

# Complex refractive indices and single scattering albedo of global dust aerosols in the shortwave spectrum and relationship to iron content and size

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#### Complex refractive indices and single scattering albedo of global dust 2 aerosols in the shortwave spectrum and relationship to iron content and size

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# **Abstract**

The optical properties of airborne mineral dust depend on its mineralogy, size distribution, shape, and might vary between different source regions. To date, large differences in refractive index values found in the literature have not been fully explained. In this paper we present a new dataset of complex refractive indices (m=n-ik) and single scattering albedos (SSA) for 19 mineral dust aerosols over the 370-950 nm range in dry conditions. Dust aerosols were generated from natural parent soils from eight source regions (Northern Africa, Sahel, Middle East, Eastern Asia, North and South America, Southern Africa, and Australia). These were selected to represent the global scale variability of the dust mineralogy. Dust was re-suspended into a 4.2 m³ smog chamber where its spectral shortwave scattering (βsca) and absorption ( $\beta_{abs}$ ) coefficients, number size distribution, and bulk composition were measured. The complex refractive index was estimated by Mie calculations combining optical and size data, while the spectral SSA was directly retrieved from  $\beta_{sca}$  and  $\beta_{abs}$  measurements. Our results show that the imaginary part of the refractive index (k) and the SSA largely vary from sample to sample, with values for k in the range 0.001 to 0.009 at 370 nm and 0.0003 to 0.002 at 950 nm, and values for SSA in the range 0.70 to 0.96 at 370 nm and 0.95 to 0.99 at 950 nm. In contrast, the real part of the refractive index (n) is mostly source (and wavelength) independent, with an average value between 1.48 and 1.55. The sample-to-sample variability in our dataset of k and SSA is mostly related to differences in the dust's iron content. In particular, a wavelength-dependent linear relationship is found between the magnitude of k and SSA and the mass concentrations of both iron oxide and total elemental iron. As an intrinsic

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- 46 property of matter, k is independent of size. When the iron oxide content exceeds >3%, the SSA linearly
- decreases with increasing fraction of coarse particles at short wavelengths (< 600 nm).
- 48 We recommend that source-dependent values of the SW spectral refractive index and SSA are used
- 49 in models and remote sensing retrievals instead of generic values. In particular, the close relationships
- 50 found between k/SSA and the iron content in dust enable establishing predictive rules for spectrally-
- 51 resolved SW absorption based on particle composition.

52 53

#### Introduction

- 54 With teragrams of annual emissions, a residence time of about 1-2 weeks in the atmosphere, and a
- 55 planetary-scale transport, mineral dust aerosols are a global phenomenon (Uno et al., 2009; Ginoux et
- 56 al., 2012), and contribute significantly to the global and regional aerosol loading (Ridley et al., 2016)
- 57 and direct radiative effect (Miller et al., 2014).
- 58 However, large uncertainties still persist on the magnitude and overall sign of the dust direct radiative
- 59 effect (Boucher et al., 2013; Highwood and Ryder, 2014; Kok et al., 2017). One of the major sources of
- 60 this uncertainty is our insufficient knowledge of the dust's absorption properties in the shortwave (SW)
- and longwave (LW) spectral ranges (e.g., Balkanski et al., 2007; Samset et al., 2018), given that mineral
- dust contains large particles and a variety of minerals absorbing over both spectral regions (e.g. iron
- 63 oxides, clays, quartz and calcium-rich species, Sokolik and Toon, 1999; Lafon et al., 2006; Di Biagio
- et al., 2014a, b). Global and regional scale mapping of dust absorption remains limited and more infor-
- 65 mation is required (Samset et al., 2018).
- 66 Aerosol absorption is represented both by the imaginary part (k) of the complex refractive index (m=n-
- 67 ik) of its constituent material, and by the single scattering albedo (SSA, i.e., the ratio of the scattering
- 68 to extinction coefficient) of the particle population, as well as by the mass absorption efficiency (MAE,
- units of m<sup>2</sup> g<sup>-1</sup>), i.e., the absorption coefficient per unit of aerosol mass concentration.
- 70 In the shortwave spectral range, absorption by dust accounts for up to ~10–20% of its total extinction.
- 71 Dust absorption is highest in the UV-VIS, and almost nil towards the near IR (Cattrall et al., 2003;
- 72 Redmond et al., 2010), due to the combined contribution of large particles in the size distribution and
- 73 the dust's mineralogy, notably the presence of iron oxides (Karickhoff and Bailey, 1973; Lafon et al.,
- 74 2006; Derimian et al., 2008; Moosmüller et al., 2012; Formenti et al., 2014a; 2014b; Engelbrecht et al.,
- 75 2016; Caponi et al., 2017). The mineralogy of mineral dust varies according to that of the parent soils
- 76 (Nickovic et al., 2012; Journet et al., 2014). Consequently, dust aerosols of different origins should be
- 77 more or less absorbing in the SW, and have different imaginary spectral refractive index and SSA. Field
- and laboratory measurements, including ground-based and spaceborne remote sensing, show that k
- varies at a regional scale by almost two orders of magnitude (0.0001-0.008 at 550 nm) with corre-
- sponding SSAs between 0.80 and 0.99 at 550 nm (Volz 1972; Patterson et al., 1977; Shettle and Fenn,
- 1979; Dubovik et al., 2002; Haywood et al., 2003; Sinyuk et al., 2003; Linke et al., 2006; Osborne et al.,
- 82 2008; Müller et al., 2009; Otto et al., 2009; Petzold et al., 2009; Schladitz et al., 2009; McConnell et al.,
- 83 2010; Formenti et al., 2011; Wagner et al., 2012; Ryder et al., 2013a; Engelbrecht et al., 2016; Rocha-

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85 size fraction or in the method used to retrieve optical parameters), differences persist when the same 86 measurement approach and retrieval method are applied, e.g., in AERONET inversions, supporting the 87 dependence of dust k and SSA with location (Dubovik et al., 2002; Koven and Fung, 2006; Su and 88 Toon, 2011). In contrast, the real part (n) of the dust refractive index, mostly related to particle scatter-89 ing, is estimated to be less variable than the imaginary part, with values between 1.47-1.56 at 550 nm 90 (i.e., Volz, 1972; Patterson et al., 1977; Balkanski et al., 2007; Petzold et al., 2009). 91 Differences in k or SSA caused by the spatial variability of the iron content may affect the sign of the 92 dust radiative effect (heating vs cooling) (Liao and Seinfeld, 1998; Claquin et al. 1999; Miller et al., 93 2014), and its global and regional implications (Myhre and Stordal, 2001; Colarco et al., 2014; Das et 94 al., 2015; Jin et al., 2016; Bangalath and Stenchikov, 2016; Strong et al., 2018). For instance, various 95 studies suggest that the direct radiative effect of dust has a strong impact on the Western African Mon-96 soon (Yoshioka et al., 2007; Konaré et al., 2008) and the Indian Summer Monsoon (Vinoj et al., 2014; 97 Das et al., 2015; Jin et al., 2016). However, there is no consensus whether dust increases or decreases 98 precipitation over these regions (Solmon et al., 2008; Jin et al., 2016; Strong et al., 2018). As an exam-99 ple, Solmon et al. (2008) indicate that dust reduces precipitation over most of the Sahelian region, but 100 increases it over the Northern Sahel-Southern Sahara. This pattern is, however, very sensitive to the 101 dust absorbing properties, and a decrease of few percent in dust absorption may even cancel out the 102 increase of precipitation over the Sahel. Similarly, Jin et al. (2016) show that by varying k from zero to 103 0.008 at 600 nm (i.e., the highest value currently used in models) the dust effect on the Indian Summer 104 Monsoon may shift from negative (reduction of precipitation) to positive (increase of precipitation) val-105 ues. 106 In spite of this sensitivity, present climate models adopt a globally-constant spectral complex refractive 107 index (and SSA) for dust, and hence still implicitly assume the same dust mineralogical composition at 108 the global scale. Reference values for the refractive index are usually taken from Volz (1972), Patterson 109 et al. (1977), D'Almeida et al. (1991), Shettle and Fenn (1979), Sokolik et al. (1993), Sinyuk et al. (2003), 110 or OPAC (Hess et al., 1998; Koepke et al., 2015). A parameterization of the spectrally-resolved dust 111 refractive index as a function of the mineralogical composition of the particles is desirable to replace 112 the globally constant values in current climate models, in particular for those models that started to incorporate the representation of dust mineralogy into their schemes (Scanza et al., 2015; Perlwitz et 113 114 al., 2015a, 2015b). 115 Improving our knowledge of the spectral SW refractive index of mineral dust and its relation to particle composition and origin is also key for the detection of dust aerosols in the atmosphere and the quanti-116 117 fication of its mass loading, and total and absorption spectral optical depth from active and passive 118 remote sensing (e.g., Ridley et al., 2016). As an example, the retrieval of the dust SSA and optical depth 119 over bright desert surfaces with the MODIS (Moderate Imaging Resolution Spectroradiometer) Deep 120 Blue algorithm (Hsu et al., 2004) applies the Critical Surface Reflectance Method (Kaufman, 1987) to retrieve dust properties from measured Top of Atmosphere (TOA) spectral reflectance. This algorithm 121 122 depends critically on a priori information on the spectral refractive index (Kaufman et al., 2001; Yoshida

Lima et al., 2018). Albeit some variability being instrumental or analytical (differences in the sampled

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et al., 2013). Similarly, active remote sensing techniques (lidar, light detection and ranging) require the knowledge of the extinction-to-backscatter ratio (the lidar ratio), which is also a strong function of the complex index of refraction or SSA of the aerosol particles (e.g., Gasteiger et al., 2011; Shin et al., 2018). Gasteiger et al. (2011) have shown in fact that a 5% change in the SSA at 532 nm can modify by up to 20% the lidar ratio of dust, which means a 20% change in the estimated profile of the dust extinction coefficient and retrieved optical depth from lidar measurements.

In this paper we address these issues by reporting the results of a new laboratory investigation of the shortwave refractive index and SSA of dust from various source regions worldwide, in the framework of the RED–DUST project (Di Biagio et al., 2017a; hereafter DB17; Caponi et al., 2017; hereafter C17). Dust optical properties at discrete wavelengths between 370 and 950 nm are derived in conjunction with the particle elemental and mineralogical composition, including total elemental iron and iron oxides. We investigate the relationship of k and SSA to the iron content to provide a parameterization of the dust absorption as a function of its mineralogy, which can be applied to climate models. The dependence of dust absorption on the particle coarse size fraction is also investigated to evaluate the change of dust absorption with atmospheric transport time.

#### 2. Experimental set-up and instrumentation

All experiments discussed here and previously described in DB17 and C17 were conducted in the 4.2 m<sup>3</sup> stainless-steel CESAM chamber (French acronym for Experimental Multiphasic Atmospheric Simulation Chamber) (Wang et al., 2011). Mineral dust aerosols were generated by mechanical shaking of parent soils using about 15 g of soil sample (first sieved to <1000 µm and then dried at 100 °C) placed in a 1 L Büchner flask and shaken for about 30 min at 100 Hz by means of a sieve shaker (Retsch AS200). The dust suspension in the flask was injected into the chamber by flushing with N2 at 10 L min<sup>-1</sup> for about 10–15 min. After injection in the chamber, the largest fraction of the dust aerosol (>1.5 µm diameter) remained in suspension for approximately 60 to 120 min thanks to a four-blade stainless steel fan located at the bottom of the chamber, which also ensured homogeneous conditions within the chamber volume. The submicron dust fraction, instead, remained constant with time during experiments, as shown in Fig. 3 reporting the timeline of the measured effective dust fine diameter. The evolution of the physico-chemical and optical properties of the suspended dust was measured by different instruments connected to the chamber. The spectral particle volume dry scattering (β<sub>sca</sub>) and absorption (β<sub>abs</sub>) coefficients were measured, respectively, by a 3-wavelength nephelometer (TSI Inc. model 3563, operating at 450, 550, and 700 nm; 2 L min<sup>-1</sup> flow rate, 2-s time resolution) and a 7wavelength aethalometer (Magee Sci. model AE31, operating at 370, 470, 520, 590, 660, 880 and 950 nm; 2 L min-1 flow rate, 2-min time resolution). The size distribution of aerosols was measured by means of a scanning mobility particle sizer (SMPS, TSI, DMA Model 3080, CPC Model 3772; mobility diameter range 0.019-0.882 µm; 2.0/0.2 L min-1 sheath-aerosol flow rates, 135-s time resolution), a WELAS optical particle counter (OPC) (PALAS, model 2000, white light source between 0.35 and 0.70 μm; optical-equivalent diameter range 0.58-40.7 μm; 2 L min<sup>-1</sup> flow rate, 1-min time resolution) and a SkyGrimm OPC (Grimm Inc., model 1.129, 0.655 µm operating wavelength; optical-equivalent diameter range 0.25-32 μm; 1.2 L min-1 flow rate, 6-s time resolution). Aerosol elemental and mineralogical

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162 composition, including iron oxides, was derived by analysis of dust samples collected on polycarbonate
 163 filters (47-mm diameter Nuclepore, Whatman, nominal pore size 0.4 μm) mounted on a custom–made
 164 stainless–steel sample holder (operated at 6 L min<sup>-1</sup>) for most of the duration of each experiment.

All instruments (size, SW optics, filters) sampled air from the chamber. To equalize the airflow extracted by the different instruments, a particle–free  $N_2/O_2$  mixture airflow was continuously injected into the chamber. Inlets for all extractive measurements consisted of a stainless steel tube located inside CESAM, and an external connection of silicone tubing (TSI Inc.) from the chamber to the instruments, for a total length varying between 0.4 and 1.2 m. As detailed in DB17 and shown in Fig. S1 in the supplement, the transmission efficiency due to aspiration and transmission in the sampling lines as a function of particle diameter was estimated to calculate the effective dust fraction sensed by each instrument, taking into account the sampling flow rate, tubing diameter, tubing geometry, and particle shape and density. For the nephelometer and the aethalometer, the length of the sampling line from the intake point in the chamber to the instrument entrance was about 1.2 m, which resulted in a 50% cutoff of the transmission efficiency at 3.9  $\mu$ m particle geometric diameter and 100% cutoff at 10  $\mu$ m. For the filter sampling system, the length of the sampling line of about 0.5 m resulted in a 50% (100%) cutoff at 6.5  $\mu$ m (15  $\mu$ m) particle diameter. For the WELAS, the only OPC considered for size distribution in the coarse fraction (see Sect. 2.2), the 50% (100%) cutoff was reached for particles of 5  $\mu$ m (8  $\mu$ m) diameter.

All experiments were conducted at ambient temperature and relative humidity <2%. In addition to overnight evacuation, the chamber was manually cleaned between experiments to avoid contaminations from remaining dust. Background concentrations of aerosols in the chamber were less than 2.0 µg m<sup>-3</sup> (that is 10<sup>2</sup> to 10<sup>5</sup> times less than the concentration of dust aerosols in suspension in the chamber

A flowchart of the procedure used to treat and combine optical, size, and compositional data, and the algorithm for SSA and complex refractive index retrieval is shown in Fig. 1. Full details of data treatment for size distribution measurements and filter compositional data are provided in DB17 and C17, and in the following we only mention the main points of interest for the present paper. Full details on the data

treatment of the SW optical data are provided in Sect. 2.1 and 3.

The optical and size datasets were acquired at different temporal resolutions and then averaged over compatible 10–min intervals, whereas the compositional data represent the experiment integral. The SSA and complex refractive index data were retrieved both at 10–min resolution and as experiment averages to relate them to both size and compositional data. Table 1 summarizes the uncertainties on the measured and derived parameters described in the following.

### 2.1 SW optical measurements

during experiments)

## 196 2.1.1 Aerosol scattering coefficient

The aerosol scattering coefficients ( $\beta_{sca}$ ) at 450, 550, and 700 nm are measured by the nephelometer at angles between 7° and 170° and need to be corrected for the restricted field–of–view of the instrument (truncation correction) to retrieve  $\beta_{sca}$  at 0°–180°. The truncation correction factor ( $C_{trunc}$ ), i.e., the

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ratio of the β<sub>sca</sub> at 0°-180° and 7°-170°, was estimated by Mie calculations for homogeneous spherical particles using the size distribution measured simultaneously behind SW inlets (see Sect. 2.2). In the calculations, the real part of the complex refractive index of dust was assumed to be wavelength-independent and fixed at a value of 1.53, while the imaginary part was set to 0.003 at 450 and 550 nm and to 0.001 at 700 nm, according to pre-existing information (Sinyuk et al., 2003; Schladitz et al., 2009; Formenti et al., 2011; Rocha-Lima et al., 2018). For the different dust samples, Ctrunc ranged between 1.2 and 1.7 and decreased with wavelength and the dust residence time in the chamber, following the relative importance of the coarse component in the dust population (Anderson and Ogreen, 1998). The uncertainty on Ctrunc, calculated by repeating the optical calculations by using the size distribution of dust within its error bars as input to the optical code, is less than ±5% at all wavelengths (in the approximation of Mie spherical and homogeneous particles). In order to assess the consistency of the derived truncation correction, we made a sensitivity study in which we recalculated Ctrunc by varying the refractive index at input to Mie calculations in the range of n and k values obtained in this study (i.e., values at the 10% and 90% percentile as reported in Table 5 for the whole dataset, that is n between 1.49 and 1.54 and k between 0.001 and 0.006 at 450, 550, and 700 nm). The results of this sensitivity study indicate that, for fixed dust size distribution, the truncation correction Ctrunc varies less than 1% for n between 1.49 and 1.54, and <5% for k between 0.001 and 0.006, and so that it is quite insensitive to the exact assumed n and k values.

- Once corrected for truncation, the spectral  $\beta_{sca}$  was extrapolated at the aethalometer wavelengths. With this aim, the Scattering Ångström Exponents, SAE<sub>450-550</sub> and SAE<sub>550-700</sub>, were calculated as the linear fit of  $\beta_{sca}$  vs  $\lambda$  at 450-550 nm and 550-700 nm, respectively. The SAE<sub>450-550</sub> and SAE<sub>550-700</sub> coefficients were used to extrapolate  $\beta_{sca}$  at wavelengths respectively lower and higher than 550 nm. Extrapolated  $\beta_{sca}$  values were used to derive an average SAE of dust for the entire investigated spectral range.
- 223 2.1.2 Aerosol absorption coefficient
- The aerosol absorption coefficient (β<sub>abs</sub>) at 370, 470, 520, 590, 660, 880, and 950 nm was retrieved from aethalometer measurements. The aethalometer measures the attenuation (ATT) through an aerosol–laden quartz filter, related to the spectral attenuation coefficient (β<sub>ATT</sub>) as:

$$\beta_{ATT}(\lambda) = \frac{\Delta ATT(\lambda)}{\Delta t} \frac{A}{V}$$
 (1)

- 228 where A is the area of the aerosol collection spot  $(0.5 \pm 0.1)$  cm<sup>2</sup> and V the air sample flow rate (0.002)
- 229 m³ min-1). The slope  $\frac{\Delta ATT(\lambda)}{\Delta t}$  is the linear fit of the measured attenuation as a function of time calcu-
- lated over 10-min intervals. The spectral attenuation coefficient was converted into an absorption co efficient β<sub>abs</sub> following the formula by Collaud Coen et al. (2010):

$$\beta_{abs}(\lambda) = \frac{\beta_{ATT}(\lambda) - \alpha(\lambda)\beta_{sca}(\lambda)}{C_{ref}R(\lambda)}$$
 (2)

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233 The  $\alpha(\lambda)\beta_{sca}(\lambda)$  term accounts for the fraction of the measured attenuation due to side and backward 234 scattering and not to light absorption. The Collaud-Coen correction scheme has been recently shown 235 to yield quite accurate values of the absorption coefficients and absorption Ångström exponents from aethalometer data (Saturno et al., 2017). The value of  $\alpha(\lambda)$  was calculated with the formula by Arnott et 236 237 al. (2005) and varied between 0.002 and 0.02 (<±1% from formal error propagation on the Arnott for-238 mula), while  $\beta_{sca}(\lambda)$  is the scattering coefficient from the nephelometer extrapolated to the aethalometer 239 wavelengths. Cref accounts for multiple scattering by the filter fibers, aerosol laden or not. Its spectral 240 value, obtained by the linear extrapolation of Cref at 450 and 660 nm estimated for mineral dust by Di 241 Biagio et al. (2017b), varied between 4.30 at 370 nm to 3.32 at 950 nm. We assume for the extrapolated 242 Cref an uncertainty of ±10% as estimated in Di Biagio et al. (2017b). The correction factor, R, accounts 243 for the decrease in the aethalometer sensitivity with the increase of the aerosol filter loading. The value 244 of R depends on the absorptivity properties of the sampled aerosol and can be calculated as a function 245 of the particle SSA. In this study, we calculated R by estimating a first-guess SSA\* as the ratio of the 246 nephelometer-corrected  $\beta_{sca}$  and  $\beta_{ext}$  obtained as the sum of  $\beta_{sca}$  and the  $\beta_{abs}$  non-corrected for filter 247 loading effect. The R was estimated by using the Collaud-Coen et al. (2010) formulation. For the range of estimated SSA\* (about 0.60 to 0.99), R varied between 0.5 and 1.0 (±1-10%). 248

- 249 The Absorption Ångstrom Exponent (AAE) was calculated as the power–law fit of  $\beta_{abs}$  versus  $\lambda$ .
- 250 Due to an instrumental problem, aethalometer data were not always available, with a typical 30-min
- 251 interruption usually 10 to 30 minutes after the beginning of experiments.

#### 252 2.2 Size distribution

253 The aerosol number size distribution was obtained from SMPS, WELAS and SkyGrimm measurements 254 over different diameter ranges. The measured electrical mobility and optical equivalent diameters from 255 the SMPS and the OPCs were first converted into geometrical diameters (Dg) as described in DB17 256 and summarized in Table 1. The OPCs conversion assumes for dust a complex refractive index that in

our study was set in the range 1.47-1.53 for n and 0.001-0.005 for k for both the SkyGrimm and the 257

258 WELAS (following DB17). After conversion, the estimated D<sub>g</sub> range was 0.01–0.50 μm for the SMPS,

259 0.65-73.0 µm for the WELAS, and 0.29-68.2 µm for the SkyGrimm. Due to a calibration issue, data for

260 the SkyGrimm in the range D<sub>9</sub> > 1µm were discarded, so that the WELAS is the only instrument consid-

261 ered in the super-micron range. A very low counting efficiency was observed for the WELAS below 1

262 µm and data in this size range were also discarded.

263 The SMPS, WELAS, and SkyGrimm data were combined, as detailed in DB17, to obtain the full size

264 distribution of the dust aerosols suspended in the CESAM chamber, (dN/dloqD<sub>a</sub>)<sub>CESAM</sub>, and the size

distribution behind SW optical instruments inlets, (dN/dlogDg)SWoptics, after taking into account particle 265 266

losses along sampling lines (see Supplementary material and Fig. S1). As previously discussed, due

to the particle losses in the sampling line from the chamber to the nephelometer/aethalometer, the

(dN/dlogD<sub>g</sub>)<sub>SWoptics</sub> size distribution is cut at 10 μm, so no particles above this diameter reach the SW 268

269 instruments.

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The measured size distributions,  $(dN/dlogD_g)_{CESAM}$  and  $dN/dlogD_g)_{SWoptics}$ , were used to estimate the mass concentration of aerosols and their effective diameter (D<sub>eff</sub>) in the CESAM chamber and behind

the SW instrument inlets as:

$$D_{\text{eff}} = \frac{\int D_g^3 \frac{dN}{d\log D_g} d\log D_g}{\int D_g^2 \frac{dN}{d\log D_g} d\log D_g}$$
(4)

The effective dust density ρ in Eq. (3) was set at 2.5 g cm<sup>-3</sup>, a value that is approximately in the middle
of the range of desert dust densities reported in the literature, i.e., 2.1–2.75 g cm<sup>-3</sup> (Maring et al., 2000;
lwasaka et al., 2003; Reid et al., 2003). The effective diameter was evaluated separately for the fine
and coarse fractions of dust by integrating Eq. (4) for diameters ≤1 μm (D<sub>eff,fine</sub>) and >1 μm (D<sub>eff,coarse</sub>),

279 respectively.

OPC correction.

The dust size distribution,  $(dN/dlogD)_{SWoptics}$ , measured at each 10–min time step for each sample was fitted with a sum of five lognormal functions. For each mode, the parameters of the lognormal functions, i.e., the total number concentration  $(N_i)$ , the geometric median diameter  $(D_{g,i})$ , and the geometric standard deviation of the distribution  $(\sigma_i)$ , were retrieved. The uncertainties in the retrieved parameters were estimated by repeating the fit using size data within their uncertainties. The resulting parameters of the fits at the peak of the injection in the chamber are reported in Table S1, and an example of size fitting is shown in Fig. S2.

The procedure described here to estimate  $(dN/dlogD_g)_{CESAM}$  and  $(dN/dlogD_g)_{SWoptics}$  implies that assumptions are made on the values of n and k to correct OPCs data, and this may introduce a circularity in the estimates of the refractive index of dust that use  $(dN/dlogD_g)_{SWoptics}$  as input in optical calculations (see Sect. 3.2). In order to analyze the dependence of the results on this assumption, we made a sensitivity calculation by varying the values of n and k used for OPCs corrections within the range of values retrieved in this study (10% and 90% percentiles in Table 5, i.e., 1.49–1.54 for n and 0.001–0.006 for k). We concluded that changing n and k in this range has a very low impact on the retrieved number size distribution behind the SW inlets  $(dN/dlogD_g)_{SWoptics}$  compared to the original assumptions made in our calculations (<5% changes in the retrieved size number distribution at the different diameters between the original correction and the correction by varying n and k). This is due to the fact that when changing  $D_g$  due to changes in the n and k in the OPCs correction, the loss function also modifies to values corresponding to the new  $D_g$ . Given that the loss function increases/decreases for increasing/decreasing  $D_g$ , the combined changes in  $D_g$  and the loss function compensate so that the net number concentration behind the SW inlets varies less than a few percent. These results therefore suggest that the procedure to estimate the complex refractive index of dust is nearly independent of the assumed

#### 2.3 Dust elemental and mineralogical composition and iron content

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304 The elemental and mineralogical composition of the dust aerosols was estimated by combining different 305 techniques: X-ray diffraction (XRD, Panalytical model Empyrean diffractometer) to estimate the parti-306 cles' mineralogical composition in terms of clays, quartz, calcite, dolomite, gypsum, and feldspars; 307 wavelength dispersive X-ray fluorescence (WD-XRF, Panalytical PW-2404 spectrometer) to deter-308 mine the dust elemental composition (Na, Mg, Al, Si, P, K, Ca, Ti, Fe); and X-ray absorption near-edge 309 structure (XANES) to retrieve the content of iron oxides and their speciation between hematite and goethite. The dust mass collected on Nuclepore filters during the experiments varied between 0.3 and 310 311 6 mg m<sup>-3</sup> as calculated from elemental concentrations according to Lide (1992).

Full details on the XRD, WD–XRF, and XANES measurements and data analysis are provided in DB17 and C17. In this study, we discuss the dust elemental iron mass concentration, MC<sub>Fe%</sub>, i.e., the percent mass of elemental iron with respect to the total dust mass concentration, and the iron oxides mass concentration, MC<sub>Fe-ox%</sub>, i.e., the percent mass fraction of iron oxides with respect to the total dust mass concentration, estimated as the sum of goethite (MC<sub>Goet%</sub>) and hematite (MC<sub>Hem%</sub>) species.

#### 3. Strategy for data analysis

## 318 3.1 Calculation of the spectral extinction coefficient and SSA from scattering and absorption

#### 319 coefficients

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- The spectral scattering and absorption coefficients,  $\beta_{sca}$  ( $\lambda$ ) and  $\beta_{abs}$  ( $\lambda$ ), measured by the nephelometer and the aethalometer were used to estimate 10–min averages of the spectral extinction coefficient,  $\beta_{ext}$
- 322 ( $\lambda$ ), at the 7– $\lambda$  of the aethalometer between 370 and 950 nm as:

$$\beta_{\text{out}}(\lambda) = \beta_{\text{obs}}(\lambda) + \beta_{\text{cos}}(\lambda) \tag{5}.$$

- 324 The Extinction Ångström Exponent (EAE) was calculated as the power–law fit of  $\beta_{\text{ext}}$  versus  $\lambda$ .
- 325 The spectral single scattering albedo of dust at 10-min resolution (SSA<sub>10-min</sub>) was retrieved as:

$$SSA_{10-min}(\lambda) = \frac{\beta_{sca}(\lambda)}{\beta_{ext}(\lambda)}$$
 (6).

The experiment–averaged SSA ( $\lambda$ ) was calculated for each soil type based on the following formula (Moosmüller et al., 2012):

$$SSA(\lambda) = \left(1 + \frac{1}{m(\lambda)}\right)^{-1} \quad (7)$$

where m ( $\lambda$ ) represents the slope of the linear fit between the 10–min averages of  $\beta_{sca}$  ( $\lambda$ ) and  $\beta_{abs}$  ( $\lambda$ ) measured along the whole duration of each experiment. An example of  $\beta_{sca}$  ( $\lambda$ ) versus  $\beta_{abs}$  ( $\lambda$ ) fitting to retrieve the spectral SSA is shown in Fig. S3 in the Supplement. The correlation coefficient R<sup>2</sup> of the  $\beta_{sca}$  versus  $\beta_{abs}$  fit usually ranges between 0.97 and 1 at all wavelengths. As will be discussed later in the paper, the single scattering albedo of dust depends on the particle coarse size fraction, and during our experiments SSA<sub>10-min</sub> was not derived continuously for the different samples due to the aethalometer measurement interruptions. The application of Eq. (7) avoids any bias in the calculated averaged SSA for different soils due to size effects. For two of the analyzed samples (Tunisia and Namib–2),

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however, the linear fitting procedure was not applicable due to the fact that, respectively, only two and one absorption measurements from the aethalometer were available just after the peak of the injection, with no data afterwards. Average SSA data for Tunisia were thus estimated as the mean of the two available SSA<sub>10-min</sub> data points, while the single SSA<sub>10-min</sub> measurement at the peak of the injection was reported for Namib–2. This difference in time sampling should be kept in mind when comparing SSA data for these two samples to the rest of the dataset.

#### 3.2 Retrieval of the spectral complex refractive index

An optical calculation was performed to estimate the complex refractive index (m=n-ik) of dust aerosols based on optical and size data. The retrieval algorithm consisted in recalculating the spectral scattering  $\beta_{sca}$  ( $\lambda$ ) and absorption  $\beta_{abs}$  ( $\lambda$ ) coefficients measured at each 10-min interval by using the fitted (dN/dlogD)<sub>SWoptics</sub> size distribution as input and by varying the real and imaginary parts of the complex refractive index in the calculations until the best agreement between measurements and calculations was found. At each wavelength the root mean square deviation (RMSD) was calculated as:

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$$RMSD(\lambda) = \sqrt{\left[\frac{\beta_{sca,measured}(\lambda) - \beta_{sca,calculated}(\lambda)(n,k))}{\beta_{sca,calculated}(\lambda)(n,k))}\right]^{2} + \left[\frac{\beta_{abs,measured}(\lambda) - \beta_{abs,calculated}(\lambda)(n,k))}{\beta_{abs,calculated}(\lambda)(n,k))}\right]^{2}}$$
352 (8)

The RMSD was minimized at each wavelength to obtain n–k pairs that most closely reproduce the measured scattering and absorption coefficients. Optical calculations were performed at the 7 wavelengths of the aethalometer between 370 and 950 nm using Mie theory for homogeneous spherical particles. In the calculations, the real part of the refractive index was varied in the range 1.40–1.60 at steps of 0.01, while the imaginary part was varied in the range 0.0001–0.050 at steps of 0.0001. For each sample, this resulted in 10500 computations per wavelength and per 10–min time step. The uncertainty on the real and imaginary parts of the refractive index was estimated with a sensitivity study. For this purpose, the values of n and k were also obtained by using as input the observed  $\beta_{\text{sca}}(\lambda)$ ,  $\beta_{\text{abs}}(\lambda)$ , and (dN/dlogD)<sub>SWoptics</sub>, plus or minus one standard deviation on their measurement. The deviations of the values of n and k retrieved in the sensitivity study with respect to those obtained in the first inversions were assumed to correspond to the one standard deviation uncertainty of 10–min retrieved values.

Experiment–averaged values of the spectral n and k were estimated as the average of single n and k values retrieved at 10–min steps (indicated as  $n_{10-min}$  and  $k_{10-min}$ ). In fact, differently from the SSA, the refractive index did not seem to depend on the particle coarse size fraction (Sect. 4.5).

A control experiment was performed with submicron ammonium sulphate aerosols (see DB17 and supplementary Fig. S4) with the aim of validating the proposed methodology to estimate the aerosol complex refractive index for a non–absorbing aerosol type. For ammonium sulphate particles with a monomodal size distribution centered at  $0.06~\mu m$ , as measured with the SMPS, the retrieved real part of the refractive index was  $1.56~(\pm 0.01)$  in the 450-700~nm wavelength range, as expected from literature (Toon et al., 1976; Flores et al., 2009; Denjean et al., 2014).

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#### 3.3 Assumptions on the retrieval of SSA and complex refractive index

375 The approach used to retrieve the SSA and the complex refractive index of dust and the accuracy of

the results depend on the accuracy of the input data and the assumptions in the optical calculations.

377 We discuss here two points of the applied procedure, in part already mentioned in the previous para-

378 graphs.

1/ The size distribution from OPCs and also the scattering coefficient from the nephelometer used as input to the n and k retrieval procedure and SSA calculation depend more or less directly on the dust refractive index. These instruments need in fact to be corrected for instrumental artefacts and these corrections require an a priori knowledge of the n and k, which in our approach were set to fixed values (1.47–1.53 for n and 0.001–0.005 for k for OPCs optical to geometrical diameter conversion, and 1.53 for n and 0.001–0.003 for k for nephelometer truncation correction). This choice may in principle introduce a certain degree of uncertainty and circularity into the derived n, k, and SSA for dust. Nonetheless, we note that the range of refractive index values used to correct OPCs and nephelometer data falls in the range of variability of the refractive index values obtained in this study (see Sect. 4.3), which suggests that the values used for the corrections are appropriate. Additionally, as previously discussed, both the size distribution (dN/dlogDg)SWoptics and the scattering coefficient are not very sensitive to the assumptions about n and k used for the calculations (less than 5% changes in both the number size distribution behind SW inlets and the scattering coefficient from changing n and k within the range of estimated values in this study) which further demonstrates the robustness of the proposed approach.

2/ The retrieval procedure for n and k, as well as the calculations for OPCs optical—to—geometrical diameter and the nephelometer truncation correction, simplifies the non—spherical heterogeneous dust aerosols (e.g., Chou et al. 2008; Okada et al., 2011; Nousiainen and Kandler, 2015) into homogeneous spherical particles that can be represented by Mie theory. In the present study, we decided not to use a more advanced shape—representing theory, given that the shape distribution and morphology of the dust samples was not measured during experiments. Improper assumptions on the particle shape and morphology may in fact induce even larger errors than using Mie theory, in particular for super—micron aerosols (Kalashnikova and Sokolik, 2004; Nousiainen and Kandler, 2015). It should be pointed out, however, that dust is usually assumed to be spherical in global climate models (e.g., Myhre and Stordal, 2001; Balkanski et al., 2007; Jin et al., 2016), and different studies still show contrasting results on the true impact of dust non—sphericity on radiative fluxes and heating rates from global model simulations (Mishchenko et al., 1995; Yi et al., 2011; Räisänen et al., 2012; Colarco et al., 2014). On the other hand, shape effects can be important for the retrieval of aerosol properties from remote sensing techniques using spectral, angular, and polarized reflectance measurements (e.g., Feng et al., 2009).

## 4. Results

Nineteen soil samples from different desert areas in Northern Africa, Sahel, Eastern Africa and the Middle East, Central Asia, Eastern Asia, North America, South America, Southern Africa, and Australia were selected for experiments from a collection of 137 soil samples from source areas worldwide. The main information on the provenance of these soils is provided in Table 2. The nineteen selected soils, the same as analyzed in DB17, represent the major dust source regions depicted in Ginoux et al. (2012).

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Amongst the database of 137 samples from all the world regions that constitute significant dust emitters, this range in mineralogical composition represents the largest variability in iron oxides contents that can be found worldwide. This is illustrated in Fig. 2 where we represent the variability of hematite and goethite content in the nineteen selected soils and compare it with the range of variability of the global desert soils from the database of Journet et al. (2014).

### 4.1 Physical and chemical properties of analysed dust samples

#### 4.1.1 Dust mass concentration and size distribution

Figure 3 shows a typical example of a time series of aerosol mass concentration and effective fine and coarse diameters measured inside the CESAM chamber and behind the SW instruments inlets during the experiments, as well as the corresponding  $\beta_{sca}$  and  $\beta_{abs}$  at 370 nm. The Figure shows the rapid increase of the mass concentration within CESAM during dust injection in the chamber, and its subsequent decrease during experiments due to both size-selective gravitational settling, occurring mostly within the first 30 min of experiments, and dilution by sampling. The scattering and absorption coefficients of dust decrease with time after injection in tandem with the decrease of the mass concentration and the size-dependent depletion in the chamber. The dust mass concentration inside CESAM at the peak of the injection is between 2 mg m<sup>-3</sup> (Mali) and 310 mg m<sup>-3</sup> (Bodélé) and falls to values between 0.9 mg m<sup>-3</sup> (Mali) and 20 mg m<sup>-3</sup> (Bodélé) behind SW instruments inlets. These values are comparable to those measured close to sources during dust storms (Rajot et al., 2008; Kander et al., 2009). After a 2-hour experiment, the dust mass concentration has decreased to values of 0.2 to 2.5 mg m<sup>-3</sup> (inside CESAM) and of 0.1 to 1.9 mg m<sup>-3</sup> (behind the SW inlets), within a range of values comparable to what has been measured after medium- to long-range dust transport in the real atmosphere (Weinzerl et al., 2011; Denjean et al., 2016b). These data therefore indicate that in a 2-hour experiment in CESAM it is possible to reproduce the temporal changes of the mass load observed in the real atmosphere for dust from its emission to a receptor site. Concurrently with the mass concentration, the effective diameter, Deff,coarse, of the coarse fraction of the dust aerosol also rapidly decreases with time due the progressive deposition of the coarsest particles in the chamber. For the different analyzed soils, Deff,coarse varies in the range of 4-8 µm (peak of injection)

concurrently with the mass concentration, the effective diameter, D<sub>eff,coarse</sub>, of the coarse fraction of the dust aerosol also rapidly decreases with time due the progressive deposition of the coarsest particles in the chamber. For the different analyzed soils, D<sub>eff,coarse</sub> varies in the range of 4–8 μm (peak of injection) to 3–4 μm (after 2 hours) inside the CESAM chamber, and in the range of 3–4 μm (peak of injection) to 2–3 μm (after 2 hours) behind the SW inlets. In contrast, D<sub>eff,fine</sub> remains quite constant during the experiments, with a value between 0.6 and 0.7 μm for all soils. The values of D<sub>eff,coarse</sub> obtained in this study inside the CESAM chamber are in line with those measured close to African sources (4–12 μm, Rajot et al., 2008; Weinzerl et al., 2009; Ryder et al., 2013a) and for dust transported across the Mediterranean (5–8 μm, Denjean et al., 2016a). Conversely, the values of D<sub>eff,coarse</sub> behind the SW instruments inlets are mostly in agreement with those reported for dust transported across the Atlantic ocean (~3 μm, Maring et al., 2003; Müller et al., 2011; Denjean et al., 2016b). Our values of D<sub>eff,fine</sub> are higher compared to values reported by Denjean et al. (2016a) for dust aerosols transported over the Mediterranean (0.2 to 0.5 nm), reflecting the fact that we analyse pure dust whereas these authors often encountered dust externally mixed with pollution particles.

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The comparison of Deff,coarse values suggests that while the size of dust in CESAM is mostly representative of dust close to sources, as already pointed out in DB17, the size measured behind the SW instruments inlets is mostly representative of transport conditions. Figure 4 illustrates this point by showing the volume size distributions of the generated dust aerosols at the peak of injection seen by the SW optical instruments, compared to the average size of dust measured in CESAM (DB17) and field observations close to sources (e.g., Niger) and after long-range transport (Cape Verde, Suriname, and Puerto Rico). The size distribution of dust inside CESAM includes a coarse mode up to tenths of micrometres and well reproduces field observations close to sources, as shown in comparison to the Niger case. Due to particle losses along tubes, the coarsest particles above 10 µm diameter are not seen by the SW instruments. The overall shape of the dust size distribution sensed by the SW instruments is comparable to that measured during atmospheric long-range transport, even if the fraction of particles above 10 µm diameter is significantly under-represented compared to observations. (i.e., Betzer et al., 1988; Formenti et al., 2001; Maring et al., 2003; Ryder et al., 2013b; Jeong et al., 2014; Denjean et al., 2016b).

#### 4.1.2 Iron and iron oxide dust content

The mass concentrations of iron oxides, hematite, goethite, and total elemental iron for the different analysed samples are reported in Table 3. There is a considerable variability in the iron and iron oxide content for our samples. Total iron in the dust samples is in the range from 2.4% (Namib-1) to 10.6% (Australia). Iron oxides account for 11% and 62% of the iron mass, whereas the percent of iron oxides to the total dust mass varies between 0.7% (Bodélé Depression) and 5.8% (Niger). These data are in the range of values reported in the literature (Reid et al., 2003; Scheuvens et al., 2013; Formenti et al., 2011, 2014a). For the samples from the Sahara and the Sahel, goethite is the dominant iron oxide species, in agreement with Lafon et al. (2006) and Formenti et al. (2014a; 2014b). In contrast, over other regions, hematite dominates over goethite, as already reported by some studies (Arimoto et al., 2002; Shen et al., 2006; Lu et al., 2011).

# 4.2 Spectral- and time-dependent dust extinction and absorption coefficients, complex refractive index, and SSA

Figure 5 illustrates a typical spectral- and time-dependent set of measured optical properties. The spectral extinction coefficient, absorption coefficient, SSA, and real and imaginary parts of the complex refractive index obtained at 10-min resolution for the Morocco and Algeria samples are shown at the peak of the dust injection in CESAM and 30 and 90 min after the peak. Figure 5 shows that absorption decreases with wavelength, but not extinction. The SSA increases from 370 to 590 nm while it shows almost constant values between 590 and 950 nm. The imaginary refractive index decreases with λ following the decrease of  $\beta_{abs}$ . The real part of the refractive index is almost constant with wavelength. The extinction and absorption coefficients decrease in absolute value with time, as already shown in Fig. 3, but their spectral dependence remains quite constant with time, even if it varies from soil to soil. The experiment-averaged Absorption, Scattering, and Extinction Angström Exponents, representing

the spectral variation of the absorption, scattering and extinction coefficients, vary between the values of 1.5 and 2.4 (AAE), -0.4 and 0.4 (SAE), and -0.2 and +0.5 (EAE) for the different samples. These

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490 values are in line with those previously reported by Moosmüller et al. (2012) and C17 for dust from 491 various locations. The retrieved n and k also show negligible changes of their spectral shape with time 492 and their magnitude remains approximately constant. In contrast, the SSA increases with time, in par-493 ticular below 600 nm wavelength, and so its spectral shape changes. This is mostly due to the decrease 494 of the coarse size fraction with residence time in the chamber, as will be analysed in Sect. 4.5. Similarly 495 to the absorption, scattering, and extinction coefficients, the spectral shape of k and SSA is somewhat different between the various samples, with the sharpest spectral variations observed for the most ab-496 497 sorbing samples and a less pronounced spectral variation for the less absorbing ones, as evident, for 498 example, by comparing the SSA data for Morocco and Algeria in Fig. 5.

# 4.3 Spectral complex refractive index and SSA for the different source regions and comparison to literature data

Figure 6 shows the experiment-averaged n, k, and SSA between 370 and 950 nm for the nineteen aerosol samples analyzed in this study. Data of n, k, and SSA and their uncertainties for each sample are reported in Tables 4 and 5 together with the average values for each of the eight different source regions and for the full dataset. Figure 6 shows that there are significant differences, both in magnitude and spectral shape, between the imaginary refractive index and SSA for the different samples. The highest values of k (and lowest values of SSA) are obtained for the Niger, Mali, Namib-2 and Australia samples, which also show the highest values of both the iron oxide content between 3.6% and 5.8% and hematite content between 2.0% and 4.8% (k is in the range 0.0048-0.0088 at 370 nm and 0.0012-0.0021 at 950 nm, and SSA is in the range 0.70-0.75 at 370 nm and 0.95-0.97 at 950 nm). The lowest values are obtained for the Bodélé, Namib-1, and Arizona samples, which have iron oxide contents between 0.7% and 1.5% (k is 0.001 at 370 and 0.0003 at 950 nm, and SSA is in the range 0.91-0.96 at 370 nm and 0.97-0.99 at 950 nm). Both k and SSA vary from region to region, with the largest absorptions (highest k, lowest SSA) for the Sahel and Australia and the lowest absorption (lowest k, highest SSA) in North and South America and the Middle East; k and SSA values also vary within the same region, as illustrated in Fig. 6 for the Sahelian and Southern African samples. The real part of the refractive index, on the other hand, is not only almost wavelength-independent, as anticipated, but also relatively invariant from sample to sample. Its average over the 370-950 nm spectral range is between 1.48 (Gobi) and 1.55 (Ethiopia and Namib-2).

The full envelope of n, k, and SSA obtained for the entire set of analysed samples is shown in Fig. 7a and 7b (left panels). The real refractive index is relatively invariant, the spectral k varies by up to an order of magnitude (0.001–0.009 at 370 nm and 0.0003–0.002 at 950 nm). The SSA changes accordingly for the different dust samples at the different wavelengths (30% change at 370 nm corresponding to values between 0.70–0.96 and 4% change at 950 nm for values within 0.95–0.99). The population mean is 1.52 for n (as spectral average) and varies in the range 0.0033–0.0009 for k and 0.85–0.98 for the SSA between 370 and 950 nm (0.0016 and 0.94 as spectral averages for k and SSA) (Fig. 7 and

526 Tables 4 and 5).

The comparison between the full envelope of n, k, and SSA in this study with literature data is shown in Fig. 7a and 7b (right panels). Literature values considered for comparison include estimates from

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ground-based, aircraft, and satellite observations, laboratory studies, AERONET inversions, and estimates from mixing rules based on the dust mineralogical composition. Given that the sample selection in our experiments fully envelopes the global variability of mineralogy of natural dust, we could expect that our dataset would also fully envelope the global-scale variability of the dust absorption and scattering properties in the SW. When comparing with available literature data we found that our n and SSA datasets very well envelope the range of values indicated in the literature, with only a few outlier points. In contrast, for the imaginary refractive index the reported range of variability from the literature is significantly larger than that found in our study, with our range of k being mostly at the lower bound of previous results. Nonetheless, our range of k values fully envelopes the ensemble of remote sensing and field campaign data on airborne dust from the previous literature reported in Fig. 7a. The global average spectral values for k in our study (thick black line) perfectly match the Dubovik et al. (2002) dataset from a synthesis of AERONET observations from various locations worldwide. Likewise, our k average is also very close to the dataset by Balkanski et al. (2007), estimated from mineralogical composition assuming 1.5% in volume of hematite in dust (a value similar to our population average of 1.8% (in mass) for the dust hematite content), a value shown to allow a reconciliation of climate modelling and satellite observations of the dust direct SW radiative effect. Looking at Fig. 7a, the datasets which show the largest deviations from our estimated range of k are the ones by: (i) Volz (1972), Patterson et al. (1977) and Hess et al. (1998; i.e., the OPAC database, which is the same k dataset used in the new OPAC 4.0 version, Koepke et al., 2015), which are also amongst the most commonly used references for the dust imaginary refractive index in many climate models; (ii) the dataset by Wagner et al. (2012), from laboratory chamber experiments; and (iii) the dataset by Steigmann and Yang (2017), estimated from a machine learning technique. The reasons for these discrepancies in the k values are difficult to assess, since they can be related to both instrumental and analytical aspects. In the studies by Volz (1972) and Patterson et al. (1977), for instance, the complex refractive index was obtained by transmittance and diffuse reflectance on pellet samples, a technique that requires the dust to be pressed in a matrix of non-absorbing material. In this case a discrepancy arises from the different optical behaviour between dust compressed in a pellet and the airborne particles. Moreover, Volz (1972) and Patterson et al. (1977) analyse dust aerosols collected after mid- to long- transport, so after the dust have been possibly mixed with absorbing species. For the case of Wagner et al. (2012), the choice of the optical theory to retrieve k (T-matrix instead of Mie theory as used in our work, but also used in the majority of past field and remote sensing literature studies considered) is expected to cause the observed differences with our dataset.

## 4.4 Imaginary refractive index and SSA versus iron and iron oxide content

The sample–to–sample variability of the imaginary part of the refractive index k and the SSA observed in Fig. 6 and 7 is related to the dust composition by investigating the dependence on the particle iron content. In Fig. 8 we show the experiment–averaged k and SSA at 370, 520, and 950 nm versus the mass concentration of iron oxides (hematite+goethite, MC<sub>Fe-ox%</sub>), hematite (MC<sub>Hem%</sub>), goethite (MC<sub>Goeth%</sub>), and total elemental iron (MC<sub>Fe%</sub>) measured for the different dust samples analyzed in this study. The data are linearly fitted to relate k and SSA to MC<sub>Fe-ox%</sub>, MC<sub>Hem%</sub>, MC<sub>Goeth%</sub>, and MC<sub>Fe%</sub>. The results of the fits at all wavelengths between 370 and 950 nm are reported in Table 6, together with the

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570 the obtained chi square divided by the number of degrees of freedom). The data in Fig. 8 and Table 6 indicate that there is an excellent correlation between both k and SSA and MCFe-ox% at the different 571 572 wavelengths (R2>0.75). A weaker correlation is found when relating k and SSA to MCHem% and MCFe%

statistical indicators of the goodness of fit (correlation coefficient, R<sup>2</sup>, and reduced chi square,  $\chi^2_{red}$ , i.e.,

(R2 between 0.40 and 0.74 for k and between 0.49 and 0.78 for the SSA), and MC<sub>Goeth</sub> (R2 between 573

574 0.17 and 0.62). The better correlation of k and SSA to MCFe-ox% compared to MCFe% is expected since

575 dust optical properties in the visible wavelengths are mostly sensitive to the fraction of iron oxides,

576 rather than to iron incorporated into the crystal structure of silicates (i.e., Karickhoff and Bailey, 1973; 577 Lafon et al., 2006; Moosmüller et al., 2012; Klaver et al., 2011; Engelbrecht et al., 2016; C17). The

578 quantities that most robustly satisfy a linear relationship are k and MCFe-ox%, as indicated by the reduced

579 chi square  $\chi^2_{red}$  that is around 1 at all different wavelengths. The  $\chi^2_{red}$  increases to values also larger

than 2 in the other cases, indicating the poorer robustness of the fit in these cases. 580

581 We also investigated the dependence of the spectral k and SSA on the mass concentration of other 582 minerals contained in dust, such as clays, calcite, quartz, and feldspars, and also on the mass concen-

583 tration of different elements. We found that there is no statistically significant correlation between k and

584 SSA and the mass concentration of any of these compounds (not shown), with R2 values between 0.002

585 and 0.46 at the different wavelengths for all cases.

586 These results therefore clearly show that iron, particularly in the form of iron oxides, is the main driver

587 of dust shortwave absorption.

### 4.5 Imaginary refractive index and SSA versus dust coarse size fraction

589 The dependence of the spectral k and SSA on the dust coarse fraction is investigated by relating it to

590 the Deff,coarse calculated from the size distribution data behind the SW instruments inlets. The k<sub>10-min</sub> and

591 SSA<sub>10-min</sub> at 370, 520, and 950 nm versus Deff,coarse are shown in Fig. 9 for all experimental data, which

592 we separated into three classes based on their iron oxide content (MC<sub>Fe-ox</sub>% ≤ 1.5%, 1.5% < MC<sub>Fe-ox</sub>%

593 < 3%, MC<sub>Fe-ox%</sub> ≥ 3%). Figure 9 shows that even if the correlation is not very strong (R<sup>2</sup><0.54), there is

a clearly decreasing tendency for the  $SSA_{10-min}$  with increasing  $D_{eff,coarse}$ , particularly at 370 and 520 nm 595 for strongly absorbing samples with iron oxide content larger than 3%. The SSA<sub>10-min</sub> is mostly inde-

596 pendent of changes of Deff,coarse at 950 nm. Conversely, k<sub>10-min</sub> has a very poor correlation with Deff,coarse

(R2<0.35) and thus does not depend on size. Similar results were also obtained for the real part (not

598 shown).

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599 These results confirm previous observations (Sokolik and Toon, 1999; McConnell et al., 2008, 2010;

600 Kandler et al., 2011; Ryder et al., 2013a; 2013b) that the refractive index, an intrinsic property of matter,

601 is independent of size, while the SSA increases as the coarse dust size fraction decreases. This is due

602 to the fact that absorption efficiency for a single particle (Qabs) increases with particle diameter while the

603 scattering efficiency (Qsca) decreases. Ryder et al. (2013a) also showed that the dependence of SSA

604 on size is linear, but important only when the coarse fraction is high (if particles larger than about 3 µm

in diameter are present), otherwise the SSA depends mainly on composition. 605

#### 5. Summary and concluding remarks

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In this paper we presented new measurements of the spectral SW complex refractive index (m=n-ik)
and single scattering albedo (SSA) for nineteen mineral dust aerosols generated in the laboratory from
natural soil samples from major desert dust source areas in northern Africa, the Sahel, Middle East,
eastern Asia, North and South America, southern Africa, and Australia, and selected to represent the
heterogeneity of the dust composition at the global scale, in particular the range of iron oxide concentrations. The envelope of refractive indices and SSA data obtained in this study can thus be taken as
representative of the variability of the global dust aerosol.

Experiments described here were conducted in the 4.2 m³ CESAM chamber, a realistic and dynamic environment where dust aerosols can be generated and maintained in suspension for few hours while monitoring the evolution of their physical, chemical, and optical properties. The generated dust aerosols are characterized by a realistic size distribution, including both the sub–micron and the super–micron fraction, and they have an atmospherically representative mass concentration and composition, including iron oxides and elemental iron content.

The results of the present study can be summarized as follows:

- 1. The spectral k and SSA retrieved in this study vary from sample to sample within the same region but also from a region to another. For k, values vary between 0.001-0.009 at 370 nm and 0.0003-0.002 at 950 nm. For SSA, values vary from 0.70 to 0.96 at 370 nm and from 0.95 at 0.99 at 950 nm. In contrast, n is wavelength-independent and almost uniform for the different sources, with values between 1.48 and 1.55. Values for n and SSA fall within the range of published literature estimates, while for k we obtain a much narrower range of variability than the ensemble of literature results, as illustrated in Fig. 7. In particular, we found lower values of k compared to most of the literature values currently used in climate models, such as Volz et al. (1972), Patterson et al. (1977), and the OPAC database (Hess et al., 1998; Koepke et al., 2015). Miller et al. (2014) state that the values of Dubovik et al. (2002) from AERONET, Patterson et al. (1977) for far-travelled dust, and OPAC probably bracket the global solar absorption by dust. In contrast, our results indicate that dust absorption is lower than previously thought, and its average is close to the values reported by Dubovik et al. (2002) from AERONET observations and Balkanski et al. (2007) for a dust with a 1.5% volume fraction of hematite. Our range of variability of an order of magnitude for k and between 4% and 30% for the spectral SSA is actually large enough to change the sign of the global dust direct effect at the TOA (Miller et al., 2004), as well as its regional implications (e.g., Solmon et al., 2008; Jin et al., 2016), and has to be taken into account in climate modelling.
- The documented changes in k and SSA also impact remote sensing retrievals. To give an example,
   following Gasteiger et al. (2011), our observed variability of about 10% for the SSA at 532 nm would
   translate to about 40% variability in the retrieved extinction profiles and optical depths from lidar
   observations for dust from varying sources.
- 3. The sample–to–sample variability observed in this study is mostly related to the iron oxide and elemental iron content in dust. At each investigated wavelength the magnitude of k and SSA is linearly correlated to the mass concentration of total iron oxides, hematite, goethite, and total elemental iron. Small variations of these compounds translate into large variations of k and SSA. This

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suggests that it is sufficient to know the content of iron oxide or elemental iron in dust to predict its spectral k and SSA, which represents a huge simplification for parameterizing their regional and global variability.

4. We also investigated the dependence of k and SSA on the size distribution of dust. While k is independent of size (suggesting that a constant value can be used along transport), below 600 nm the SSA linearly decreases for increasing D<sub>eff,coarse</sub> for strongly absorbing samples with more than 3% iron oxide content. The investigated range of D<sub>eff,coarse</sub> is within about 2 and 4 μm, and thus comparable to values obtained along a transport path over the Atlantic Ocean for dust during about 2 to 6 days following emission (Denjean et al., 2016a).

Based on our results, we recommend that dust simulations, as well as remote sensing retrievals, use source-dependent values of the spectral SW refractive index and SSA instead of generic values. We propose, as a first step, a set of regionally-averaged n, k, and SSA values to represent dust from each of the eight regions analysed here as well as a global average value from the ensemble of our data (Tables 4 and 5). Furthermore, the relationships found between k and SSA and the iron oxides or elemental iron content in dust open the perspective to establish predictive rules to estimate the spectrallyresolved SW absorption of dust based on composition. We recommend the use of iron oxide content rather than iron content as it is better correlated with k and SSA. The relationship found in this study, nonetheless, refer to the bulk composition of the dust aerosols and to a size range typical of 2 to 6 days of transport in the atmosphere. As demonstrated in C17 with the mass extinction efficiency, the relationships linking the dust absorption to iron content vary as a function of the analysed size fraction due to the fact that iron bearing minerals are more concentrated in the clay fraction (<2.0 µm) than in the coarsest fraction of the dust (Kandler et al., 2009; C17). Further investigation should be therefore addressed to evaluate the dependence of the spectral k and SSA versus iron content as a function of the size distribution of the particles. This will allow to determine if the k and SSA versus iron relationships change or not in different phases of the aerosol lifetime, so from the source areas (when the coarsest fraction is dominant), to long-range transport conditions (when most of the coarse particle fraction above few µm has settled out). We point out, however, that the use of mineralogy to estimate k and SSA based on linear relationships, as obtained in our study, requires nonetheless that the model-predicted dust composition accurately reflects that of the natural atmospheric aerosols. To this aim, realistic soil mineralogy databases and accurate modelling of the soil to aerosol size fractionation need to be developed in model schemes.

Finally, this study had the objective to investigate the variability of the dust SW optical properties at the global scale in link to the global variability of the dust composition. It is noteworthy that observations over Southern Africa and the Sahel from the present study indicate that the k and SSA variabilities over these regions are comparable to the ones obtained for the global scale. For other regions, such as North America and Australia, only one sample was analyzed, with no information on the regional-scale variability of k and SSA. Additionally, for some of the analyzed areas, such as the Bodélé depression, even local scale variability (on the order of few km) may be of relevance, given the documented local scale changes of the particles' mineralogy and iron content (Bristow et al., 2010). More efforts should

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be therefore devoted to better characterize the variability of dust spectral optical properties at the regional scale with the aim of better assessing the dust impact on the climate of different areas of the world.

#### Data availability

Complex refractive index and single scattering albedo data for the different analyzed samples are provided in Tables 4 and 5. Processed CESAM data are immediately available upon request to the contact author. They will also soon be made available through the EUROCHAMP–2020 data center (https://data.eurochamp.org/). The following IDL routines were used in the analysis: mpfitexy.pro (available at <a href="https://github.com/williamsmj/mpfitexy">https://github.com/williamsmj/mpfitexy</a>) was used to linearly fit data taking into account uncertainties on both x and y; mie\_single.pro (available at <a href="http://www.atm.ox.ac.uk/code/mie/mie\_single.html">http://www.atm.ox.ac.uk/code/mie/mie\_single.html</a>) was used for optical calculations using Mie theory; mpcurvefit.pro (available at <a href="http://cow.physics.wisc.edu/~craigm/idl/idl.html">http://cow.physics.wisc.edu/~craigm/idl/idl.html</a>) was used for size lognormal fitting.

#### **Author contributions**

C. Di Biagio, P. Formenti, Y. Balkanski, and J. F. Doussin designed the experiments and discussed the results. C. Di Biagio performed the experiments and performed the full data analysis with contributions by P. Formenti, L. Caponi, M. Cazaunau, E. Pangui, and J.F. Doussin. The soil samples used for experiments were collected by M. O. Andreae, K. Kandler, T. Saeed, S. Piketh, D. Seibert, and E. Williams. E. Journet participated to the selection of the soil samples for experiments. Sophie Nowak performed the XRD measurements. C. Di Biagio and P. Formenti wrote the manuscript with comments from all co–authors.

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1087 **Table 1.** Measured and retrieved quantities and their estimated uncertainties. For further details, refer1088 to Sect. 2, as well DB17 and C17.

.089 Pa	rameter	Time resolution	Uncertainty	Uncertainty calcula- tion	Comments
	Scattering coefficient at 450, 550, and 700 nm, $\beta_{sca}$	10-min data	5–12%	Quadratic combination of photon counting and gas calibration uncertainty (5%), angular corrections uncertainty (<5%) and standard deviation over 10–min intervals (2-10%).	The uncertainty on $\beta_{sca}$ ( $\lambda$ ) usually decreases with increasing dust residence time in the chamber as a result of the reduction of the coarse component.
	Absorption coefficient at 370, 470, 520, 590, 660, 880, and 950 nm, β <sub>abs</sub> (λ)	10-min data	22–30% at 370 nm 23–87% at 950 nm	Error propagation formula¹ on Eq. (2) considering the uncertainties on $\beta_{ATT}(\lambda)$ from 10-min fitting procedure (error propagation formula¹ on Eq. 1, ~20%), and uncertainties on $\alpha(\lambda)$ (1%), $\beta_{sca}(\lambda)$ (5-12%), $C_{ref}$ (10%), and R (1-10%).	
	Extinction coeffi- cient, $\beta_{\text{ext}}(\lambda) = \beta_{\text{sca}}(\lambda) + \beta_{\text{abs}}(\lambda)$	10–min data	~25%	Sum of $\beta_{sca}$ ( $\lambda$ ) and $\beta_{abs}$ ( $\lambda$ ) uncertainties	
	Single Scattering Albedo, SSA ( $\lambda$ ) = $\beta_{sca}$ ( $\lambda$ ) / ( $\beta_{sca}$ ( $\lambda$ ) + $\beta_{abs}$ ( $\lambda$ ))	10–min data	9–12%	Error propagation formula $^1$ considering single uncertainties on $\beta_{\text{sca}}$ and $\beta_{\text{abs}}$ .	
Optical SW	Single Scattering Albedo, SSA $(\lambda)$ = $(1+1/m(\lambda))^{-1}$	Experiment averaged	1–12% at 370 nm 1–3% at 950 nm	Error propagation formula on Eq. (6) considering the uncertainty on $m(\lambda)$ , i.e., the slope of the linear fit between $\beta_{\text{sca}}$ and $\beta_{\text{abs}}$ over the whole duration of each experiment.	
	Complex refrac- tive index (n-ik)	10-min data	<5% for n <50% for k	Deviations of the values of n and k retrieved in the sensitivity study (see Sect. 3.2) with respect to those obtained in the first inversions were assumed to correspond to the one standard deviation uncertainty to 10—min retrieved values.	
	Complex refractive index (n-ik)	Experiment averaged	<8% for n 13–75 % for k	Quadratic combination of the standard deviation of n and k over the experiment and the deviation on the experiment—averaged values between those obtained from central inversions and inversions using input data ± their uncertainty.	
Size distribution	SMPS geometrical diameter $(D_g), \\ D_g = D_m  / \chi$	-	~6%	Error propagation formula' considering the uncertainty on the estimated shape factor χ (~6%)	The electrical mobility to geometrical diameter conversion was performed by assuming for dust a dynamic shape factor of 1.75 ± 0.10, as determined by SMPS—SkyGrimm comparison

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	1		T		1. d. d
					in their overlapping
	SkyGrimm geo- metrical diameter (D <sub>g</sub> )	-	<15.2%	Standard deviation of the D <sub>g</sub> values obtained for different refractive indices values used in the optical to geomet- rical conversion	range (see DB17) The conversion of optical to geometrical diameters for the SkyGrimm and the WELAS was performed by Mie calculations by varying the
	WELAS geomet- rical diameter (D <sub>9</sub> )	-	<7%	The same as for the SkyGrimm	dust refractive index in the range 1.47–1.53 for the real part and 0.001–0.005 for the imaginary part. Then $D_g$ is set at the mean $\pm$ 1 standard deviation of the values obtained for the different values of n and k (see DB17). Refractive index is assumed to be constant with particle size and wavelength—independent.
	(dN/dlogD) <sub>SWoptics</sub>	10-min data	~20-90%	Error propagation formula¹ considering the dN/dlogD <sub>g</sub> st. dev. over 10-min and the uncertainty on particle loss function along sampling tubes L(D <sub>g</sub> ) (~50% at 2 µm, ~10% at 8 µm)	The uncertainty of L(D <sub>g</sub> ) was estimated with a sensitivity study by varying the values of the input parameters to the Particle Loss Calculator software within their uncertainties (see DB17)
	D <sub>eff,fine</sub>	10-min data	<5%	Deviation obtained by	,
	D <sub>eff,coarse</sub>	10-min data	5–40%	repeating the calculations by using the size distribution ± its uncertainty.	
	Elemental iron mass concentra- tion (MC <sub>Fe%</sub> )	Experiment averaged	10%		
Mineralogi- cal composi-	Iron oxides mass concentration (MC <sub>Fe-ox%</sub> )	Experiment averaged	15%	Uncertainties calculated as discussed in DB17	
tion	Goethite mass concentration (MC <sub>Goet%</sub> )	Experiment averaged	<10%	and C17	
	Hematite mass concentration (MC <sub>Hem%</sub> )	Experiment averaged	<10%		
		2			

<sup>1</sup>  $\sigma_f = \sqrt{\sum_{i=1}^n \left(\frac{\partial f}{\partial x_i} \sigma_{x_i}\right)^2}$ 

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Table 2. Summary of information on the soil samples and sediments used in this study.

Geographical area	Sample	Coordinates	Desert area
	Tunisia	33.02°N, 10.67°E	Maouna
	Morocco	31.97°N, 3.28°W	east of Ksar Sahli
Northern Africa – Sahara	Libya	27.01°N, 14.50°E	Sebha
	Algeria	23.95°N, 5.47°E	Ti-n-Tekraouit
	Mauritania	20.16°N, 12.33°W	east of Aouinet Nchir
	Niger	13.52°N, 2.63°E	Banizoumbou
Sahel	Mali	17.62°N, 4.29°W	Dar el Beida
	Bodélé	17.23°N, 19.03°E	Bodélé depression
Eastern Africa and the	Ethiopia	7.50°N, 38.65°E	Lake Shala National Park
Middle East	Saudi Arabia	27.49°N, 41.98°E	Nefud
Middle East	Kuwait	29.42°N, 47.69°E	Kuwaiti
Eastern Asia	Gobi	39.43°N, 105.67°E	Gobi
Eastern Asia	Taklimakan	41.83°N, 85.88°E	Taklimakan
North America	Arizona	33.15 °N, 112.08°W	Sonoran
South America	Atacama	23.72°S, 70.40°W	Atacama
South America	Patagonia	50.26°S, 71.50°W	Patagonia
Southern Africa	Namib-1	21.24°S, 14.99°E	Namib
Southern Affica	Namib-2	19.00°S, 13.00°E	Namib
Australia	Australia	31.33°S, 140.33°E	Strzelecki

**Table 3.** Chemical characterization of the dust aerosols in the PM<sub>10.6</sub> size fraction. Column 3 shows MC<sub>Fe%</sub>, the fractional mass of elemental iron with respect to the total dust mass concentration (±10 %), and column 4 reports MC<sub>Fe-ox%</sub>, the mass fraction of iron oxides with respect to the total dust mass concentration (±15 %) and its speciation in hematite MC<sub>Hem%</sub> and goethite MC<sub>Goeth%</sub> (<±10%). The iron oxide measurements were not made on the Taklimakan sample. Mean values and standard deviations based on single sample data are reported for the full dataset.

Geographical area	Sample	MC <sub>Fe%</sub>	MC <sub>Fe-ox%</sub>	MC <sub>Hem%</sub>	MC <sub>Goet%</sub>
	Tunisia	4.1	2.2	1.2	1.1
	Morocco	3.6	1.4	0.4	1.0
Northern Africa – Sahara	Libya	5.2	3.1	0.9	2.2
normon, mod Canara	Algeria	6.6	2.7	1.4	1.4
	Mauritania	8.1	3.3	3.3	0.0
	Niger	6.1	5.8	2.3	3.5
Sahel	Mali	6.6	3.7	2.0	1.7
	Bodélé	4.1	0.7	0.7	0.0
Factors Africa and the Middle	Ethiopia	6.8	2.0	2.0	0.0
Eastern Africa and the Middle East	Saudi Arabia	3.8	2.6	1.8	0.8
Lasi	Kuwait	5.0	1.5	1.5	0.0
Eastern Asia	Gobi	4.8	0.9	0.9	0.0
Lasterii Asia	Taklimakan	5.8	_	ı	_
North America	Arizona	5.3	1.5	1.5	0.0
South America	Atacama	4.7	1.6	1.6	0.0
South America	Patagonia	5.1	1.5	0.9	0.6
Southern Africa	Namib-1	2.4	1.1	0.8	0.3
Southern Amica	Namib-2	10.6	4.8	4.8	0.0
Australia	Australia	7.2	3.6	3.6	0.0
Full dataset mean (st. dev.)		5.6 (1.9)	2.4 (1.4)	1.8 (1.1)	0.7 (1.0)

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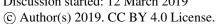




**Table 4.** Real (n) and imaginary (k) parts of the refractive index estimated for the nineteen analysed dust samples and mean values calculated for the eight regions and for the full dataset. Data for single soils are reported as experiment—averaged values and their uncertainty is calculated as indicated in Table 1. Mean values and standard deviations at each wavelength based on single sample data are reported for the eight regions and the full dataset. The median and 10% and 90% percentile values are also reported for the full dataset. For North America and Australia, for which only one dust sample was analysed, the reported data correspond to the single sample available from these regions. For the real part, the average over the whole shortwave range (nsw) is indicated.

Sample/Re- gion	nsw	$\sigma_{nsw}$				k							$\sigma_{k}$			
	037–0.95 μm	037– 0.95 μm	0.37 μm	0.47 μm	0.52 μm	0.59 μm	0.66 μm	0.88 μm	0.95 μm	0.37 μm	0.47 μm	0.52 μm	0.59 µm	0.66 μm	0.88 µm	0.95 μm
Tunisia	1.51	0.06	0.0045	0.0035	0.0026	0.0018	0.0015	0.0013	0.0012	0.0030	0.0026	0.0018	0.0012	0.0010	0.0008	0.0007
Morocco	1.49	0.03	0.0023	0.0016	0.0012	0.0008	0.0007	0.0006	0.0007	0.0006	0.0004	0.0003	0.0002	0.0002	0.0002	0.0002
Lybia	1.5	0.04	0.0029	0.0019	0.0014	0.0007	0.0006	0.0007	0.0007	0.0006	0.0004	0.0002	0.0001	0.0002	0.0002	0.0002
Algeria	1.52	0.04	0.0025	0.0016	0.0012	0.0007	0.0005	0.0006	0.0006	0.0010	0.0006	0.0004	0.0003	0.0003	0.0003	0.0003
Mauritania	1.5	0.03	0.0043	0.0033	0.0026	0.0014	0.0013	0.0010	0.0010	0.0010	0.0009	0.0008	0.0003	0.0003	0.0004	0.0003
Northern Af- rica – Sa- hara (mean and st. dev.)	1.51	0.03	0.0033	0.0024	0.0018	0.0011	0.0009	0.0008	0.0008	0.0010	0.0010	0.0007	0.0005	0.0004	0.0003	0.0003
Niger	1.51	0.04	0.0088	0.0061	0.0048	0.0034	0.0031	0.0028	0.0021	0.0043	0.0031	0.0023	0.0018	0.0015	0.0010	0.0013
Mali	1.52	0.05	0.0048	0.0038	0.0030	0.0023	0.0024	0.0021	0.0021	0.0008	0.0006	0.0004	0.0003	0.0003	0.0003	0.0003
Bodélé	1.49	0.03	0.0011	0.0007	0.0006	0.0004	0.0004	0.0003	0.0003	0.0006	0.0004	0.0003	0.0002	0.0002	0.0001	0.0001
Sahel (mean and st. dev.)	1.51	0.03	0.0049	0.0035	0.0028	0.0020	0.0020	0.0017	0.0015	0.0038	0.0027	0.0021	0.0015	0.0014	0.0013	0.0011
Ethiopia	1.55	0.06	0.0026	0.0020	0.0016	0.0013	0.0011	0.0007	0.0006	0.0009	0.0008	0.0007	0.0005	0.0004	0.0002	0.0002
Saudi Arabia	1.54	0.06	0.0028	0.0021	0.0015	0.0007	0.0006	0.0006	0.0006	0.0006	0.0005	0.0004	0.0002	0.0001	0.0001	0.0001
Kuwait	1.50	0.04	0.0016	0.0010	0.0008	0.0006	0.0005	0.0005	0.0004	0.0005	0.0003	0.0003	0.0002	0.0002	0.0003	0.0002
Eastern Af- rica and the Middle East (mean and st. dev.)	1.53	0.05	0.0023	0.0017	0.0013	0.0009	0.0007	0.0006	0.0005	0.0007	0.0006	0.0005	0.0004	0.0003	0.0001	0.0001
Gobi	1.48	0.05	0.0041	0.0025	0.0018	0.0012	0.0011	0.0012	0.0012	0.0017	0.0009	0.0006	0.0004	0.0004	0.0005	0.0005
Taklimakan	1.54	0.07	0.0018	0.0012	0.0009	0.0006	0.0005	0.0005	0.0005	0.0008	0.0005	0.0004	0.0002	0.0002	0.0002	0.0002
Eastern Asia (mean and st. dev.)	1.51	0.05	0.0030	0.0019	0.0014	0.0009	0.0008	0.0008	0.0009	0.0016	0.0009	0.0006	0.0005	0.0005	0.0005	0.0005
Arizona	1.51	0.05	0.0011	0.0009	0.0007	0.0005	0.0005	0.0005	0.0004	0.0005	0.0004	0.0003	0.0002	0.0002	0.0002	0.0002
North Amer- ica (mean and st. dev.)	1.51	0.05	0.0011	0.0009	0.0007	0.0005	0.0005	0.0005	0.0004	0.0005	0.0004	0.0003	0.0002	0.0002	0.0002	0.0002
Atacama	1.54	0.07	0.0016	0.0015	0.0012	0.0008	0.0006	0.0006	0.0006	0.0005	0.0004	0.0003	0.0002	0.0002	0.0002	0.0002
Patagonia	1.53	0.07	0.0024	0.0016	0.0011	0.0009	0.0006	0.0007	0.0006	0.0008	0.0005	0.0003	0.0003	0.0003	0.0003	0.0002
South Amer- ica (mean and st. dev.)	1.54	0.06	0.0020	0.0015	0.0011	0.0008	0.0006	0.0007	0.0006	0.0006	0.0001	0.0001	0.0001	0.0000	0.0001	0.0000
Namib-1	1.53	0.06	0.0012	0.0009	0.0006	0.0004	0.0003	0.0004	0.0004	0.0006	0.0004	0.0003	0.0002	0.0001	0.0002	0.0001
Namib-2	1.55	0.07	0.0072	0.0054	0.0044	0.0025	0.0018	0.0014	0.0014	0.0027	0.0019	0.0016	0.0009	0.0007	0.0006	0.0006
Southern Af- rica (mean and st. dev.)	1.54	0.06	0.0042	0.0031	0.0025	0.0014	0.0011	0.0009	0.0009	0.0042	0.0032	0.0027	0.0015	0.0010	0.0007	0.0007
Australia	1.54	0.06	0.0058	0.0042	0.0033	0.0017	0.0013	0.0013	0.0012	0.0022	0.0011	0.0010	0.0006	0.0006	0.0004	0.0003
Australia (mean and st. dev.)	1.54	0.06	0.0058	0.0042	0.0033	0.0017	0.0013	0.0013	0.0012	0.0022	0.0011	0.0010	0.0006	0.0006	0.0004	0.0003
Full dataset (mean and st. dev.)	1.52	0.04	0.0033	0.0024	0.0018	0.0012	0.0010	0.0009	0.0009	0.0021	0.0016	0.0013	0.0008	0.0007	0.0006	0.0005
Full dataset median Full dataset	1.52		0.0026	0.0019	0.0014	0.0008	0.0006	0.0007	0.0006							
10% percen- tile	1.49		0.0012	0.0009	0.0007	0.0005	0.0004	0.0004	0.0004							
Full dataset 90% percen- tile	1.54		0.0061	0.0044	0.0035	0.0023	0.0019	0.0015	0.0015							

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1129 **Table 5.** As in Table 4 for the single scattering albedo (SSA) data.

Sam-				SSA							$\sigma_{SSA}$			
ple/Region	0.37	0.47	0.52	0.59	0.66	0.88	0.95	0.37	0.47	0.52	0.59	0.66	0.88	0.95
Tuninin	μm	μm	μm	μm	μm	μm 0.97	μm	μm	0.47 μm	μm	μm	μm	μm	μm
Tunisia Morocco	0.85 0.92	0.90 0.95	0.93 0.96	0.95 0.98	0.95 0.98	0.97	0.97 0.99	0.03	0.02	0.02	0.01	0.01	0.01	0.01
	0.92	0.93	0.95	0.98	0.98		0.99	0.01	0.01		0.00	0.00	0.00	0.00
Lybia Algeria	0.89	0.93	0.95	0.98	0.98	0.98	0.98	0.02	0.01	0.01	0.00	0.00	0.00	0.00
Mauritania	0.85	0.92	0.94	0.96	0.97	0.98	0.98	0.02	0.01	0.01	0.00	0.00	0.00	0.00
Northern Africa – Sahara (mean and	0.88	0.90	0.94	0.97	0.97	0.98	0.98	0.02	0.02	0.02	0.01	0.01	0.01	0.01
st. dev.) Niger	0.72	0.85	0.89	0.91	0.92	0.94	0.95	0.09	0.09	0.07	0.05	0.05	0.03	0.02
Mali	0.72	0.85	0.89	0.91	0.92	0.94	0.95	0.09	0.09	0.07	0.05	0.05	0.03	0.02
Bodélé	0.75	0.98	0.03	0.99	0.99	0.99	0.99	0.04	0.03	0.02	0.02	0.02	0.01	0.01
Sahel (mean and	0.81	0.89	0.92	0.99	0.95	0.96	0.99	0.04	0.02	0.02	0.04	0.04	0.03	0.01
st. dev.) Ethiopia	0.80	0.86	0.90	0.92	0.94	0.97	0.97	0.03	0.03	0.02	0.02	0.01	0.01	0.01
Saudi Ara- bia	0.88	0.93	0.96	0.98	0.98	0.98	0.98	0.03	0.02	0.01	0.01	0.01	0.00	0.00
Kuwait	0.95	0.97	0.98	0.98	0.99	0.99	0.99	0.02	0.01	0.01	0.01	0.01	0.01	0.00
Eastern Africa and the Middle East (mean and st. dev.)	0.88	0.92	0.94	0.96	0.97	0.98	0.98	0.07	0.05	0.04	0.03	0.03	0.01	0.01
Gobi	0.88	0.92	0.94	0.96	0.97	0.97	0.97	0.04	0.03	0.02	0.01	0.01	0.01	0.01
Taklimakan	0.82	0.88	0.92	0.95	0.96	0.96	0.96	0.03	0.02	0.02	0.01	0.01	0.01	0.01
Eastern Asia (mean and st. dev.)	0.85	0.90	0.93	0.96	0.96	0.97	0.97	0.04	0.03	0.02	0.01	0.01	0.01	0.01
Arizona	0.93	0.96	0.97	0.98	0.98	0.99	0.99	0.01	0.01	0.01	0.00	0.00	0.00	0.00
North America (mean and st. dev.)	0.93	0.96	0.97	0.98	0.98	0.99	0.99	0.01	0.01	0.01	0.00	0.00	0.00	0.00
Atacama	0.89	0.93	0.94	0.97	0.97	0.98	0.98	0.03	0.02	0.02	0.01	0.01	0.01	0.01
South America (mean and st. dev.)	0.88	0.91 <b>0.92</b>	0.94	0.96	0.97 <b>0.97</b>	0.98	0.98	0.02	0.02	0.01	0.01	0.01	0.00	0.01
Namib-1	0.91	0.95	0.96	0.98	0.98	0.99	0.99	0.02	0.01	0.01	0.00	0.00	0.00	0.00
Namib-2	0.74	0.82	0.86	0.92	0.94	0.96	0.97	0.03	0.02	0.02	0.01	0.01	0.01	0.01
Southern Africa (mean and st. dev.)	0.83	0.88	0.91	0.95	0.96	0.98	0.98	0.12	0.09	0.07	0.04	0.03	0.02	0.02
Australia	0.70	0.81	0.85	0.91	0.93	0.96	0.97	0.04	0.03	0.02	0.01	0.01	0.01	0.01
Australia (mean and st. dev.)	0.70	0.81	0.85	0.91	0.93	0.96	0.97	0.04	0.03	0.02	0.01	0.01	0.01	0.01
Full da- taset (mean and st. dev.)	0.85	0.91	0.93	0.96	0.96	0.97	0.98	0.08	0.05	0.04	0.03	0.02	0.01	0.01
Full da- taset me- dian	0.88	0.92	0.94	0.96	0.97	0.98	0.98							

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Full da- taset 10% percentile	0.74	0.84	0.88	0.92	0.94	0.96	0.96				
Full da- taset 90% percentile	0.93	0.96	0.97	0.98	0.99	0.99	0.99				

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**Table 6.** Results of the linear fit between k and SSA and the mass concentration of iron oxides,  $MC_{Fe-ox\%}$ , hematite,  $MC_{Hem\%}$ , goethite,  $MC_{Goeth\%}$ , and elemental iron,  $MC_{Fe\%}$  in dust. Column 1 indicates the wavelength; (a  $\pm$   $\sigma$ a) indicates the retrieved slope and its estimated uncertainty; (b  $\pm$   $\sigma$ b) indicates the retrieved intercept and its estimated uncertainty;  $R^2$  denotes the correlation coefficient and  $\chi^2_{red}$  is the reduced chi–square of the fit.

	k =	a MC <sub>Fe-ox %</sub> + b		$SSA = a MC_{Fe-ox \%} + b$					
Wavelength (nm)	a±σa	b±σb	$R^2$ ; $\chi^2_{red}$	a±σa	b±σb	$R^2$ ; $\chi^2_{red}$			
370	(11.9 ± 2.4) 10 <sup>-4</sup>	$(2.4 \pm 4.6) \ 10^{-4}$	0.88 ; 0.6	(-5.8 ± 0.8) 10 <sup>-2</sup>	(1.00 ± 0.02)	0.83 ; 1.7			
470	(9.0 ± 1.7) 10 <sup>-4</sup>	$(1.7 \pm 3.2) 10^{-1}$		$(-3.8 \pm 0.6) 10^{-2}$	$(1.00 \pm 0.01)$				
		4	0.89 ; 0.8			0.78 ; 1.8			
520	$(6.8 \pm 1.3) \ 10^{-4}$	$(1.3 \pm 2.4) 10^{-4}$	0.90 ; 0.9	$(-2.9 \pm 0.4) 10^{-2}$	$(1.01 \pm 0.01)$	0.76 ; 2.0			
590	$(4.5 \pm 0.9) \ 10^{-4}$	$(0.9 \pm 1.6) \ 10^{-4}$	0.85 ; 1.4	$(-1.8 \pm 0.3) 10^{-2}$	$(1.00 \pm 0.01)$	0.75 ; 2.3			
660	$(4.3 \pm 0.8) \ 10^{-4}$	$(0.8 \pm 1.4) \ 10^{-4}$	0.81 ; 1.6	$(-1.3 \pm 0.2) 10^{-2}$	$(1.00 \pm 0.00)$	0.75 ; 2.2			
880	$(3.4 \pm 0.6) \ 10^{-4}$	(0.6 ± 1.2) 10 <sup>-4</sup>		$(-0.76 \pm 0.16)  10^{-1}$	$(1.00 \pm 0.00)$				
			0.79 ; 1.0	2		0.79 ; 1.4			
950	(3.2 ± 0.6) 10 <sup>-4</sup>	$(0.6 \pm 1.0) 10^{-4}$		$(-0.62 \pm 0.13) 10^{-1}$	$(0.99 \pm 0.00)$				
		L	0.77 ; 1.1			0.78 ; 1.1			
	K:	= <i>a</i> MC <sub>Hem %</sub> + b		SSA	$= a MC_{Hem \%} + b$				
Wavelength (nm)	a±σa	b ± σb	R <sup>2</sup> ; χ <sup>2</sup> <sub>red</sub>	a±σa	b± σb	$R^2$ ; $\chi^2_{red}$			
370	(9.7 ± 2.7) 10 <sup>-4</sup>	(2.7 ± 4.0) 10 <sup>-4</sup>	0.67 ; 1.9	(-4.4 ± 0.6) 10 <sup>-2</sup>	$(0.95 \pm 0.01)$	0.73 ; 3.5			
470	(8.3 ± 1.9) 10 <sup>-4</sup>	$(1.9 \pm 2.7) \ 10^{-4}$	0.72 ; 1.9	$(-3.0 \pm 0.4) 10^{-2}$	$(0.97 \pm 0.01)$	0.76; 3.2			
520	(6.9 ± 1.5) 10 <sup>-4</sup>	$(1.5 \pm 2.0) \ 10^{-4}$	0.74 ; 2.0	(-2.2 ± 0.3) 10 <sup>-2</sup>	$(0.98 \pm 0.00)$	0.78; 3.3			
590	$(3.7 \pm 0.8) \ 10^{-4}$	(0.9 ± 1.2) 10 <sup>-4</sup>	0.61 ; 2.1	(-1.3 ± 0.2) 10 <sup>-2</sup>	$(0.99 \pm 0.00)$	0.71 ; 2.7			
660	$(3.7 \pm 0.8) 10^{-4}$	$(0.8 \pm 1.1) 10^{-4}$	0.51 ; 2.6	$(-0.9 \pm 0.2) 10^{-2}$	$(0.99 \pm 0.00)$	0.62 ; 2.5			
880	$(2.9 \pm 0.7) \cdot 10^{-4}$	$(0.7 \pm 1.1) 10^{-4}$	0.43 ; 2.1	$(-0.6 \pm 0.1) 10^{-2}$	$(0.99 \pm 0.00)$	0.57 ; 1.8			
950	$(2.6 \pm 0.6) 10^{-4}$	$(0.6 \pm 0.9) \ 10^{-4}$	0.46 ; 2.1	$(-0.5 \pm 0.1) 10^{-2}$	$(0.99 \pm 0.00)$	0.49; 1.7			
	k =	a MC <sub>Goeth %</sub> + b		SSA	= a MC <sub>Goeth %</sub> + b				
Wavelength (nm)	a±σa	b ± σb	$R^2$ ; $\chi^2_{red}$	a±σa	b±σb	$R^2$ ; $\chi^2_{red}$			
370	(9.0 ± 2.5) 10 <sup>-4</sup>	(2.5 ± 2.2) 10 <sup>-4</sup>	0.47 ; 1.8	(-13.4 ± 6.9) 10 <sup>-3</sup>	$(0.90 \pm 0.01)$	0.32 ; 6.8			
470	(5.5 ± 1.7) 10 <sup>-4</sup>	(1.7 ± 1.5) 10 <sup>-4</sup>	0.43 ; 2.3	$(-8.3 \pm 4.7) \ 10^{-3}$	$(0.94 \pm 0.00)$	0.21 ; 6.2			
520	(3.4 ± 1.1) 10 <sup>-4</sup>	(1.1 ± 1.2) 10 <sup>-4</sup>	0.41 ; 2.5	(-4.9 ± 3.2) 10 <sup>-3</sup>	$(0.96 \pm 0.00)$	0.17 ; 6.4			
590	$(0.5 \pm 0.6) \ 10^{-4}$	$(0.6 \pm 0.8) 10^{-4}$	0.50 ; 3.2	$(0.9 \pm 2.0) 10^{-3}$	$(0.97 \pm 0.00)$	0.23 ; 5.5			
660	$(2.2 \pm 0.8) 10^{-4}$	$(0.8 \pm 0.7) \ 10^{-4}$	0.55 ; 3.6	$(0.2 \pm 1.6) \ 10^{-3}$	$(0.98 \pm 0.00)$	0.34 ; 4.4			
880	$(2.6 \pm 0.8) \ 10^{-4}$	$(0.8 \pm 0.6) \ 10^{-4}$	0.62 ; 2.4	(-1.1 ± 1.4) 10 <sup>-3</sup>	$(0.98 \pm 0.00)$	0.47; 3.0			
950	$(2.6 \pm 0.8) 10^{-4}$	$(0.8 \pm 0.6) \ 10^{-4}$	0.55 ; 2.5	(-2.1 ± 1.4) 10 <sup>-3</sup>	$(0.98 \pm 0.00)$	0.54 ; 2.6			
	k	= <i>a</i> MC <sub>Fe %</sub> + b		SSA = a MC <sub>Fe %</sub> + b					
Wavelength (nm)	a±σa	b± σb	$R^2$ ; $\chi^2_{red}$	a±σa	b± σb	$R^2$ ; $\chi^2_{red}$			
370	(6.0 ± 1.4) 10 <sup>-4</sup>	(1.4 ± 0.7) 10 <sup>-4</sup>	0.60 ; 1.5	(-2.7 ± 0.4) 10 <sup>-2</sup>	(1.02 ± 0.02)	0.67 ; 3.1			
470	$(4.7 \pm 1.0) 10^{-4}$	$(1.0 \pm 0.5) 10^{-4}$	0.62 ; 1.7	$(-1.8 \pm 0.3) \ 10^{-2}$	(1.02 ± 0.01)	0.72 ; 2.8			
520	$(3.9 \pm 0.8) \cdot 10^{-4}$	$(0.8 \pm 3.9) \ 10^{-4}$	0.65 ; 1.6	$(-1.3 \pm 0.2) \cdot 10^{-2}$	(1.01 ± 0.01)	0.72; 2.9			
590	$(2.5 \pm 0.5) \cdot 10^{-4}$	$(0.5 \pm 2.4) 10^{-4}$	0.56 ; 1.7	$(-0.8 \pm 0.1) \cdot 10^{-2}$	$(1.01 \pm 0.01)$	0.70 ; 2.4			
660	$(2.0 \pm 0.4) \cdot 10^{-4}$	$(0.4 \pm 1.7) \cdot 10^{-4}$	0.48 ; 1.9	$(-0.5 \pm 0.1) \cdot 10^{-2}$	$(1.00 \pm 0.00)$	0.62 ; 2.0			
880	$(1.8 \pm 0.4) 10^{-4}$	$(0.4 \pm 2.0) 10^{-4}$	0.40 ; 1.8	$(-0.4 \pm 0.1) 10^{-2}$	$(1.00 \pm 0.00)$	0.54 ; 1.6			
950	$(1.4 \pm 0.3) \cdot 10^{-4}$	$(0.3 \pm 1.4) \cdot 10^{-4}$	0.45 ; 2.0	$(-0.3 \pm 0.1) 10^{-2}$	$(1.00 \pm 0.00)$	0.49 ; 1.5			

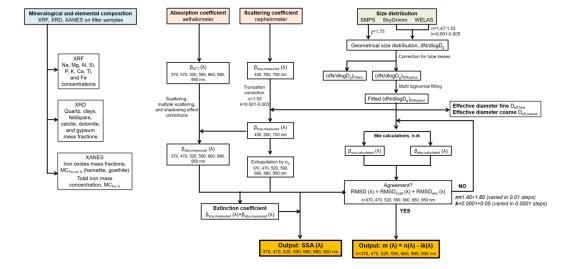
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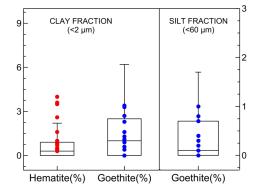


**Figure 1.** Flowchart illustrating the procedure for data treatment and retrieval of physical and chemical (size, composition) and spectral optical properties (single scattering albedo, SSA, and complex refractive index) of mineral dust aerosols.

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**Figure 2.** Box and whisker plot showing the full variability of hematite and goethite mass fractions in the clay and silt soil fractions in the dust source regions at the global scale by considering desert source areas as reported in Ginoux et al. (2012) and DB17. Data are from the soil mineralogical database by Journet et al. (2014). Dots indicate hematite and goethite soil content (extracted from Journet et al.) for the nineteen soils used in the CESAM experiments. The Journet database assumes that the iron oxides in the silt fraction consist only of goethite.

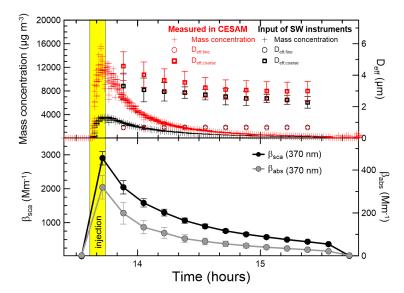


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Figure 3. Top panel: time series of the aerosol mass concentration (cross symbols) and effective fine ( $D_{eff,fine}$ , open dots) and coarse diameter ( $D_{eff,coarse}$ , open squares) measured inside the CESAM chamber (red symbols) and at the input of the SW instruments (black symbols) for one experiment (Morocco dust). Bottom panel: time series of the scattering  $\beta_{sca}$  and absorption  $\beta_{abs}$  coefficients at 370 nm for the same experiment. Mass concentrations are reported as 6-sec data, while all other quantities are 10-min averages.

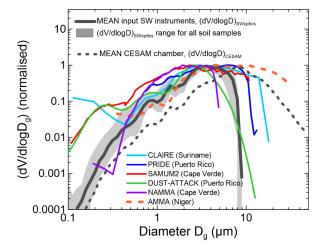


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**Figure 4.** Comparison of dust size distributions sensed by the SW optical instruments (behind the SW instruments inlet (dV/dlogD<sub>g</sub>)<sub>SWoptics</sub>), with field data for long–range transported dust. The thick black line represents the mean value of (dV/dlogD<sub>g</sub>)<sub>SWoptics</sub> at the peak of the dust injection in CESAM for experiments with the different samples. The grey shaded area indicates the range of (dV/dlogD<sub>g</sub>)<sub>SWoptics</sub> for all samples. The dotted black line shows the average of the dust size distribution at the peak of the injection inside the CESAM chamber from DB17. Field data are from: Formenti et al. (2001) (CLAIRE campaign in Suriname, South America), Maring et al. (2003) and Denjean et al. (2016b) (PRIDE and DUST-AT-TACK campaigns in Puerto Rico, Caraibes), Müller et al. (2011) and Chen et al. (2011) (SAMUM2 and NAMMA campaigns in Cape Verde, eastern Atlantic). For comparison, data taken close to the source in Niger from Formenti et al. (2011) during the AMMA campaign are also shown. All data are reported as volume size distributions normalised at the maximum.



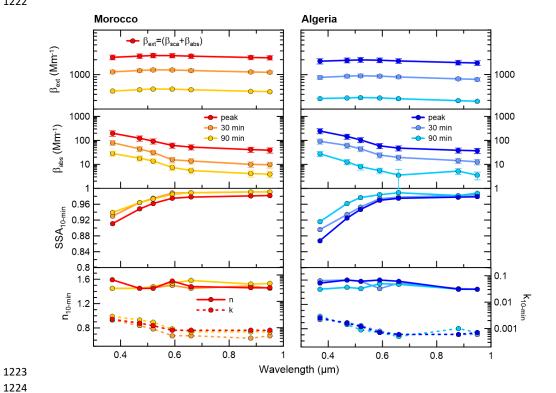
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**Figure 5.** Spectral extinction coefficient, absorption coefficient, SSA, and real (n) and imaginary (k) parts of the refractive index at the peak of the dust injection in the chamber and after 30 and 90 minutes for Morocco and Algeria dust samples. Data are reported at the seven aethalometer wavelengths (370, 470, 520, 590, 660, 880, and 950 nm) as 10–min averages. In the top panel we report the extinction calculated as the sum of scattering and absorption coefficients. For the sake of clarity error bars are not shown for SSA, n, and k data.



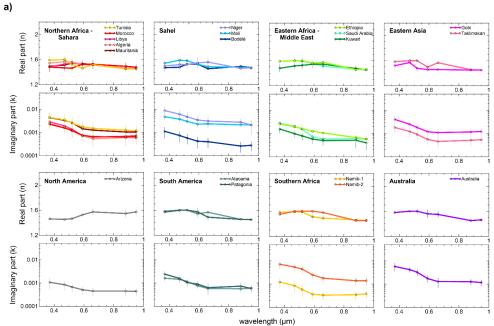


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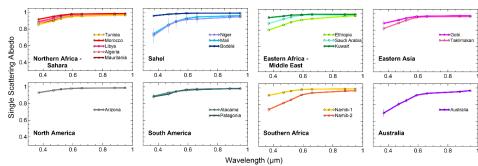




**Figure 6.** (a) Real (n) and imaginary (k) parts of the dust complex refractive index and (b) single scattering albedo (SSA<sub>AVG</sub>) at seven wavelengths between 370 and 950 nm obtained for the 19 aerosol samples analyzed in this study. Data for the refractive index correspond to the time average of the 10 min values obtained between the peak of the injection and 120 min later. The error bar corresponds to the absolute uncertainty in n and k, estimated to be <8% for n and between 13 and 75 % for k. SSA<sub>AVG</sub> data correspond to the fit of the 10 min values of β<sub>sca</sub> versus β<sub>abs</sub>, and the uncertainty is between 1% and 12% at 370 nm and between 1% and 3% at 950 nm.







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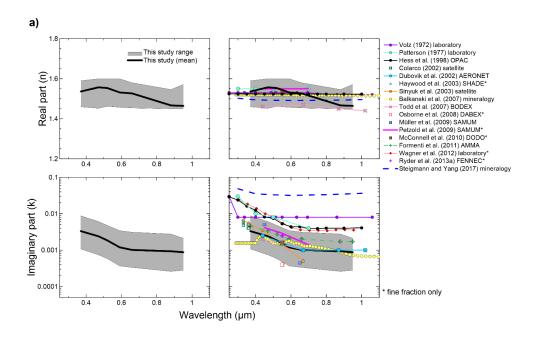
1249 Figure 7. Comparison of results obtained in this study (a) with literature-compiled values (b) of the dust 1250 real and imaginary parts of the refractive index (n, k) and single scattering albedo (SSA) in the SW spectral range. The regions in gray in panel a) indicate the full range of variability obtained in this study, 1251 1252 and the gray thick lines are the means of k and SSA obtained for the different aerosol samples. Litera-1253 ture values in panel b) include estimates from ground-based and aircraft observations during field cam-1254 paigns, laboratory studies, AERONET inversions, and estimates from dust mineralogical composition. 1255 Data are in some cases for the full dust size distribution, while in other only the fine fraction below about 1256 2 μm is measured (identified with \*). 1257 The main provenance of the dust and datasets from the literature is provided in the following: Volz et 1258 al. (1972) is data for rainout dust collected in Germany; Patterson et al. (1977) is Saharan dust; Hess 1259 et al. (1998) is data from the OPAC database; Colarco et al. (2002) is data for dust from Dakar, Sal, and Tenerife; Dubovik et al. (2002) included data from Bahrain-Persian Gulf and Solar Village-Saudi 1260 1261 Arabia AERONET stations; Haywood et al. (2003) is dust from Mauritania; Sinyuk et al. (2003) is data 1262 from Cape Verde, Dakar, and Burkina Faso; Clarke et al. (2004) is Asian dust offshore of China, Japan, 1263 and Korea; Linke et al. (2006)-A is dust from Cairo; Linke et al. (2006)-B is dust from Morocco; Balkanski 1264 et al (2007) is calculated from mineralogical composition assuming a 1.5% hematite mass fraction in 1265 dust; Todd et al. (2007) is from Bodélé; Osborne et al. (2008) is from Niger; Otto et al. (2009), Petzold 1266 et al. (2009), Schladitz et al. (2009), and Muller et al. (2010, 2011) is dust originated mostly in Morocco; 1267 McConnell et al. (2008, 2010) is dust from Niger/Senegal; Chen et al. (2011) is dust from Western 1268 Sahara; Formenti et al. (2011) in the k plot is an average of airborne observations for the AMMA campaign in Niger, while for the SSA plot, Formenti et al. (2011)-A is from observations in the Saharan Air 1269 1270 Layer, -B is from Bodélé/Sudan, and -C is a Sahelian uplift episode; Johnson et al. (2011) is dust from 1271 Western Sahara; Moosmüller et al. (2012) analysed samples from Middle East, Mali and Spain, and 1272 here we report the average of their obtained values; Wagner et al. (2012) obtained k values for several 1273 samples from Cairo and Burkina Fasu and here we report and average of its results; Ryder et al. (2013) 1274 is dust from Western Sahara and Mauritania and we report in both k and SSA plots the average of their 1275 observations; Engelbrecht et al. (2016) analysed many dust samples from all over the world, here we 1276 report their measured minimum and maximum of the dust SSA that are -A from California and -B from 1277 the Etosha Pan in Namibia; Steigmann and Yang (2017) modelled the refractive index of dust based 1278 on assumed mineralogical compositions typical for Northern and Southern Sahara and Western and 1279 Eastern Asia dust, and here we report the average of their results for both n and k. Uncertainties in the 1280 field observations have been omitted for the sake of clarity. The legend identifies the line styles used in 1281 1282 (The different acronyms spell out as: AERONET = Aerosol Robotic Network; OPAC = Optical Properties 1283 of Aerosols and Clouds; SHADE = Saharan Dust Experiment; BODEX = The Bodélé Dust Experiment; 1284 DABEX = Dust and Biomass Experiment; SAMUM = Saharan Mineral Dust Experiment; DODO = Dust 1285 Outflow and Deposition to the Ocean; AMMA = African Monsoon Multidisciplinary Analysis; NAMMA = 1286 NASA African Monsoon Multidisciplinary Analysis; ACE-Asia = Asian Pacific Regional Aerosol Charac-1287 terization Experiment; GERBILS = Geostationary Earth Radiation Budget Intercomparison of Longwave 1288 and Shortwave radiation).

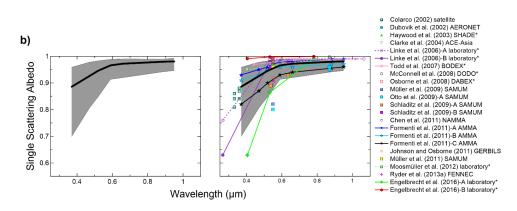
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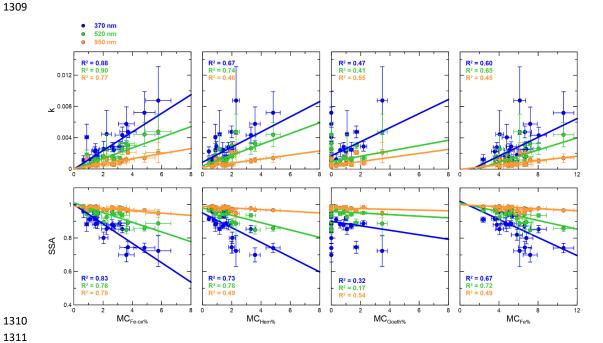


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**Figure 8.** Experiment–averaged imaginary part of the refractive index (k, top panels) and single scattering albedo (SSA, bottom panels) at 370, 520, and 950 nm versus the mass concentration of iron oxides ( $MC_{Fe-ox\%}$ ), hematite ( $MC_{Hem\%}$ ), goethite ( $MC_{Goeth\%}$ ), and elemental iron ( $MC_{Fe\%}$ ) measured for the different dust samples analysed in this study. The calculated linear fit regression lines are shown, together with the correlation coefficients of the fits ( $R^2$ ). The legend indicates the line styles used in the plots. Data for the Taklimakan sample were excluded from the k and SSA plots versus  $MC_{Fe-ox\%}$ ,  $MC_{Hem\%}$ , and  $MC_{Goeth\%}$  due to the absence of data for this sample.



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**Figure 9.** 10–min averaged imaginary refractive index (k<sub>10-min</sub>, top panels) and single scattering albedo (SSA<sub>10-min</sub>, bottom panels) at 370, 520, and 950 nm versus effective coarse diameter (D<sub>eff,coarse</sub>) estimated at the input of the SW instruments. Data were classified in three classes based on the iron oxide content of the dust samples. The linear fit curves and the correlation coefficients for the linear regression fits for each dataset are also reported. The legend identifies the line styles used in the plots.

