignment of dust age? If there is a continuum of transport times, then some cases near the temporal boundaries of each category might be mischaracterized, a source of uncertainty. This uncertainty is fine if acknowledged, and I like the supplement because age is more clearly acknowledged as an estimate.

We thank the reviewer for this comment. We have added the following sentence in the main text (lines 109-113) to highlight this aspect “To note that potential uncertainties may arise in this classification, in particular for datasets lying at the boundaries of the SOURCE, MRT and LRT categories, and we acknowledge this aspect as a source of error in our analysis. We invite the reader to refer to the Supplementary material (Text S4) for thorough description of the assumptions made in some cases to associate each dataset to a category.”.

line 168: I understand the importance of harmonizing the different types of diameter present in the literature (e.g. geometric vs. aerodynamic). What is unclear is what is meant by "differences in number concentrations" in this context.

The reviewer is right. In fact, as while the diameter requires harmonization due to different definitions, for what concerns the number concentration is not really a harmonization, but just a normalization to remove differences in absolute particle concentrations arising from different samples atmospheric conditions. In order to clarify this point the sentence has been rewritten as (lines 171-173): “The original observations were treated to provide with a harmonized dataset in terms of the definition of particle diameter and data were normalized to remove differences in sampled number concentrations.”

line 197: CRI = 1.53 + 0.003i. What is the citation for this? The CRI depends upon wavelength, and should match the wavelength used by the instrument. Is this range of instrumental wavelength small enough that a single CRI is representative?

The range of wavelength of OPC instruments considered in this study covers the intervals 440 to 1054 nm, as detailed in Text S2. The value of 1.53-0.003i represents the average value reported by Di Biagio et al. (2019) for dust from global regions in the 370-950 nm wavelength range. In that study it is shown that the real part of the refractive index is almost wavelength-independent and does not change for different source regions globally, with an average value of 1.52 ± 0.04. The imaginary part is instead regionally- and wavelength-dependent and decreases with increasing wavelength, varying between minimum values of 0.0004 and 0.006 in the 370-950 nm range for different source areas. The CRI in this study is therefore set to cover the broad regional- and wavelength-dependency of the imaginary refractive index, representing a moderately absorbing mineral dust. This text has been added in the main text to clarify this point (lines 200-202): "This CRI value is at the average of the dust refractive indices reported in the 370-950 nm spectral range in Di Biagio et al. 2019) for dust of global origin."

The following text has been adapted in Text S3 to cover the same consideration: "The corrections factor by Formenti et al. (2021) for spherical dust and assuming a refractive index of 1.53−0.003i in the visible (value that is at the average of the dust global values reported in Di Biagio et al. 2019 in the range 370-950 nm, therefore covering the range of operation OPC wavelengths as details in Text S2) are applied to correct the different datasets."

line 224: Do these correction factors depend upon the observed distribution of shape parameters compiled by Huang et al. 2020: e.g. the aspect ratio and height-to-width ratio? If so, a brief description of these assumed shapes descriptors would be useful.

A detailed description of the retrieval of these parameters in the original publications is already provided in the Text S3, also linking to the information on the size- and global-distribution of the aspect ratio and height-to-width ratio. For example, as extracted from Text S3: "Huang et al. (2020) compiled global AR and HWR and found that both parameters deviate from unity and seem to be size independent and being lognormally distributed. They determine a median globally averaged value of 1.7 ± 0.03 for AR and 0.40 ± 0.07 for HWR. For aspherical dust, based on the application of Eq. (S1) and applying a Monte Carlo simulation taking into account the global distribution of AR and HWR, Huang et al. (2021) derived a global average conversion factor of 1.56 to convert D_{area} into a D_{geom} ($D_{geom}= D_{area}/1.56$)."

line 244: "Additionally, the 25% and 75% percentiles are also calculated, despite keeping in mind their limited representativeness given the reduced number of samples in the datasets, especially for SOURCE and LRT classes." You should also note that each of the individual measurement studies have their own uncertainty that will vary with the instrument and duration of the measurement period that is not accounted for here. (I realize that this information is not always available or possible to estimate robustly.)

We thank the reviewer for this comment. We have added a small text to specify this aspect in Sect. 2.4 "Limitations of the chosen approach": The text reads as follows (lines 293-295): "An additional source of error is the individual measurement uncertainty, which varies with the specific setup, instrument and spatial and temporal extent of the measurement."

line 270: delete 'used"

By reading the sentence we feel that keeping "used" clarify the meaning of the sentence. So, if the reviewer is fine with this, we would keep the word.

286: take -> taken

Done

line 287: "and when the first and/or the last bin of the corrected size showed unrealistic divergence" I do not understand what is meant by unrealistic divergence.

Sometimes when applying corrections to size distribution data it may happen that the number concentration dN/dlogD in a single bin becomes unrealistically-high or unrealistically-low because the width of the bin is changing. As measurements at the boundaries are sometimes poorly constrained because of the difficulty in defining the exact lower and upper limits of the first/last channels, some divergence can arise. However, these values are often not realistic (i.e. an order of magnitude or more different from the original values) and only the results of a numerical calculation. As maybe the word "unrealistic divergence" is not easy-understandable, we modified in "significant divergence". We hope this is clearer now.

line 316: "A main mode located at ~10 µm" This is outside the range of normalization cited on lines 180-181. Maybe note that this mode near 10 um (and possibly beyond) is based upon only a few studies?

We thank the reviewer for suggesting t put this aspect in evidence. We modified the text as (line 325): "A main mode located at ~10 µm (in volume) is observed for dust at emission and close to sources, as based from the few studies allowing to measure up to the coarse fraction."

line 328: the normalized size distribution?

The text has been changed accordingly.

line 355: "Dust particles below 0.4 µm in diameter are seldom measured close to source regions, but are found in observations at mid– and long–range transport conditions." Could you speculate about why this is? Is this emergence of a fine fraction the result of contamination by non-dust aerosols that is more apparent far from the source where dust concentrations are smaller? Alternatively, is it a consequence of the normalization, where larger particles are emphasized closer to the source and smaller particles are obscured?

Some elements of discussion on this aspects are already provided in the main text (lines 327-336): "As a matter of fact, the sparse datasets measuring very fine particles at the SOURCE show that particles with diameters below 0.4 µm (however measured only down to 0.2 µm, as shown in Fig. 2) represent approximately 20% of the total particles' number, increasing to more than 90% in MRT and LRT. Instruments such as SMPS and DMPS used in MRT and LRT studies measure particles as small as 0.02 µm in diameter. Previous single–particle compositional observations showing that the particle number concentration in the size range between 0.1 and 0.4 µm is largely contributed by aluminosilicate dust particles at emission, while internal or external mixing with aerosols other than dust gains importance with time and altitude of transport (Chou et al., 2008; Kandler et al., 2007, 2009; Weinzierl et al., 2009; 2017; Klaver et al., 2011; Denjean et al., 2016a; 2016b)."

The specific range of normalization is not expected to modify the shape and relative modes of the size distribution, as it implies only a size-invariant multiplicative factor, therefore we do not expect this to induce emphases on the fine or coarse modes.

Finally, as for Refere #1 we have also checked with the EasyData team who confirmed that the portal uses a https protocol. The issue is now solved after the intervention of the respective IT teams (EasyData and NASA). We thank R. Miller for his assistance on the matter.

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmos. Chem. Phys., 19, 15503–15531, https://doi.org/10.5194/acp-19-15503-2019, 2019.