| 1<br>2 | Large synthesis of in situ field measurements of the size distribution of mineral dust aerosols across their lifecycle                         |
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#### 9 Abstract

10 Mineral dust aerosol is important in the Earth system and the correct representation of its size distribution is fundamental for shaping the current state and the evolution of climate. Despite many 11 12 observational dust size data that are available in the literature, using this body of information to properly guide the development and validation of climate models and remote sensing retrievals remains 13 14 challenging. In this study we collect, evaluate, harmonize, and synthetize 58 size distribution data from 15 the past 50 years of in situ field observations with the aim of providing a consistent dataset to the 16 community to use for constraining the representation of dust size across its lifecycle. Four levels (LEV) 17 of data treatment are defined, going from original data (LEVO), data interpolated and normalized on a 18 standardized diameter grid (LEV1), and data in which original particle diameters are converted into a 19 common geometrical definition under both spherical (LEV2a) and aspherical (LEV2b) assumptions. Size 20 distributions are classified as emission/source (SOURCE, <1 day from emission; number of datasets in 21 this category, N=12), mid-range transport (MRT, 1-4 days of transport; N=36) and long-range transport 22 (LRT, >4 days of transport; N=10). The harmonized dataset shows consistent features suggesting the 23 conservation of airborne particles with time and a decrease of the main coarse mode diameter from a 24 value of the order of 10  $\mu$ m (in volume) for SOURCE dust to a value of the order of 1-2  $\mu$ m for LRT 25 conditions. An additional mode becomes evident below 0.4  $\mu$ m for MRT and LRT dust. Data for the three levels (LEV1, LEV2a, LEV2b) and the three categories (SOURCE, MRT, LRT), together with statistical 26 27 metrics (mean, median, 25% and 75% percentiles, and standard deviation) are made available as: 28 SOURCE (https://doi.org/10.57932/58dbe908-9394-4504-9099-74a3e77140e9; Formenti and Di Biagio, 2023a);

- 29 MRT (<u>https://doi.org/10.57932/31f2adf7-74fb-48e8-a3ef-059f663c47f1;</u> Formenti and Di Biagio, 2023b);
- 30 LRT (<u>https://doi.org/10.57932/17dc781c-3e9d-4908-85b5-5c99e68e8f79</u>; Formenti and Di Biagio, 2023c).
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## 32 Introduction

Airborne mineral dust aerosols emitted by the aeolian erosion of bare soils contribute in a major way to the Earth's radiative budget and environmental processes, including the human health. Because of their native mineralogical composition and size distribution, they interact with solar and infrared radiation, influence the formation and brightness of liquid and ice clouds, and affect the composition of the atmosphere and the ocean, while also transporting pollutants, viruses and bacteria across the continents and the oceans (Knippertz and Stuut, 2014, and the many references therein).

As a consequence, a large effort has started in the last decades to include the representation of those properties in climate and air quality models. Indeed, the complex mineralogy of mineral dust, depending on that of the parent soils (Claquin et al., 1999; Journet et al., 2014; Gonçalves Ageitos et al., 2023a), is now accounted for in models (Scanza et al., 2015; Perlwitz et al., 2015a; 2015b; Menut et al., 2020; Kok et al., 2017; Di Biagio et al., 2020; Gómez Maqueo Anaya et al., 2024) and starts to be retrieved by remote sensing (Green et al., 2020; Zhou et al., 2020; Di Biagio et al., 2023).

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- On the other hand, representing the span and the variability in time and space of the dust aerosol sizedistribution remains a challenge.
- The particle size distribution of mineral dust extends over several orders of magnitudes. Iron-rich particles as small as 14 nm in diameter have been observed in the laboratory from deflating soils by
- 49 Baddock et al. (2013). During sandstorm in Algeria, Gomes et al. (1990) measured an increase of the
- 50 mass concentration of particles between 100 nm and 1  $\mu$ m, and attributed this to clays disaggregated
- 51 by sandblasting. Measurements of the size-resolved vertical dust flux by Gillette et al. (1972; 1974a;
- 52 1974b) based on microscopy analyses of samples from Texas and Nebraska showed the presence of
- 53 particles up to several microns in dust emissions.
- The representation of the accumulation and coarse modes in mineral dust has long been based on the columnar measurements by the sun/sky photometers of the Aerosol Robotic Network (AERONET) network, which provides with normalized size distributions of mineral dust considered as chemically homogeneous particles the 0.1—30 µm optically–equivalent diameter (Dubovik et al., 2002; 2006; Holben et al., 2011), and which, incidentally, serve also the look–up tables of the remote sensing retrievals of dust from space (e.g., Cuesta et al., 2015; Zhou et al., 2020).
- Nevertheless, in situ observations at ground–based stations and on aircraft in more recent years have shown that particles of several tens, sometimes hundreds, of micron are airborne at emission, and
- remain so after several days of transport (Reid et al., 2003; Formenti et al., 2003; Rajot et al., 2008;
- 63 Chou et al., 2008; Kandler et al., 2007; 2009; Wagner et al., 2009; Klaver et al., 2011; Ryder et al., 2013;
- 64 2015; Rosenberg et al., 2014; Denjean et al., 2016; Wienzerl et al., 2017; van der Does et al., 2018).
- 65 These observations have been instrumental to a number of advances. Using them as ensemble dataset,
- to smooth local atmospheric variability, they have served as a basis to a new classification of the dust
- size distribution in four modes, namely fine dust (diameter  $\leq 2.5 \ \mu$ m), coarse dust (2.5 < diameter  $\leq 10$
- $\mu$ m), super coarse dust (10 < diameter ≤ 62.5 μm) and giant dust (diameter > 62.5 μm), extending above
- 69 the size range retrieved by AERONET (Adeyemi et al., 2023). Additionally, they have also fostered the
- 70 revision of the numerical schemes of emissions and deposition, and identified the numerous processes
- 71 and properties (non–spherical shape of particles, electric forces, atmospheric turbulence), that could
- 72 counteract the size-selective removal by gravitational settling and keep particles airborne longer than
- expected (Kok, 2011; Huneeus et al., 2011; Mahowald et al., 2011; Kok et al., 2017; Di Biagio et al, 2020;
  Zhao et al., 2022; Adebiyi and Kok, 2020; Adebiyi et al., 2020; Huang et al., 2021; Meng et al., 2022;
- 75 Adeyemi et al., 2023).
- 76 In support of those activities, in this paper we present a large and standardized compilation of *in situ*
- observations of the particle size distribution of mineral dust conducted during the past 50 years of
- 78 research. This dataset extends the currently published compilations of measurements (Meng et al.,
- 2022; Adeyemi et al., 2020; 2023) to provide with a state-of-the art of the current knowledge in support
   to the development of models, and ground-based and satellite remote sensing. Analysis of this dataset
- may provide with an integrated view of the size distribution of dust particles across their life cycle to
- 82 evaluate their impacts in the Earth/human system.

# 83 **2. Methods**

# 84 **2.1** Constitution of the dataset

Data presented in this paper result from in situ ground-based and aircraft observations of airborne dust conducted during field campaigns during the past 50 years of dust research. Data from deposition samples (e.g., van der Does et al. 2018 or Varga 2021) are not considered in this analysis.

Only datasets being published and properly referenced in the open peer-reviewed literature were retained. We also privileged datasets for which the methodology of acquisition, calibration and data 90 treatment was well described so that the data quality can be assessed. Finally, we search for data as 91 much as possible representative of different source and transport regions of the world.

The observations contributing to the dataset are listed in **Table S1** and the spelling of the acronyms of the field campaigns is reported in **Text S1** in the supporting material. Data are geo-localized in **Figure 1**, where they are classified with respect to their time after emission. Geographical coordinates are reported in Table S2.





Figure 1. Geographical location of the datasets contributing to size distribution observations for the source, the
 mid-range transport (MRT) and the long-range transport (LRT) categories. The legend indicates the line style
 used in the plot. The number of data for each category is indicated in the parenthesis in the legend.

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105 Observations obtained at the time of dust emission or within 1 day after emission are classified as 106 SOURCE. Observations corresponding to 1 to 4 days after emission and/or geographically acquired near-source regions (for example, offshore North Africa) are classified as mid-range transport (MRT). 107 108 Observations at times exceeding 4 days after emission or geographically distant from source regions 109 (for example, observations in the Caribbean) are classified as long-range transport (LRT). To note that 110 potential uncertainties may arise in this classification, in particular for datasets lying at the boundaries 111 of the SOURCE, MRT and LRT categories, and we acknowledge this aspect as a source of error in our 112 analysis. We invite the reader to refer to the Supplementary material (Text S4) for thorough description 113 of the assumptions made in some cases to associate each dataset to a category.

114 The SOURCE dataset (Fig 1, black points) consists in 12 observations in Northern Africa, North America, 115 and Asia, and one data set in Australia. They include works by Gillette et al. (1972, 1974), Gillette (1974), 116 Fratini et al. (2007), Rajot et al. (2008), Sow et al. (2009), Shao et al. (2011), Ryder et al. (2013a, 2013b), 117 Rosenberg et al. (2014), Huang et al. (2019), and Khalfallah et al. (2020), a set of data recently used by 118 Kok et al. (2017), Di Biagio et al. (2020) and Huang et al. (2021) to constrain the shape of dust size 119 distribution at emission in model studies, and the most recent work by Gonzales-Florez et al. (2023). 120 The MRT class (Fig. 1, blue points) is contributed by 36 datasets from field campaigns (ACE2, ACE-Asia, ADRIMED, AER-D, AMMA, DABEX, DARPO, DIAPASON, DODO1-2, FENNEC, GAMARF, GERBILS, INDOEX, 121 NAMMA, RHaMBLe, SALTRACE, SAMUM1–2, TRACE–P, and UAE2) in Western Africa, Capo Verde, the 122 123 Mediterranean basin, the eastern tropical Atlantic, Saudi Arabia, Japan, and Indian Ocean, downwind 124 sources either over the ocean or over desert areas. Additional datasets from studies performed in the Sahara, the Atlantic Ocean, Canary Islands and Japan (Schütz, 1981; D'Almeida et al., 1987; Maring et al., 2000; Kobayashi et al., 2007) are added to the dataset. The LRT class (Fig. 1, red points) lays on 10 datasets of observations across the Atlantic Ocean and South America and is contributed by observations from Bacex, CLAIRE, Dust-Attack, Go-Amazon, PRIDE, and SALTRACE campaigns and intercontinental dust transport data from Schütz (1981).

### 130 **2.2.** Instrumentation contributing to the in situ dataset

The natural dynamical range of the particle size and concentration of mineral dust can only be represented by a combination of instruments based on different intrinsic particle properties such as density, electrical charge, shape and composition (e.g., Reid et al., 2003a; Formenti et al., 2011; Wendisch and Brenguier, 2013; Mahowald et al., 2014, Adeyemi et al., 2023). As a consequence, the datasets considered in this paper are contributed by different in situ instruments, also described in **Text S2** in the supporting material, namely:

- Optical particle counters (OPC) using the dependence of light scattering on particle size and providing
  with the particle concentration as a function of the optical equivalent diameter (e.g., Reid et al.,
  2003b; Clarke et al., 2004; Osborne et al., 2008; Formenti et al., 2011; Ryder et al., 2013a, 2018;
  Khalfallah et al., 2020).
- Particle collection by filtration or impaction followed by individual particle characterization by transmission (TEM) and/or scanning electron microscopy (SEM) sizing particles as function of their equivalent projected-area diameter and coulter geometric sizing methods, (e.g., Gillette et al., 1972, 1974a, 1974b; Reid et al., 2003a; Khobayashi et al., 2007; Kandler et al., 2009; Chou et al., 2008).
- Multi-stage filtration or impaction sampling coupled with gravimetric or chemical analysis providing
   with the mass size distribution as equivalent aerodynamic diameter (e.g., Formenti et al., 2001; Reid
   et al., 2003b).
- Differential and Scanning Mobility Particle Sizer (DMPS and SMPS) providing the size of particles in the submicron range as the electrical mobility equivalent diameter of a charged particle moving in a static electric field (e.g., Maring et al., 2000, 2003; Bates et al., 2002; Müller et al., 2010; Denjean et al., 2016a, 2016b).
- Aerodynamic particle sizers (APS), measuring the equivalent aerodynamic diameter of a sphere of
   unit density having the same terminal velocity in an accelerated airflow as the irregularly shaped
   dust particles (e.g., Maring et al., 2003; Reid et al., 2003b; 2008; Struckmeier et al., 2016)
- 155 Each of those instrument types size particles on an equivalent diameter (optical, projected-area, 156 aerodynamic, mobility) that depends on their respective working principle. Converting those 157 operational size definitions into a homogenized one is part of the treatment applied in this work, which 158 follows the theory proposed and discussed in the literature and benefits of recent progresses in characterizing/synthetizing dust properties relevant for these treatments (e.g., Hinds, 1999, De Carlo et 159 160 al., 2004 ; Mahowald et al., 2014; Di Biagio et al., 2019; Huang et al., 2020, 2021; Formenti et al., 2021). 161 Diameter definitions and formulas to convert each of them into a geometrical diameter, both under the 162 assumption of spherical and aspherical dust, is provided in **Text S3** and summarized in **Table S3**.
- **Text S4** presents relevant information on each dataset considered in the present analysis. This includes a brief description of the field operations, the experimental conditions, the type of original data (number, volume or mass concentration size distribution, size-resolved emission fluxes), the instrumentation, and the data treatment applied to the measurements (averages, diameter corrections, etc.) in the original publication. Original data were obtained, as much as possible, through a personal contact with the data providers or from the original publications. This is also indicated in **Text S4**.
- 169 **2.3. Data treatment, harmonization, and synthesis**

170 The original observations were treated to provide with a harmonized dataset in terms of the definition 171 of particle diameter and data were normalized to remove differences in sampled number 172 concentrations. Four level of data treatment are defined as described below.

173 1/ Level-O (LEVO): original data, taken at the native resolution or the resolution from digitalization 174 process and converted into volume distribution assuming spherical particles ( $\pi/6*D^{3*}dN/dlogD$ ), where 175 D is the particle diameter used in the publication and dN/dlogD is the particle number concentration. 176 To remove differences in concentration, and in absence of information on original bin width, *LEVO* data 177 are normalized to the maximum of the volume size distribution;

178 2/ Level-1 (LEV1): data from LEVO are interpolated over a common size range of equi-logarithmically 179 spaced diameters (dlogD = 0.05) encompassing the original diameter range for each dataset and 180 normalized so that the integral is equal to 1 over a common diameter range. The diameter range for 181 integral normalization was set to be the largest as possible and to be covered by more than 90% of the 182 datasets in each category. For SOURCE data it resulted that the diameter range for common integral 183 normalization is within 1.58 and 7.1 μm, and for MRT and LRT it is between 0.71 and 8.9 μm.

3/ Level-2a (LEV2a): based on LEV1, the LEV2a data treatment aims at harmonizing the size distributions
 by converting the operational original particle diameters, which depend on the physical principle of each
 instrument, into a common-defined sphere-equivalent geometric diameter. Data from LEV1 are
 treated as in the following with respect to their diameter corrections:

- o data already provided as geometrical diameters (from coulter counters, i.e., only one dataset in our study) are left unchanged;
- 190 o data provided as projected-area diameters (i.e. from microscopy) are left unchanged;
- 191 o data provided as aerodynamic diameters (from APS or cascade impactors) are corrected 192 assuming a shape factor ( $\chi$ ) of 1 (under spherical assumption), therefore a size-invariant 193 conversion factor of 1.58 (see Eq. S2) is applied to the dataset assuming dust density of 2.5 g 194 cm<sup>-3</sup> (D<sub>geom</sub>=D<sub>aerod</sub>/1.58). If original aerodynamic diameter data are already converted into 195 geometrical diameter, we replace the original correction with the conversion factor of 1.58. 196 Since the correction is a multiplicative factor the dlogD of the bins remain unchanged;
- 197 data provided as optical diameters (from OPCs) are converted into sphere-equivalent 198 geometric diameters applying the optical to geometrical correction by assuming homogeneous 199 spherical particles and a value of CRI of 1.53–0.003i. This CRI value is at the average of the dust 200 refractive indices reported in the 370-950 nm spectral range in Di Biagio et al. 2019) for dust of 201 global origin. Data for applying the correction for the different model of OPCs considered were 202 taken from Formenti et al. (2021) and conversion factors were recalculated at the dlogD path 203 of 0.05 assumed in the interpolated sizes. For the GRIMM 1.108 for which calibration is not 204 provided in Formenti et al. (2021) we used the data taken from Formenti et al. (2011) 205 interpolated at the 0.05 dlogD path of our diameters. In order to avoid discontinuities appearing and because of the new dlogD do not significantly differ on average from the value of 0.05 for 206 207 D<sub>geom</sub> calculated from D<sub>opt</sub> interpolated data, we do not update the dlogD, so that the conversion only imply a shift of the diameter. More details on the choices applied for corrections in 208 209 different cases are provided in Text S4. Original datasets already converted into geometrical 210 diameter, are left unchanged. However, it is worth noting that the ensemble of data already 211 applying an optical to geometrical correction uses a CRI varying between 1.53 and 1.55 for the 212 real part and 0.001 and 0.004 for the imaginary part and work under the hypothesis of 213 homogeneous spherical particles (Mie theory), therefore consistent with our treatment. 214 Exceptions are Khalfallah et al. (2020) using a CRI of 1.43-0.00i as for quartz particles, and 215 González–Flórez et al. (2023) using a CRI of 1.49–0.0015i and also applying calculations in 216 ellipsoidal assumption instead of Mie theory. The only dataset not theoretically submitted to the optical to geometric correction is the one provided by Renard et al. (2018) using an OPC 217 218 built with a specific geometry making the measurements very low sensitive to CRI calibration.

4/ Level-2b (LEV2b): based on LEV1, the LEV2b data treatment aims at harmonizing the size distributions
by converting the operational original particle diameters into a common-defined geometrical diameter
by taking into account that mineral dust is aspherical. Data from LEV1 are treated as in the following
with respect to their diameter corrections:

- o data already provided as geometrical diameters from coulter counters are left unchanged. This
   technique is in fact only slightly affected by shape effects, as discussed by Kobayashi et al.
   (2007);
- o data provided as projected-area diameters are corrected using the size-invariant correction
   factor of 1.56 from Huang et al. (2021) (D<sub>geom</sub>=D<sub>area</sub>/1.56) (see Eq. S1);
- o data provided as aerodynamic diameter are corrected assuming a size-invariant conversion
   factor of 1.45 following Huang et al. (2021) (D<sub>geom</sub>=D<sub>aerod</sub>/1.45) (see Eq. S2);
- 230 data provided as optical diameters and already treated as for LEV2a data, are further corrected 0 231 by applying a size-dependent aspherical to spherical ratio (ASR(D<sub>geom</sub>)) correction function,  $ASR(D_{geom})=(D_{geom})_{aspherical}/(D_{geom})_{spherical}$ , to take into account non–sphericity effects in optical to 232 233 geometrical conversion. The ASR function (Fig. S1) is obtained by combining the optical to 234 geometrical diameter conversion factors for different OPCs calculated by Formenti et al. (2021) 235 and Huang et al. (2021) both in the assumption of spherical homogeneous particles (Dgeom)spherical 236 and tri-ellipsoids dust (D<sub>geom</sub>)<sub>aspherical</sub>. More details are provided in Text S3. Original datasets 237 derived from OPC measurements already provided as geometrical diameter but under 238 assumption of sphericity are also corrected by applying the ASR(D<sub>geom</sub>) converting function. The only exception are González-Flórez et al. (2023), that already apply tri-axial ellipsoids 239 240 calculations in their optical to geometric conversion, and Renard et al. (2018), not requiring 241 optical to geometrical conversion.
- As for LEV1, the LEV2a and LEV2b data, for which a known interpolation path is used, are normalized so that the integral of the volume size distribution is 1 over a common diameter range ( $1.58 - 7.1 \mu m$  for SOURCE,  $0.71 - 8.9 \mu m$  for MRT, LRT).
- For each category (SOURCE, MRT, LRT) and for each data level (LEV1, LEV2a, LEV2b), the mean, median, and standard deviation of the particle volume concentration per size class are calculated where at least 2 datasets are available in the diameter range. 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.

## 250 **2.4. Limitations of the chosen approach**

251 Some precisions should be given when considering the LEV2a and LEV2b treatment reported in this 252 work. First, the implicit assumption when applying LEV2a and LEV2b dataset corrections is that dust is the dominant aerosol species and possible effects due to internal or external mixing of dust with other 253 254 aerosol types are not taken into considerations (i.e., in the complex refractive index or shape factor 255 assumptions). Second, for those datasets that are obtained from the combination of different 256 techniques, namely DMPS+APS (Bates et al., 2002; Maring et al., 2000, 2003; Müller et al., 2010), 257 OPC+APS (Chen et al., 2011), SMPS + OPC (de Reus et al., 2000; Otto et al;, 2007; Denjean et al., 2016a, 258 2016b), DMPS + APS + microscopy (Kandler et al., 2011), or multiple OPC instruments (Reid et al., 2003b; 259 McConnell et al., 2008; Johnson and Osborne, 2011; Ryder et al., 2013a, 2013b, 2018; Rosenberg et al., 260 2014; Weinzierl et al., 2009, 2011, 2017), the choice is that of applying artefact corrections for the dominant instrument, often the one in the extended coarse mode range, and consider this correction 261 262 applicable to the whole diameter range. This is because when multiples instruments are used to build a 263 size distribution it is then not easy to reconstruct the steps of data analysis and merging from the original 264 work. It follows the subsequent considerations:

265 1/ the corrections applied for the aerodynamic and projected-area diameter apply a constant 266 size-invariant scaling factor to the ensemble of the size distribution data. In this approximation, if 267 the SMPS/DMPS is combined with aerodynamic or microscopy data, a correction factor between

- 1.45 and 1.58, depending on the level and the technique as detailed in the previous section, is
  applied in place of the factor 1 (spherical assumption) or 1.19 (aspherical assumption) (see Eq. S3)
  expected to convert the mobility diameter to geometrical diameter in LEV2a and LEV2b data. As a
  consequence, the submicron size is 20 to 58% finer than expected only due to mobility to geometrical
  conversion.
- 273 2/ A similar approach is used to correct datasets where OPC is the main used technique to size dust 274 particles together with the SMPS. For LEV2a data the Mie correction is applied to the full size 275 distribution, but being the size-dependent correction mostly inactive for submicron particles (i.e. 276 D<sub>geom</sub> ~ D<sub>opt</sub> for most OPCs), the approach is mostly equivalent at considering a mobility diameter 277 correction with a shape factor of 1. For LEV2b data, using OPC corrections induce a limited right 278 shifting of the size distribution compared to the one that would be obtained from mobility 279 conversion because of the magnitude of the ASR function (Fig. S1) compared to the shape factor of 280 1.19 assumed for aspherical dust.
- 3/ When datasets relying on multiple OPCs measurements, the assumption is that the "dominant" OPC that is the OPC covering the largest range and the coarsest sizes in particular, is considered. Given that optical to geometrical corrections are not relevant for submicron particles and that the magnitude of the correction typically increases for increasing sizes, this assumption is not expected to determine significant biases in the data. To mention additionally a general ambiguity of the optical to geometrical correction around the diameter of 1 µm where a plateau in the scattering calibration function for several OPCs models can be found (i.e. Formenti et al., 2021).
- 288 More details on the specific assumptions and choices done for each dataset are provided in **Text S4**.

Further, for LEV2a and LEV2b data for which corrections are applied on the data, caution is taken at the boundary of the size distribution and when the first and/or the last bin of the corrected size showed

- 291 significant divergence, these data are removed from the dataset.
- An additional source of erroris the individual measurement uncertainty, which varies with the specific setup, instrument and spatial and temporal extent of the measurement.
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## **3.** Presentation and discussion of the dataset

296 Illustration of the data for different levels is provided in Figure 2. Figure 3 presents the synthesis of the 297 LEV2b data and the comparison of SOURCE, MRT and LRT distributions. The contribution of different 298 size classes to the total particle number, surface and volume is summarised in Table 1. Size classes have 299 been defined according to the classification of Adeyemi et al. (2023) defining fine dust ( $D \le 2.5 \mu m$ ), 300 coarse dust ( $2.5 < D \le 10 \mu m$ ), super coarse dust ( $10 < D \le 62.5 \mu m$ ) and giant dust ( $D > 62.5 \mu m$ ). Within 301 the fine dust class, we further calculate the fractions of particles smaller than 0.4  $\mu m$ .

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Figure 2. Data for SOURCE, MRT, and LRT dust at level 1, 2a, and 2b as described in Sect. 2.3 (labelled as LEV1,
 LEV2a, LEV2b, respectively). Single datasets, all normalized at the integral of 1, are plotted as black lines. The mean
 (thick black, blue, and red line for SOURCE, MRT, and LRT, respectively) are shown at all levels. Note that the mean
 is calculated only where at least 2 datasets are available in the diameter range.

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**Figure 3.** Comparison of normalized mean volume size distribution for the SOURCE, MRT, and LRT categories in our study reported as LEV2b data (mean ± standard deviation). For the sake of comparison, and differently from data in Fig. 2, the

313 SOURCE, MRT, and LRT synthesis datasets reported here are normalized at the integral equal to 1 over a common diameter 314 range corresponding to 0.35–17.8 μm. This is done to remove differences linked to different integration range for SOURCE data

315 compared to MRT and LRT.

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| Datase  | t      | D ≤ 2.5 μm<br>(D ≤ 0.4 μm) | 2.5 < D ≤ 10 μm | 10 < D ≤ 62.5 μm | D > 62.5 μm |
|---------|--------|----------------------------|-----------------|------------------|-------------|
|         | SOURCE | 95.4% (20.4%)              | 4.5%            | 0.1%             | 0.4%        |
| Number  | MRT    | 99.8% (96.1%)              | 0.2%            | 0.0%             | 0.0%        |
|         | LRT    | 99.9% (94.5%)              | 0.1%            | 0.0%             | 0.0%        |
|         | SOURCE | 45.0% (1.1%)               | 39.4%           | 15.5%            | 0.14%       |
| Surface | MRT    | 65.4% (16.8%)              | 30.7%           | 3.6%             | 0.29%       |
|         | LRT    | 84.6% (23.1%)              | 15.1%           | 0.2%             | 0.00%       |
|         | SOURCE | 10.8% (0.1%)               | 34.9%           | 52.7%            | 1.6%        |
| Volume  | MRT    | 22.1% (1.1%)               | 44.3%           | 25.7%            | 8.0%        |
|         | LRT    | 53.4% (3.6%)               | 44.5%           | 2.0%             | 0.0%        |

318 319 **Table 1.** Percentages of number, surface and volume size distribution in the diameter ranges  $D \le 0.4 \ \mu$ m,  $D \le 2.5 \ \mu$ m,  $2.5 < D \le 10 \ \mu$ m,  $10 < D \le 62.5 \ \mu$ m, and  $D > 62.5 \ \mu$ m for the mean of the size obtained for the SOURCE, MRT, and LRT LEV2b datasets.

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321 As shown in Fig. 2 and 3 the shape of the dust size distribution at emission and along transport shows 322 main consistent features. A main mode located at ~10 µm (in volume) is observed for dust at emission 323 and close to sources, as based from the few studies allowing to measure up to the coarse fraction. The 324 main dust mode decreases to  $\sim$ 5  $\mu$ m and  $\sim$ 2  $\mu$ m for MRT and LRT conditions, respectively. Below 0.4  $\mu$ m 325 the dust volume size shows an additional mode, particularly visible for MRT and LRT. As a matter of fact, 326 the sparse datasets measuring very fine particles at the SOURCE show that particles with diameters 327 below 0.4 µm (however measured only down to 0.2 µm, as shown in Fig. 2) represent approximately 328 20% of the total particles' number, increasing to more than 90% in MRT and LRT. Instruments such as 329 SMPS and DMPS used in MRT and LRT studies measure particles as small as 0.02  $\mu$ m in diameter. 330 Previous single-particle compositional observations showing that the particle number concentration in 331 the size range between 0.1 and 0.4 µm is largely contributed by aluminosilicate dust particles at 332 emission, while internal or external mixing with aerosols other than dust gains importance with time 333 and altitude of transport (Chou et al., 2008; Kandler et al., 2007, 2009; Weinzierl et al., 2009; 2017; 334 Klaver et al., 2011; Denjean et al., 2016a; 2016b).

The normalized size distribution of dust particles between 0.4 and 10  $\mu$ m is rather consistent and invariant along the dust cycle. This is true in particular when restricting to the 2.5 to 10  $\mu$ m size range when differences are minimal and contribution to total volume is in between 34.9% and 44.5%. Below that range, which is between 0.4 and 2.5  $\mu$ m, the contribution of particles for LRT is significantly higher (53.4% in volume) than for SOURCE (10.8%) and MRT (22.1%), likely as, because of the normalization, it compensates the decrease of particles larger than 10  $\mu$ m.

The magnitude of the particle volume above 10 µm remains unchanged almost up to 100 µm for both the SOURCE and the MRT conditions, which also present similar particle volume. This mode decreases very strongly for LRT conditions, when it represents only 2% of the total volume, compared to almost 55% and 34% for SOURCE and MRT, respectively.

The dataset presented in this work, synthetizing available in situ observations, allows evaluation of the 345 346 natural variability of dust size distribution along its lifecycle. To be emphasized, however, that while 347 consistent differences in the mean size distribution curves are obtained going from SOURCE to LRT, as 348 shown in Fig. 3, the inherent range of variability for each category, represented by the standard 349 deviation of the data, is also non-negligible and reflects the large range of documented size 350 distributions, together with the limited statistics available. This is particularly true for both super-coarse 351 and giant dust at MRT and LRT. Lower variability is identified below 0.4 µm because of the restricted 352 number of dataset available for MRT and LRT conditions, and there is an absence of data for SOURCE 353 dust below this size range.

354 Finally, to facilitate the use of these data within models and remote sensing schemes, Table 2 provides 355 the parameters of lognormal size distributions fitting the LEV2a and LEV2b mean values of the three 356 dust categories. Lognormal functions are set to reproduce the main shape of the dust distribution above 357 0.4 µm, neglecting the specific features below this diameter where information is lower and the size 358 affected by particle mixing with other compounds, especially for MRT and LRT. We found that a single 359 broad mode can be employed to represent the main features of the volume size distributions above 0.4 360 μm. Plots of the fitting functions are provided in supplementary Fig. S4. Because there is an inherent 361 level of subjectivity in the choice of the number of modes and their parameters, we invite the individual 362 researchers using the data to implement the parameterizations in accordance to their scientific needs.

- 363
- 364

| Dataset         | Lognormal mode            |          |  |          |      |  |
|-----------------|---------------------------|----------|--|----------|------|--|
|                 | N <sub>tot</sub> (# cm⁻³) | NMD (μm) | V <sub>tot</sub> (nm <sup>3</sup> cm <sup>-3</sup> ) | VMD (µm) | σ    |  |
| SOURCE – LEV2a  | 5.08 10 <sup>-10</sup>    | 0.355    | 7.76   | 26.69    | 3.32 |  |
| SOURCE – LEV 2b | 9.8 10 <sup>-10</sup>     | 0.300    | 3.38   | 11.71    | 3.02 |  |
| MRT – LEV 2a    | 2.11 10 <sup>-9</sup>     | 0.150    | 2.55   | 11.64    | 3.33 |  |
| MRT – LEV 2b    | 6.82 10 <sup>-9</sup>     | 0.100    | 1.57   | 5.79     | 3.20 |  |
| LRT – LEV 2a    | 2.35 10 <sup>-9</sup>     | 0.280    | 1.39   | 3.88     | 2.55 |  |
| LRT – LEV 2b    | 2.96 10 <sup>-9</sup>     | 0.350    | 1.15   | 2.34     | 2.22 |  |

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366

367 **Table**. Parameters (total number and volume concentration,  $N_{tot}$  (# cm<sup>-3</sup>),  $V_{tot}$  (nm<sup>3</sup> cm<sup>-3</sup>), number and volume median diameter, 368 NMD and VMD (µm), geometric standard deviation,  $\sigma$ ) for the log-normal modes used to parameterize the LEV2b volume size 369 distributions of the SOURCE, MRT, and LRT categories. Parameters refers to the following equations:  $\frac{dV}{dlogD} =$ 370  $\frac{\pi}{6}D^3 \frac{N_{tot}}{\sqrt{2\pi}log\sigma} exp\left(-\frac{(logD-logNMD)^2}{2(log\sigma)^2}\right)$  and  $\frac{dV}{dlogD} = \frac{V_{tot}}{\sqrt{2\pi}log\sigma} exp\left(-\frac{(logD-logVMD)^2}{2(log\sigma)^2}\right)$ 

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#### 372 4. Conclusive remark

In this paper we present the most possible comprehensive synthesis of *in situ* observations of the particle size distribution of atmospheric dust aerosols. This compilation reflects the current state—of the—art and represents a standardized and synthetic benchmark to constrain and evaluate models and satellite retrievals. We highlight differences and commonalities of the dust volume distribution as a function of time in the atmosphere, both in terms of main identified modes and relative contribution of dust in different size ranges.

We did this based on a large statistics of data and permit to retrieve robust information between 0.4 and 10  $\mu$ m where most of observations exist, while above and below this size range, observations are rare. Dust particles below 0.4  $\mu$ m in diameter are seldom measured close to source regions, but are found in observations at mid– and long–range transport conditions. Their presence at emission, their size–segregated composition and state of mixing should be better documented and understood. The dynamics of the coarse mode above 10  $\mu$ m, its invariance from source to mid-range transport, and decline afterwards is reported, and can challenge models.

We acknowledge the evidence that the compilation of a reference dataset is, almost by definition, a subjective and incomplete exercise which must revised continuously with the emergence of new datasets, new field campaigns, and the improvement of sampling techniques. We henceforth encourage colleagues to provide us with new or revised datasets to feed and update the dataset in the future.

390 Data availability

- 391 The LEV1, LEV2a and LEV2b datasets discussed in this paper are available on the EaSy Data, the Earth
- 392 System Data repository (https://www.easydata.earth/#/public/home, last access: 01 June 2024) 393 maintained by the National French DATA TERRA research Infrastructure. Their respective DOIs are
- 394 summarized here below:
- 395
   SOURCE\_LEV1.dat, SOURCE\_LEV2a.dat, SOURCE\_LEV2b.dat : https://doi.org/10.57932/58dbe908-9394-4504 

   396
   9099-74a3e77140e9 (Formenti and Di Biagio, 2023a);
- 397
   MRT\_LEV1.dat,
   MRT\_LEV2a.dat,
   MRT\_LEV2b.dat:
   <a href="https://doi.org/10.57932/31f2adf7-74fb-48e8-a3ef-398">https://doi.org/10.57932/31f2adf7-74fb-48e8-a3ef-398</a>

   398
   059f663c47f1
   (Formenti and Di Biagio, 2023b);
   <a href="https://doi.org/10.57932/31f2adf7-74fb-48e8-a3ef-398">https://doi.org/10.57932/31f2adf7-74fb-48e8-a3ef-398</a>
- 399
   LRT\_LEV1.dat,
   LRT\_LEV2a.dat,
   LRT\_LEV2b.dat :
   https://doi.org/10.57932/17dc781c-3e9d-4908-85b5 

   400
   5c99e68e8f79 (Formenti and Di Biagio, 2023c).
   6
- 401 Figures of the individual datasets (including LEV0) are provided upon request.
- 402 Code availability. Data from images on published papers were digitalized with the online
   403 WebplotDigitizer software available at <a href="https://automeris.io/WebPlotDigitizer/">https://automeris.io/WebPlotDigitizer/</a>
- 404 Author contributions. PF and CDB designed the research, compiled and analysed the dataset, and wrote
   405 the manuscript.
- 406 **Competing interests**. The authors declare that they have no competing interests.
- 407 **Special issue statement.** The paper is not associated with a special issue.

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