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

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Abstract

The optical properties of airborne mineral dust depend on its mineralogy, size distribution, and 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). They 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 (β_{sca}) and absorption (β_{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 β_{sca} and β_{abs} measurements. Dust is assumed to be spherical in the whole data treatment, which introduces a potential source of uncertainty. Our results show that the imaginary part of the refractive index (k) and the SSA vary widely from sample to sample, with values for k in the range 0.0011 to 0.0088 at 370 nm, 0.0006 to 0.0048 at 520 nm, and 0.0003 to 0.0021 at 950 nm, and values for SSA in the range 0.70 to 0.96 at 370 nm, 0.85 to 0.98 at 520 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 wave-

45   length–dependent linear relationship is found between the magnitude of *k* and SSA and the mass con-
46   centrations of both iron oxide and total elemental iron, with iron oxide better correlated than total ele-
47   mental iron to both *k* and SSA. The value of *k* was found to be independent of size. When the iron oxide
48   content exceeds 3%, the SSA linearly decreases with increasing fraction of coarse particles at short
49   wavelengths (< 600 nm).

50   Compared to the literature, our values for the real part of the refractive index and SSA are in line with
51   past results, while we found lower values of *k* compared to most of the literature values currently used
52   in climate models

53   We recommend that source–dependent values of the SW spectral refractive index and SSA be used in
54   models and remote sensing retrievals instead of generic values. In particular, the close relationships
55   found between *k* or SSA and the iron content in dust enable establishing predictive rules for spectrally–
56   resolved SW absorption based on particle composition.

57

58   **Introduction**

59   With teragram amounts of annual emissions, a residence time of about 1–2 weeks in the atmosphere,
60   and a planetary–scale transport, mineral dust aerosols are a global phenomenon (Uno et al., 2009;
61   Ginoux et al., 2012), and contribute significantly to the global and regional aerosol loading (Ridley et
62   al., 2016) and direct radiative effect (Miller et al., 2014).

63   However, large uncertainties still persist on the magnitude and overall sign of the dust direct radiative
64   effect (Boucher et al., 2013; Highwood and Ryder, 2014; Kok et al., 2017). One of the major sources of
65   this uncertainty is our insufficient knowledge of the dust's absorption properties in the shortwave (SW)
66   and longwave (LW) spectral ranges (e.g., Balkanski et al., 2007; Samset et al., 2018), given that mineral
67   dust contains large particles and a variety of minerals absorbing over both spectral regions (e.g. iron
68   oxides, clays, quartz and calcium–rich species; Sokolik and Toon, 1999; Lafon et al., 2006; Di Biagio
69   et al., 2014a, b). Global and regional scale mapping of dust absorption remains limited and more infor-
70   mation is required (Samset et al., 2018).

71   Aerosol absorption is represented both by the imaginary part (*k*) of the complex refractive index ($m=n-$
72   ik) of its constituent material, and by the single scattering albedo (SSA, i.e., the ratio of the scattering
73   to extinction coefficient) of the particle population, as well as by the mass absorption efficiency (MAE,
74   units of $m^2 \ g^{-1}$), i.e., the aerosol absorption coefficient per unit mass concentration.

75   In the shortwave spectral range, absorption by dust accounts for up to ~10–20% of its total extinction.
76   Dust absorption is highest in the UV–VIS, and almost nil towards the near IR (Cattrall et al., 2003;
77   Redmond et al., 2010), due to the combined contribution of large particles in the size distribution and
78   the dust's mineralogy, notably the presence of iron oxides (Karickhoff and Bailey, 1973; Lafon et al.,
79   2006; Derimian et al., 2008; Moosmüller et al., 2012; Formenti et al., 2014a; 2014b; Engelbrecht et al.,
80   2016; Caponi et al., 2017). The mineralogy of airborne mineral dust varies according to that of the
81   parent soils (Nickovic et al., 2012; Journet et al., 2014). Consequently, dust aerosols of different origins
82   should be more or less absorbing in the SW, and have different imaginary spectral refractive index and

83   SSA. Field and laboratory measurements, including ground–based and space–borne remote sensing,
84   show that k varies at a regional scale by almost two orders of magnitude (0.0001–0.008 at 550 nm) with
85   corresponding SSAs between 0.80 and 0.99 at 550 nm (Volz 1972; Patterson et al., 1977; Shettle and
86   Fenn, 1979; Dubovik et al., 2002; Haywood et al., 2003; Sinyuk et al., 2003; Linke et al., 2006; Osborne
87   et al., 2008; Müller et al., 2009; Otto et al., 2009; Petzold et al., 2009; Schladitz et al., 2009; McConnell
88   et al., 2010; Formenti et al., 2011; Wagner et al., 2012; Ryder et al., 2013a; Engelbrecht et al., 2016;
89   Rocha–Lima et al., 2018). Albeit some variability being instrumental or analytical (differences in the
90   sampled size fraction or in the method used to retrieve optical parameters), geographic differences
91   persist when the same measurement approach and retrieval method are applied, e.g., in AERONET
92   inversions, supporting the dependence of dust k and SSA with its origin (Dubovik et al., 2002; Koven
93   and Fung, 2006; Su and Toon, 2011). In contrast, the real part (n) of the dust refractive index, mostly
94   related to particle scattering, is less variable, with values between 1.47–1.56 at 550 nm (e.g., Volz,
95   1972; Patterson et al., 1977; Balkanski et al., 2007; Petzold et al., 2009).

96   Differences in k or SSA caused by the spatial variability of the iron content may affect the sign of the
97   dust radiative effect (heating vs cooling) (Liao and Seinfeld, 1998; Claquin et al. 1999; Miller et al.,
98   2014), and its global and regional implications (Myhre and Stordal, 2001; Colarco et al., 2014; Das et
99   al., 2015; Jin et al., 2016; Bangalath and Stenchikov, 2016; Strong et al., 2018). The direct radiative
100   effect of dust has a strong impact on the Western African Monsoon (Yoshioka et al., 2007; Konaré et
101   al., 2008) and the Indian Summer Monsoon (Vinoj et al., 2014; Das et al., 2015; Jin et al., 2016). How-
102   ever, there is no consensus whether dust increases or decreases precipitation over these regions
103   (Solmon et al., 2008; Jin et al., 2016; Strong et al., 2018). As an example, Solmon et al. (2008) indicate
104   that dust reduces precipitation over most of the Sahelian region, but increases it over the Northern
105   Sahel–Southern Sahara. This pattern is, however, very sensitive to the dust absorbing properties, and
106   a decrease of few percent in dust absorption may even cancel out the increase of precipitation over the
107   Sahel. Similarly, Jin et al. (2016) show that by varying k from zero to 0.008 at 600 nm (i.e., the highest
108   value currently used in models) the dust effect on the Indian Summer Monsoon may shift from negative
109   (reduction of precipitation) to positive (increase of precipitation) values.

110   In spite of this sensitivity, present climate models adopt a globally–constant spectral complex refractive
111   index (and SSA) for dust, and hence still implicitly assume the same dust mineralogical composition at
112   the global scale. This is mainly due to the lack of a globally consistent dataset providing information of
113   the geographical variability of the dust scattering and absorption properties (e.g., Sunset et al., 2018).
114   Reference values for the refractive index are usually taken from Volz (1972), Patterson et al. (1977),
115   D'Almeida et al. (1991), Shettle and Fenn (1979), Sokolik et al. (1993), Sinyuk et al. (2003), or OPAC
116   (Optical Properties of Aerosols and Clouds, Hess et al., 1998; Koepke et al., 2015). A parameterization
117   of the spectrally–resolved dust refractive index as a function of the mineralogical composition of the
118   particles is desirable to replace the globally constant values in current climate models, in particular for
119   those models that started to incorporate the representation of dust mineralogy into their schemes
120   (Scanza et al., 2015; Perlwitz et al., 2015a, 2015b).

121 Improving our knowledge of the spectral SW refractive index of mineral dust and its relation to particle
122 composition (henceforth origin) is also key for the detection of dust aerosols in the atmosphere and the
123 quantification of its mass loading, and total or absorption spectral optical depth from active and passive
124 remote sensing (e.g., Ridley et al., 2016). As an example, the retrieval of the dust SSA and optical depth
125 over bright desert surfaces with the MODIS (Moderate Imaging Resolution Spectroradiometer) Deep
126 Blue algorithm (Hsu et al., 2004) applies the Critical Surface Reflectance Method (Kaufman, 1987) to
127 retrieve dust properties from measured Top of Atmosphere (TOA) spectral reflectance. This algorithm
128 depends critically on a priori information on the spectral refractive index (Kaufman et al., 2001; Yoshida
129 et al., 2013). Similarly, active remote sensing techniques (lidar, light detection and ranging) require the
130 knowledge of the extinction–to–backscatter ratio (the lidar ratio), which is also a strong function of the
131 complex index of refraction or SSA of the aerosol particles (e.g., Gasteiger et al., 2011; Shin et al.,
132 2018). Gasteiger et al. (2011) have shown in fact that a 5% change in the SSA at 532 nm can modify
133 by up to 20% the lidar ratio of dust, which means a 20% change in the estimated profile of the dust
134 extinction coefficient and retrieved optical depth from lidar measurements.

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

144 **2. Experimental set–up and instrumentation**

145 As previously described in DB17 and C17, all experiments discussed here and were conducted in the
146 4.2 m^3 stainless–steel CESAM chamber (French acronym for Experimental Multiphasic Atmospheric
147 Simulation Chamber) (Wang et al., 2011). Mineral dust aerosols were generated by mechanical shaking
148 of parent soils using about 15 g of soil sample (first sieved to <1000 μm and then dried at 100 °C)
149 placed in a 1 L Büchner flask and shaken for about 30 min at 100 Hz by means of a sieve shaker
150 (Retsch AS200). The dust suspension in the flask was injected into the chamber by flushing with N_2 at
151 10 L min^{-1} for about 10–15 min. After injection in the chamber, the largest fraction of the dust aerosol
152 (>1.5 μm diameter) remained in suspension for approximately 60 to 120 min thanks to a four–blade
153 stainless steel fan located at the bottom of the chamber, which also ensured homogeneous conditions
154 within the chamber volume. The submicron dust fraction, instead, remained constant with time during
155 the experiments (see Sect. 4.1.1). The evolution of the physico–chemical and optical properties of the
156 suspended dust was measured by different instruments connected to the chamber. The spectral particle
157 volume dry scattering (β_{sca}) and absorption (β_{abs}) coefficients were measured, respectively, by a 3–
158 wavelength nephelometer (TSI Inc. model 3563, operating at 450, 550, and 700 nm; 2 L min^{-1} flow rate,
159 2–s time resolution) and a 7–wavelength aethalometer (Magee Sci. model AE31, operating at 370, 470,

160 520, 590, 660, 880 and 950 nm; 2 L min^{-1} flow rate, 2–min time resolution). The size distribution of

161 aerosols was measured by means of a scanning mobility particle sizer (SMPS, TSI, DMA Model 3080,

162 CPC Model 3772; mobility diameter range 0.019–0.882 µm; 2.0/0.2 L min^{-1} sheath–aerosol flow rates,

163 135–s time resolution), a WELAS optical particle counter (OPC) (PALAS, model 2000, white light source

164 between 0.35 and 0.70 µm; optical–equivalent diameter range 0.58–40.7 µm; 2 L min^{-1} flow rate, 1–

165 min time resolution) and a SkyGrimm OPC (Grimm Inc., model 1.129, 0.655 µm operating wavelength;

166 optical–equivalent diameter range 0.25–32 µm; 1.2 L min^{-1} flow rate, 6–s time resolution). Aerosol ele-

167 mental and mineralogical composition, including iron oxides, was derived by analysis of dust samples

168 collected on polycarbonate filters (47–mm diameter Nuclepore, Whatman, nominal pore size 0.4 µm)

169 mounted in a custom–made stainless–steel sample holder (operated at 6 L min^{-1}) for most of the dura-

170 tion of each experiment.

171 All instruments (size, SW optics, filters) sampled air from the chamber. To equalize the airflow extracted

172 by the different instruments, a particle–free N_2/O_2 mixture airflow was continuously injected into the

173 chamber. Inlets for all extractive measurements consisted of a stainless steel tube located inside

174 CESAM, and an external connection of silicone tubing (TSI Inc.) from the chamber to the instruments,

175 for a total length varying between 0.4 and 1.2 m. As detailed in DB17 and shown in Fig. S1 in the

176 supplement, the transmission efficiency due to aspiration and transmission in the sampling lines as a

177 function of particle diameter was estimated to calculate the effective dust fraction sensed by each in-

178 strument, taking into account the sampling flow rate, tubing diameter, tubing geometry, and particle

179 shape and density. For the nephelometer and the aethalometer, the length of the sampling line from

180 the intake point in the chamber to the instrument entrance was about 1.2 m, which resulted in a 50%

181 cutoff of the transmission efficiency at 3.9 µm particle geometric diameter and 100% cutoff at 10 µm.

182 For the filter sampling system, the length of the sampling line of about 0.5 m resulted in a 50% (100%)

183 cutoff at 6.5 µm (15 µm) particle geometric diameter (or 50% cutoff at 10.6 µm aerodynamic diameter

184 as indicated in C17, therefore compositional analyses refer to the $PM_{10.6}$ size fraction). For the WELAS,

185 the only OPC considered for size distribution in the coarse fraction (see Sect. 2.2), the 50% (100%)

186 cutoff was reached for particles of 5 µm (8 µm) diameter.

187 All experiments were conducted at ambient temperature and relative humidity <2%. In addition to over-

188 night evacuation, the chamber was manually cleaned between experiments to avoid contaminations

189 from remaining dust. Background concentrations of aerosols in the chamber were less than 2.0 µg m^{-3}

190 (that is 10^2 to 10^5 times smaller than the concentration of dust aerosols in suspension in the chamber

191 during experiments)

192 A flowchart of the procedure used to treat and combine optical, size, and compositional data, and the

193 algorithm for SSA and complex refractive index retrieval is shown in Fig. 1. Full details of data treatment

194 for size distribution measurements and filter compositional data are provided in DB17 and C17, and in

195 the following we only mention the main points of interest for the present paper. Full details on the data

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

197 The optical and size datasets were acquired at different temporal resolutions and then averaged over

198 compatible 10–min intervals, whereas the compositional data represent the experiment integral. The

199 SSA and complex refractive index data were retrieved both at 10–min resolution and as experiment
200 averages to relate them to both size and compositional data. Table 1 summarizes the uncertainties on
201 the measured and derived parameters described in the following.

2.1 SW optical measurements

2.1.1 Aerosol scattering coefficient

204 The aerosol scattering coefficients (β_{sca}) at 450, 550, and 700 nm are measured by the nephelometer
205 at angles between 7° and 170° and need to be corrected for the restricted field–of–view of the instru-
206 ment (truncation correction) to retrieve β_{sca} at 0°–180°. The truncation correction factor (C_{trunc}), i.e., the
207 ratio of the β_{sca} at 0°–180° and 7°–170°, was estimated by Mie calculations for homogeneous spherical
208 particles using the size distribution measured simultaneously behind SW inlets (see Sect. 2.2). In the
209 calculations, the real part of the complex refractive index of dust was assumed to be wavelength–inde-
210 pendent and fixed at a value of 1.53, while the imaginary part was set to 0.003 at 450 and 550 nm and
211 to 0.001 at 700 nm, according to pre–existing information (Sinyuk et al., 2003; Schladitz et al., 2009;
212 Formenti et al., 2011; Rocha–Lima et al., 2018). For the different dust samples, C_{trunc} ranged between
213 1.2 and 1.7 and decreased with wavelength and the dust residence time in the chamber, following the
214 relative importance of the coarse component in the dust population. The uncertainty on C_{trunc}, calculated
215 by repeating the optical calculations by using the size distribution of dust within its error bars as input
216 to the optical code, is less than ±5% at all wavelengths. In order to assess the consistency of the derived
217 truncation correction, we made a sensitivity study in which we recalculated C_{trunc} by varying the refrac-
218 tive index used as input to the Mie calculations in the range of n and k values obtained in this study
219 (i.e., values at the 10% and 90% percentile as reported in Table 5 for the whole dataset, that is n
220 between 1.49 and 1.54 and k between 0.001 and 0.006 at 450, 550, and 700 nm). The results of this
221 sensitivity study indicate that, for fixed dust size distribution, the truncation correction C_{trunc} varies less
222 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
223 insensitive to the exact assumed n and k values.

224 Once corrected for truncation, the spectral β_{sca} was extrapolated at the aethalometer wavelengths. With
225 this aim, the Scattering Ångström Exponents, $SAE_{450–550}$ and $SAE_{550–700}$, were calculated as the linear
226 fit of β_{sca} vs λ at 450–550 nm and 550–700 nm, respectively. The $SAE_{450–550}$ and $SAE_{550–700}$ coefficients
227 were used to extrapolate β_{sca} at wavelengths respectively lower and higher than 550 nm. Extrapolated
228 β_{sca} values were used to derive an average SAE of dust for the entire investigated spectral range.

2.1.2 Aerosol absorption coefficient

230 The aerosol absorption coefficient (β_{abs}) at 370, 470, 520, 590, 660, 880, and 950 nm was retrieved
231 from aethalometer measurements. The aethalometer measures the attenuation (ATT) through an aer-
232 osol–laden quartz filter, related to the spectral attenuation coefficient (β_{ATT}) as:

233
$$\beta_{ATT}(\lambda) = \frac{\Delta ATT(\lambda)}{\Delta t}\frac{A}{V} \quad (1)$$

234 where A is the area of the aerosol collection spot (0.5 ± 0.1) cm^2 and V the air sample flow rate (0.002

235 m^3 min^{-1}). The slope $\frac{\Delta ATT(\lambda)}{\Delta t}$ is the linear fit of the measured attenuation as a function of time calcu-

236 lated over 10–min intervals. The spectral attenuation coefficient was converted into an absorption co-

237 efficient β_{abs} following the formula by Collaud Coen et al. (2010):

238
$$\beta_{abs}(\lambda) = \frac{\beta_{ATT}(\lambda) - \alpha(\lambda)\beta_{sca}(\lambda)}{C_{ref}R(\lambda)} \qquad (2)$$

239 The $\alpha(\lambda)\beta_{sca}(\lambda)$ term accounts for the fraction of the measured attenuation due to side and backward

240 scattering and not to light absorption. The Collaud–Coen correction scheme has been recently shown

241 to yield quite accurate values of the absorption coefficients and absorption Ångström exponents from

242 aethalometer data (Saturno et al., 2017). The value of $\alpha(\lambda)$ was calculated with the formula by Arnott et

243 al. (2005) and varied between 0.002 and 0.02 (<±1% from formal error propagation on the Arnott for-

244 mula), while $\beta_{sca}(\lambda)$ is the scattering coefficient from the nephelometer extrapolated to the aethalometer

245 wavelengths. The C_{ref} term accounts for multiple scattering by the filter fibers, aerosol laden or not. Its

246 spectral value, obtained by the linear extrapolation of C_{ref} at 450 and 660 nm estimated for mineral dust

247 by Di Biagio et al. (2017b), varied between 4.30 at 370 nm and 3.32 at 950 nm. We assume for the

248 extrapolated C_{ref} an uncertainty of ±10% as estimated in Di Biagio et al. (2017b). The correction factor,

249 R, accounts for the decrease in the aethalometer sensitivity with the increase of the aerosol filter load-

250 ing. The value of R depends on the absorptivity properties of the sampled aerosol and can be calculated

251 as a function of the particle SSA. In this study, we calculated R by estimating a first–guess SSA* as the

252 ratio of the nephelometer–corrected β_{sca} and β_{ext} obtained as the sum of β_{sca} and the β_{abs} non–corrected

253 for filter loading effect. The R was estimated by using the Collaud–Coen et al. (2010) formulation. For

254 the range of estimated SSA* (about 0.60 to 0.99), R varied between 0.5 and 1.0 (±1–10%).

255 The Absorption Ångström Exponent (AAE) was calculated as the power–law fit of β_{abs} versus λ.

256 Due to an instrumental problem, aethalometer data were not always available, with a typical 30–min

257 interruption usually 10 to 30 minutes after the beginning of experiments.

258 **2.2 Size distribution**

259 The aerosol number size distribution was obtained from SMPS, WELAS and SkyGrimm measurements

260 over different diameter ranges. The measured electrical mobility and optical equivalent diameters from

261 the SMPS and the OPCs were first converted into geometrical diameters (D_g) as described in DB17

262 and summarized in Table 1. The OPCs conversion assumes a dust complex refractive index that in our

263 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 WELAS

264 (following DB17, for more details see Table 1). After conversion, the estimated D_g range was 0.01–0.50

265 μm for the SMPS, 0.65–73.0 μm for the WELAS, and 0.29–68.2 μm for the SkyGrimm. Due to a cali-

266 bration issue, data for the SkyGrimm in the range D_g > 1μm were discarded, so that the WELAS is the

267 only instrument considered in the super–micron range. A very low counting efficiency was observed for

268 the WELAS below 1 μm and data in this size range were also discarded.

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

270 distribution of the dust aerosols suspended in the CESAM chamber, $(dN/dlogD_g)_{CESAM}$, and the size

271 distribution behind SW optical instruments inlets, $(dN/dlogD_g)_{SWoptics}$, after taking into account particle

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

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

274 $(dN/dlogD_g)_{SWoptics}$ size distribution is cut at 10 microns, so no particles above this diameter reach the

275 SW instruments.

276 The measured size distributions, $(dN/dlogD_g)_{CESAM}$ and $(dN/dlogD_g)_{SWoptics}$, were used to estimate the

277 mass concentration of aerosols and their effective diameter (D_{eff}) in the CESAM chamber and behind

278 the SW instrument inlets as:

279
$$\text{Mass concentration} = \int \frac{\pi}{6} D_g^3 \frac{dN}{dlogD_g} \rho \cdot dlogD_g \qquad (3)$$

280
$$D_{eff} = \frac{\int D_g^3 \frac{dN}{dlogD_g} dlogD_g}{\int D_g^2 \frac{dN}{dlogD_g} dlogD_g} \quad (4)$$

281 The effective dust density ρ in Eq. (3) was set at 2.5 g cm^{-3}, a value that is approximately in the middle

282 of the range of desert dust densities reported in the literature, i.e., 2.1–2.75 g cm^{-3} (Maring et al., 2000;

283 Iwasaka et al., 2003; Reid et al., 2003). The effective diameter was evaluated separately for the fine

284 and coarse fractions of dust by integrating Eq. (4) for diameters ≤1 μm ($D_{eff,fine}$) and >1 μm ($D_{eff,coarse}$),

285 respectively. For $D_{eff,coarse}$ the upper limit of the calculation is 10 μm when calculated from

286 $(dN/dlogD_g)_{SWoptics}$, i.e. measured behind the SW inlets.

287 The dust size distribution, $(dN/dlogD)_{SWoptics}$, measured at each 10–min time step for each sample was

288 fitted with a sum of five lognormal functions to smooth data inhomogeneities linked to the different

289 instrument's operating principles and artefacts. Fitting was performed using the Levenberg–Marquardt

290 algorithm. For each mode, the parameters of the lognormal functions, i.e., the total number concentra-

291 tion (N_i), the geometric median diameter $(D_{g,i})$, and the geometric standard deviation of the distribution

292 (σ_i), were retrieved. The uncertainties in the retrieved parameters were estimated by repeating the fit

293 using size data within their uncertainties. The resulting parameters of the fits at the peak of the injection

294 in the chamber are reported in Table S1, and an example of size fitting is shown in Fig. S2.

295 The procedure described here to estimate $(dN/dlogD_g)_{CESAM}$ and $(dN/dlogD_g)_{SWoptics}$ implies that as-

296 sumptions are made on the values of n and k to correct OPCs data, and this may introduce a circularity

297 in the estimates of the refractive index of dust that use $(dN/dlogD_g)_{SWoptics}$ as input in optical calculations

298 (see Sect. 3.2). In order to analyze the dependence of the results on this assumption, we made a

299 sensitivity calculation by varying the values of n and k used for OPCs corrections within the range of

300 values retrieved in this study (10% and 90% percentiles in Table 5, i.e., 1.49–1.54 for n and 0.001–

301 0.006 for k). We concluded that changing n and k in this range has a very low impact on the retrieved

302 number size distribution behind the SW inlets $(dN/dlogD_g)_{SWoptics}$ compared to the original assumptions

303 made in our calculations (<5% changes in the retrieved size number distribution at the different diame-
304 ters between the original correction and the correction by varying n and k). This is due to the fact that
305 when changing D_g due to changes in the n and k in the OPCs correction, the loss function also modifies
306 to values corresponding to the new D_g. Given that the loss function increases/decreases for increas-
307 ing/decreasing D_g, the combined changes in D_g and the loss function compensate so that the net num-
308 ber concentration behind the SW inlets varies less than a few percent. These results therefore suggest
309 that the procedure to estimate the complex refractive index of dust is nearly independent of the assumed
310 OPC correction.

311 Other sources of uncertainties are linked to the spherical assumption to perform the optical to geomet-
312 rical diameter conversion (discussed in Sect. 3.3) as well as those due to Mie resonance oscillations of
313 the calculated scattering intensities. Concerning Mie resonances, a sensitivity study was performed
314 varying the size resolution of our calculations (high/low diameter resolution in the calculations to have
315 a better/worse reproduction of Mie resonance oscillations) and show that Mie resonances impact the
316 optical to geometrical correction by less than 1%.

317 **2.3 Dust elemental and mineralogical composition and iron content**

318 The elemental and mineralogical composition of the dust aerosols in the $PM_{10.6}$ size fraction was esti-
319 mated by combining different techniques: X–ray diffraction (XRD, Panalytical model Empyrean diffrac-
320 tometer) to estimate the particles' mineralogical composition in terms of clays, quartz, calcite, dolomite,
321 gypsum, and feldspars; wavelength dispersive X–ray fluorescence (WD–XRF, Panalytical PW–2404
322 spectrometer) to determine the dust elemental composition (Na, Mg, Al, Si, P, K, Ca, Ti, Fe); and X–
323 ray absorption near–edge structure (XANES) to retrieve the content of iron oxides and their speciation
324 between hematite and goethite. The dust mass collected on Nuclepore filters during the experiments
325 varied between 0.3 and 6 mg m^{-3} as calculated from elemental concentrations according to Lide (1992).

326 Full details on the XRD, WD–XRF, and XANES measurements and data analysis are provided in DB17
327 and C17. In this study, we discuss the dust elemental iron mass concentration, $MC_{Fe\%}$, i.e., the percent
328 mass of elemental iron with respect to the total dust mass concentration, and the iron oxides mass
329 concentration, $MC_{Fe-ox\%}$, i.e., the percent mass fraction of iron oxides with respect to the total dust mass
330 concentration, estimated as the sum of goethite ($MC_{Goet\%}$) and hematite ($MC_{Hem\%}$) species.

331 **3. Strategy for data analysis**

332 **3.1 Calculation of the spectral extinction coefficient and SSA from scattering and absorption**
333 **coefficients**

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

337 $$\beta_{ext}(\lambda) = \beta_{abs}(\lambda) + \beta_{sca}(\lambda) \qquad (5).$$

338 The Extinction Ångström Exponent (EAE) was calculated as the power–law fit of β_{ext} versus λ.

339 The spectral single scattering albedo of dust at 10–min resolution (SSA_{10-min}) was retrieved as:

340
$$SSA_{10\text{-min}}\left(\lambda\right)=\frac{\beta_{sca}(\lambda)}{\beta_{ext}(\lambda)} \qquad (6).$$

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

343
$$SSA\left(\lambda\right)=\left(1+\frac{1}{m(\lambda)}\right)^{-1} \qquad (7)$$

344 where $m\,(\lambda)$ represents the slope of the linear fit between the 10–min averages of $\beta_{sca}\,(\lambda)$ and $\beta_{abs}\,(\lambda)$
345 measured along the whole duration of each experiment. An example of $\beta_{sca}\,(\lambda)$ versus $\beta_{abs}\,(\lambda)$ fitting to
346 retrieve the spectral SSA is shown in Fig. S3 in the Supplement. The correlation coefficient R^2 of the
347 $\beta_{sca}\,(\lambda)$ versus $\beta_{abs}\,(\lambda)$ fit usually ranges between 0.97 and 1 at all wavelengths. As will be discussed
348 later in the paper, the single scattering albedo of dust depends on the particle coarse size fraction, and
349 during our experiments $SSA_{10\text{–min}}$ was not derived continuously for the different samples due to the
350 aethalometer measurement interruptions. The application of Eq. (7) avoids any bias in the calculated
351 averaged SSA for different soils due to size effects. For two of the analyzed samples (Tunisia and
352 Namib–2), however, the linear fitting procedure was not applicable due to the fact that, respectively,
353 only two and one absorption measurements from the aethalometer were available just after the peak of
354 the injection, with no data afterwards. Average SSA data for Tunisia were thus estimated as the mean
355 of the two available $SSA_{10\text{–min}}$ data points, while the single $SSA_{10\text{–min}}$ measurement at the peak of the
356 injection was reported for Namib–2. This difference in time sampling should be kept in mind when com-
357 paring data for these two samples to the rest of the dataset.

358 **3.2 Retrieval of the spectral complex refractive index**

359 An optical calculation was performed to estimate the complex refractive index ($m=n–ik$) of dust aerosols
360 based on optical and size data. The retrieval algorithm consisted in recalculating the spectral scattering
361 $\beta_{sca}\,(\lambda)$ and absorption $\beta_{abs}\,(\lambda)$ coefficients measured at each 10–min interval by using the fitted
362 $(dN/dlogD)_{SWoptics}$ size distribution as input and by varying the real and imaginary parts of the complex
363 refractive index in the calculations until the best agreement between measurements and calculations
364 was found. At each wavelength the root mean square deviation (RMSD) was calculated as:

365
$$RMSD(\lambda)=\sqrt{\left[\frac{\beta_{sca,measured}\left(\lambda\right)-\beta_{sca,calculated}\left(\lambda\right)\left(n,k\right)}{\beta_{sca,calculated}\left(\lambda\right)\left(n,k\right)}\right]^2+\left[\frac{\beta_{abs,measured}\left(\lambda\right)-\beta_{abs,calculated}\left(\lambda\right)\left(n,k\right)}{\beta_{abs,calculated}\left(\lambda\right)\left(n,k\right)}\right]^2} \qquad (8)$$

366 The RMSD was minimized at each wavelength to obtain $n–k$ pairs that most closely reproduce the
367 measured scattering and absorption coefficients. Optical calculations were performed at the 7 wave-
368 lengths of the aethalometer between 370 and 950 nm using Mie theory. In the calculations, the real part
369 of the refractive index was varied in the range 1.40–1.60 at steps of 0.01, while the imaginary part was
370 varied in the range 0.0001–0.050 at steps of 0.0001. For each sample, this resulted in 10500 compu-
371 tations per wavelength and per 10–min time step. The uncertainty on the real and imaginary parts of
372 the refractive index was estimated with a sensitivity study. For this purpose, the values of n and k were
373 also obtained by using as input the observed $\beta_{sca}\,(\lambda)$, $\beta_{abs}\,(\lambda)$, and $(dN/dlogD)_{SWoptics}$, plus or minus one

374 standard deviation on their measurement. The deviations of the values of n and k retrieved in the sen-
375 sitivity study with respect to those obtained in the first inversions were assumed to correspond to the
376 one standard deviation uncertainty of 10–min retrieved values.

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

380 A control experiment was performed with submicron ammonium sulphate aerosols (see DB17 and sup-
381 plementary Fig. S4) with the aim of validating the proposed methodology to estimate the aerosol com-
382 plex refractive index for a non–absorbing aerosol type. For ammonium sulphate particles with a mono–
383 modal size distribution centered at 0.06 μm, as measured with the SMPS, the retrieved real part of the
384 refractive index was 1.56 (±0.01) in the 450–700 nm wavelength range, as expected from literature
385 (Toon et al., 1976; Flores et al., 2009; Denjean et al., 2014).

3.3 Assumptions on the retrieval of SSA and complex refractive index

387 The approach used to retrieve the SSA and the complex refractive index of dust and the accuracy of
388 the results depend on the accuracy of the input data and the assumptions in the optical calculations.
389 We discuss here two points of the applied procedure, in part already mentioned in the previous para-
390 graphs.

391 1/ The size distribution from OPCs and also the scattering coefficient from the nephelometer used as
392 input to the n and k retrieval procedure and SSA calculation depend more or less directly on the dust
393 refractive index. These instruments need in fact to be corrected for instrumental artefacts and these
394 corrections require an a priori knowledge of the n and k, which in our approach were set to fixed values
395 (1.47–1.53 for n and 0.001–0.005 for k for OPCs optical to geometrical diameter conversion, and 1.53
396 for n and 0.001–0.003 for k for nephelometer truncation correction). This choice may in principle intro-
397 duce a certain degree of uncertainty and circularity into the derived n, k, and SSA for dust. Nonetheless,
398 we note that the range of refractive index values used to correct OPCs and nephelometer data falls in
399 the range of variability of the refractive index values obtained in this study (see Sect. 4.3), which sug-
400 gests that the values used for the corrections are appropriate. Additionally, as previously discussed,
401 both the size distribution $(dN/dlogD_g)_{SWoptics}$ and the scattering coefficient are not very sensitive to the
402 assumptions about n and k used for the calculations (less than 5% changes in both the number size
403 distribution behind SW inlets and the scattering coefficient from changing n and k within the range of
404 estimated values in this study) which further demonstrates the robustness of the proposed approach.

405 2/ The retrieval procedure for n and k, as well as the calculations for OPCs optical–to–geometrical
406 diameter and the nephelometer truncation correction, simplifies the non–spherical heterogeneous dust
407 aerosols (e.g., Chou et al. 2008; Okada et al., 2011; Nousiainen and Kandler, 2015) into homogeneous
408 spherical particles that can be represented by Mie theory. In the present study, we decided not to use
409 a more advanced shape–representing theory for three main reasons. First, the spherical model has
410 been shown to produce only moderate errors when computing angular–integrated quantities
411 (Mishchenko et al., 1995; Otto et al., 2009; Sorribas et al., 2015) such as those we calculate in this

study to retrieve the OPC and truncation corrections and for n and k retrieval. For instance, Sorribas et al. (2015) showed that using a spheroidal model has a limited effect on the truncation correction. These authors estimated that using a spheroidal model permits to improve by 4 to 13% the agreement between modelled and measured spectral scattering coefficient at 450–700 nm but only for supermicron parti-cles. Conversely, for submicron dust the spherical approximation is better suited than the spheroidal model to reproduce the scattering coefficients by the nephelometer. The study by Mogili et al. (2007) also found an excellent agreement between measured shortwave extinction spectra and those calcu-lated from Mie theory simulations for dust minerals, supporting the use of Mie theory for dust optical modelling. On the other side, other studies point to the need of a non–spherical assumption to improve the modelling of dust optical properties (e.g., Otto et al., 2009). Second, we used Mie theory for the sake of comparison with the large majority of previous field and laboratory data published so far, which had used calculations with the spherical approximation Third, the shape distribution and morphology of the dust samples was not measured during experiments. Improper assumptions on the particle shape and morphology may 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 contradictory results on the true impact of dust non–sphericity on radiative fluxes and heating rates from global model simula-tions (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 tech-niques using spectral, angular, and polarized reflectance measurements (e.g., Feng et al., 2009). In synthesis, accounting for shape effects is still controversial for dust modelling and also a complex issue beyond the scope of this paper. Thus, while we acknowledge the potential uncertainties induced by spherical assumptions in our study, we do not quantify here the overall impact of this assumption on our results.

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). 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 goe-thite 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

451 Figure 3 shows a typical example of a time series of aerosol mass concentration and effective fine and
452 coarse diameters measured inside the CESAM chamber and behind the SW instruments inlets during
453 the experiments, as well as the corresponding β_{sca} and β_{abs} at 370 nm. The Figure shows the rapid
454 increase of the mass concentration within CESAM during dust injection in the chamber, and its subse-
455 quent decrease during the experiments due to both size–selective gravitational settling, occurring
456 mostly within the first 30 min of experiments, and dilution by sampling. The scattering and absorption
457 coefficients of dust decrease with time after injection in tandem with the decrease of the mass concen-
458 tration and the size–dependent depletion in the chamber. The dust mass concentration inside CESAM
459 at the peak of the injection is between 2 mg m^{-3} (Mali) and 310 mg m^{-3} (Bodélé) and falls to values
460 between 0.9 mg m^{-3} (Mali) and 20 mg m^{-3} (Bodélé) behind the SW instruments inlets. These values
461 are comparable to those measured close to sources during dust storms (Rajot et al., 2008; Kander et
462 al., 2009). After 2 hours, the dust mass concentration has decreased to values of 0.2 to 2.5 mg m^{-3}
463 (inside CESAM) and of 0.1 to 1.9 mg m^{-3} (behind the SW inlets), as after medium– to long–range dust
464 transport in the real atmosphere (Weinzerl et al., 2011; Denjean et al., 2016b). This indicates that in a
465 2–hour experiment in CESAM it is possible to reproduce the temporal changes of the dust mass load
466 observed in the real atmosphere from emission to medium/long–range transport.

467 As the mass concentration, the effective diameter of the coarse fraction, $D_{eff,coarse}$, also rapidly de-
468 creases with time due the progressive deposition of the coarsest particles in the chamber. For the var-
469 ious soils, $D_{eff,coarse}$ varies in the range of 4–8 µm (peak of injection) to 3–4 µm (after 2 hours) inside the
470 CESAM chamber, and in the range of 3–4 µm (peak of injection) to 2–3 µm (after 2 hours) behind the
471 SW inlets. In contrast, $D_{eff,fine}$ remains quite constant during the experiments, with a value between 0.6
472 and 0.7 µm for all soils. The values of $D_{eff,coarse}$ obtained in this study inside the CESAM chamber are in
473 line with those measured close to African sources (4–12 µm, Rajot et al., 2008; Weinzerl et al., 2009;
474 Ryder et al., 2013a) and for dust transported across the Mediterranean (5–8 µm, Denjean et al., 2016a).
475 Conversely, the values of $D_{eff,coarse}$ behind the SW instruments inlets are mostly in agreement with those
476 reported for dust transported at Cape Verde and across the Atlantic ocean (~3 µm, Maring et al., 2003;
477 Müller et al., 2011; Denjean et al., 2016b). Our values of $D_{eff,fine}$ are higher compared to values reported
478 by Denjean et al. (2016a) for dust aerosols transported over the Mediterranean (0.2 to 0.5 µm), reflect-
479 ing the fact that we analyse pure dust whereas these authors often encountered dust externally mixed
480 with pollution particles.

481 The comparison of $D_{eff,coarse}$ values suggests that while the size distribution in CESAM is mostly repre-
482 sentative of dust close to sources (see DB17), the size measured behind the SW instruments inlets is
483 mostly representative of transport conditions. Figure 4 illustrates this point by showing the volume size
484 distributions of the generated dust aerosols at the peak of injection seen by the SW optical instruments,
485 compared to the average size of dust measured in CESAM (DB17) and field observations close to
486 sources (e.g., Niger) and after long–range transport (Cape Verde, Suriname, Puerto Rico, and Barba-
487 dos). The size distribution of dust inside CESAM includes a coarse mode up to ~ 50 µm and well repro-
488 duces field observations close to sources, as shown in comparison to the Niger case. Due to particle
489 losses along tubes, particles above 10 µm diameter are not seen by the SW instruments. The overall
490 shape of the dust size distribution sensed by the SW instruments is comparable to that measured after

491 atmospheric long–range transport, even if the fraction of particles above 3.9 µm diameter, which is at
492 the 50% cutoff of the transmission efficiency for the SW optical instruments, is significantly under–
493 represented compared to observations (i.e., Betzer et al., 1988; Formenti et al., 2001; Maring et al.,
494 2003; Ryder et al., 2013b, 2018; Jeong et al., 2014; Denjean et al., 2016b). It should be keep in mind
495 that often also field data are affected by inlet restrictions so that they cannot measure the whole coarse
496 dust fraction (see Table 1 in Ryder et al., 2018). The lowest cutoff for field data shown in Fig. 4 are for
497 the NAMMA and PRIDE datasets and correspond to upper size limits at 5 and 10 µm in diameter,
498 respectively. Being these values above our cutoff of 3.9 µm, it means that the comparison with our size
499 dataset is meaningful within the range of our measurements. To note that only the data from AER–D
500 did not suffer from significant inlet restrictions thus leading to the observation of giant dust particles up
501 to tens of microns in the Saharan Air Layer off the coasts of Western Africa.

4.1.2 Iron and iron oxide dust content

503 Elemental iron includes the iron in the form of iron oxides and hydroxides, i.e. hematite and goethite
504 (the so–called free iron, mostly controlling SW absorption) and the iron incorporated in the crystal struc-
505 ture of silicates and alluminosilicates (illite, smectite), which does not substantially contribute to SW
506 absorption (Karickhoff and Bailey, 1973; Lafon et al., 2004). The mass concentrations of these compo-
507 nents (total iron oxides, hematite, goethite, and total elemental iron) for the different analysed samples
508 are reported in Table 3. There is a considerable variability in the iron and iron oxide content for our
509 samples. Total iron in the dust samples is in the range from 2.4% (Namib–1) to 10.6% (Namib–2). Iron
510 oxides account for 11% to 62% of the iron mass (calculated following C17, not reported in Table 3),
511 whereas the percent of iron oxides to the total dust mass varies between 0.7% (Bodélé Depression)
512 and 5.8% (Niger). These data are in the range of values reported in the literature (Reid et al., 2003;
513 Scheuvens et al., 2013; Formenti et al., 2011, 2014a). For the samples from the Sahara and the Sahel,
514 goethite is the dominant iron oxide species, in agreement with Lafon et al. (2006) and Formenti et al.
515 (2014a; 2014b). Elsewhere, hematite dominates over goethite, as reported by some studies (Arimoto
516 et al., 2002; Shen et al., 2006; Lu et al., 2011).

4.2 Spectral– and time–dependent dust extinction and absorption coefficients, complex refrac-
tive index, and SSA

519 Figure 5 illustrates a typical spectral– and time–dependent set of measured optical properties. The
520 spectral extinction coefficient, absorption coefficient, SSA, and real and imaginary parts of the complex
521 refractive index obtained at 10–min resolution for the Morocco and Algeria samples are shown at the
522 peak of the dust injection in CESAM and 30 and 90 min after the peak. Figure 5 shows that absorption
523 decreases with wavelength, but not extinction. The SSA increases from 370 to 590 nm while it is almost
524 constant between 590 and 950 nm. The imaginary part of the refractive index decreases with λ following
525 the decrease of β_{abs}. The real part of the refractive index does not depend on wavelength.

526 The extinction and absorption coefficients decrease in absolute value with time, as already shown in
527 Fig. 3. Their spectral dependence remains quite constant with time, but varies from soil to soil. The
528 experiment–averaged absorption, scattering, and extinction Ångström exponents in the 370–950 nm

529 spectral range, representing the spectral variation of the absorption, scattering and extinction coeffi-
530 cients, vary between the values of 1.5 and 2.4 (AAE), −0.4 and 0.4 (SAE), and −0.2 and +0.5 (EAE)
531 for the different samples. These values are in line with those previously reported by Moosmüller et al.
532 (2012) and C17 for dust from various locations. The retrieved n and k also show negligible changes of
533 their spectral shape with time and their magnitude remains approximately constant. In contrast, the SSA
534 increases with time, in particular below 600 nm wavelength, and its spectral shape changes. This is
535 mostly due to the decrease of the coarse size fraction with residence time in the chamber, as will be
536 analysed in Sect. 4.5. Similarly to the absorption, scattering, and extinction coefficients, the spectral
537 shape of k and SSA is somewhat different between the various samples, with the sharpest spectral
538 variations observed for the most absorbing samples and a less pronounced spectral variation for the
539 less absorbing ones, as evident, for example, by comparing the SSA data for Morocco and Algeria in
540 Fig. 5.

541 **4.3 Spectral complex refractive index and SSA for the different source regions and comparison**
542 **to literature data**

543 Figures 6 and 7 show the experiment–averaged n, k, and SSA between 370 and 950 nm for the nine-
544 teen aerosol samples analyzed in this study. Data of n, k, and SSA and their uncertainties are reported
545 in Tables 4 and 5 for each sample together with the average values for each of the eight different source
546 regions and for the full dataset. Figures 6 and 7 show that there are significant differences, both in
547 magnitude and spectral shape, between the imaginary refractive index and SSA for the different sam-
548 ples. The highest values of k (0.0048–0.0088 at 370 nm and 0.0012–0.0021 at 950 nm) and lowest
549 values of SSA (0.70–0.75 at 370 nm and 0.95–0.97 at 950 nm) are obtained for the Niger, Mali, Namib–
550 2 and Australia samples, which also show the highest values of both the iron oxide content between
551 3.6% and 5.8% and hematite content between 2.0% and 4.8%. The lowest values (k is 0.0011–0.0012
552 at 370 and 0.0003–0.0004 at 950 nm, and SSA is in the range 0.91–0.96 at 370 nm and 0.97–0.99 at
553 950 nm) are obtained for the Bodélé, Namib–1, and Arizona samples, which have iron oxide contents
554 between 0.7% and 1.5%. Both k and SSA vary from region to region, with the largest absorptions (high-
555 est k, lowest SSA) for the Sahel and Australia and the lowest absorption (lowest k, highest SSA) in
556 North and South America and the Middle East; k and SSA values also vary within the same region, as
557 illustrated for the Sahelian and Southern African samples. The real part of the refractive index, on the
558 other hand, is not only almost wavelength–independent, as anticipated, but also relatively invariant from
559 sample to sample. Its average over the 370–950 nm spectral range is between 1.48 (Gobi) and 1.55
560 (Ethiopia and Namib–2).

561 The full envelope of n, k, and SSA obtained for the entire set of analysed samples is shown in Fig. 8.
562 The real refractive index is relatively invariant, while the spectral k varies by up to an order of magnitude
563 (0.001–0.009 at 370 nm and 0.0003–0.002 at 950 nm). The SSA changes accordingly for the different
564 dust samples at the different wavelengths (30% change at 370 nm corresponding to values between
565 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
566 (as spectral average) and varies in the range 0.0033–0.0009 for k and 0.85–0.98 for the SSA between
567 370 and 950 nm (0.0016 and 0.94 as spectral averages for k and SSA) (Fig. 8 and Tables 4 and 5).

568   The comparison between the full envelope of *n*, *k*, and SSA in this study with literature data is also

569   shown in Fig. 8. Literature values considered for comparison include estimates from ground–based,

570   aircraft, and satellite observations, laboratory studies, AERONET inversions, and estimates from mixing

571   rules based on the dust mineralogical composition. Given that the sample selection in our experiments

572   fully envelopes the global variability of mineralogy of natural dust, we could expect that our dataset

573   would also fully envelope the global–scale variability of the dust absorption and scattering properties in

574   the SW. When comparing with available literature data we found that our *n* and SSA datasets very well

575   encompass the range of values indicated in the literature, with only a few outlier points. In contrast, for

576   the imaginary refractive index the reported range of variability from the literature is significantly larger

577   than that found in our study, with our range of *k* being mostly at the lower bound of previous results.

578   Nonetheless, our range of *k* values fully envelopes the ensemble of remote sensing and field campaign

579   data on airborne dust from the previous literature reported in Fig. 8a. The global average spectral values

580   for *k* in our study (thick black line) perfectly match the Dubovik et al. (2002) dataset from a synthesis of

581   AERONET observations from various locations worldwide. Likewise, our *k* average is also very close

582   to the dataset by Balkanski et al. (2007), estimated from mineralogical composition assuming 1.5% (by

583   volume) of hematite in dust, a value shown to allow a reconciliation of climate modelling and satellite

584   observations of the dust direct SW radiative effect. By comparison, the average dust hematite content

585   for the ensemble of our analysed samples is 1.8% (in mass), close to the 1.5% value proposed by

586   Balkanski et al. (2007).

587   Looking at Fig. 8, the datasets that show the largest values, which also fall outside our estimated range

588   of *k* over the entire considered wavelength range are the ones by: (i) Volz (1972), Patterson et al. (1977)

589   and Hess et al. (1998; i.e., the OPAC 3.1 version database, which is the same *k* dataset used in the

590   new OPAC 4.0 version, Koepke et al., 2015) showing larger values than our dataset over the entire

591   considered wavelength range. These datasets are amongst the most commonly used references for

592   the dust imaginary refractive index in many climate models; and (ii) the dataset by Wagner et al. (2012)

593   obtained from laboratory chamber experiments, deviating especially below 600 nm wavelength from

594   our range of *k*. The reasons for these discrepancies in the *k* values are difficult to assess, since they

595   could be related to both instrumental and analytical aspects. In the studies by Volz (1972) and Patterson

596   et al. (1977), for instance, the complex refractive index was obtained by transmittance and diffuse re-

597   flectance on pellet samples, a technique that requires the dust to be pressed in a matrix of non–absorb-

598   ing material. In this case a discrepancy arises from the different optical behaviour between dust com-

599   pressed in a pellet and the airborne particles. Moreover, Volz (1972) and Patterson et al. (1977) analyse

600   dust aerosols collected after mid– to long–range transport, thus after the dust has possibly been mixed

601   with absorbing species.

602   For the case of Wagner et al. (2012) the imaginary refractive index was retrieved from laboratory cham-

603   ber experiments on suspended dust, as in our study. Nonetheless, their approach differs in various

604   aspects from the one applied here and this can lead to the observed differences in the retrieved *k*. First,

605   the aerosol generation technique is different between the two works and this possibly leads to particles

606   with different physico–chemical features compared to our study. In Wagner et al. (2012) the dust aerosol

607   was generated by a rotating brush disperser using only the 20–75 µm sieved fraction of the soils. This

608   system acts to disaggregate the finest particles of the soil by passing it through a nozzle. Then the

609   largest aerosol grains were removed by a cyclone system (50% cutoff at 1.2 μm aerodynamic diameter),

610   so that only the submicron size fraction was measured. We show in Sect. 4.5 that k is independent of

611   size for the range of investigated effective coarse diameters between 2 and 4 μm, but the range of sizes

612   analysed in Wagner et al. (2012) is significantly lower than in our study and a size–effect cannot be

613   excluded. In fact, the relationship between dust absorption and iron content may vary depending on the

614   considered size fraction (see C17) due to the fact that iron bearing minerals are more concentrated in

615   the clay fraction (<2.0 μm) of the dust (Kandler et al., 2009). Moreover, generating dust in a different

616   way may lead to differences in the chemical and mineralogical size–dependent composition of the sam-

617   ple, therefore contributing to the observed differences. The impact of this is however difficult to evaluate.

618   Another difference concerns the choice of the optical theory to retrieve k (T–matrix in Wagner et al.

619   instead of Mie theory as used in our work). This can contribute to the observed differences, even if in a

620   limited way (Mogili et al., 2007; Sorribas et al., 2015). Third, in their retrieval Wagner et al. fixed the real

621   refractive index to a wavelength–independent value of 1.53 (as done in several other field and labora-

622   tory studies in Fig. 8) and this assumption can bias high/low the retrieved k if the actual n is higher/lower

623   than the assumed 1.53 value. So, in summary, while multiple factors could contribute to the discrepancy

624   it remains however difficult to assess which source of discrepancy is dominant.

625   4.4 Imaginary refractive index and SSA versus iron and iron oxide content

626   The sample–to–sample variability of the imaginary part of the refractive index k and the SSA observed

627   in Fig. 6 and 7 is related to the dust composition by investigating the dependence on the particle iron

628   content. In Fig. 9 we show the experiment–averaged k and SSA at 370, 520, and 950 nm versus the

629   mass concentration of iron oxides (hematite+goethite, $MC_{Fe-ox\%}$), hematite ($MC_{Hem\%}$), goethite

630   ($MC_{Goeth\%}$), and total elemental iron ($MC_{Fe\%}$) measured for the different dust samples in this study. The

631   data are linearly fitted to relate k and SSA to $MC_{Fe-ox\%}$, $MC_{Hem\%}$, $MC_{Goeth\%}$, and $MC_{Fe\%}$. The results of

632   the fits at all wavelengths between 370 and 950 nm are reported in Table 6, together with the statistical

633   indicators of the goodness of fit (correlation coefficient, R^2, and reduced chi square, χ^2_{red}, i.e., the ob-

634   tained chi square divided by the number of degrees of freedom). There is an excellent correlation be-

635   tween both k and SSA and $MC_{Fe-ox\%}$ at the different wavelengths (R^2>0.75). A weaker correlation is

636   found when relating k and SSA to $MC_{Hem\%}$ and $MC_{Fe\%}$ (R^2 between 0.40 and 0.74 for k and between

637   0.49 and 0.78 for the SSA), and $MC_{Goeth\%}$ (R^2 between 0.17 and 0.62). The better correlation of k and

638   SSA to $MC_{Fe-ox\%}$ compared to $MC_{Fe\%}$ is expected since dust optical properties in the visible wavelengths

639   are mostly sensitive to the fraction of iron oxides, rather than to iron incorporated into the crystal struc-

640   ture of silicates (Karickhoff and Bailey, 1973; Lafon et al., 2006; Moosmüller et al., 2012; Klaver et al.,

641   2011; Engelbrecht et al., 2016; C17). The quantities that most robustly satisfy a linear relationship are

642   k and $MC_{Fe-ox\%}$, as indicated by the reduced chi square χ^2_{red} that is around 1 at all different wavelengths.

643   The χ^2_{red} increases to values also larger than 2 in the other cases, indicating the poorer robustness of

644   the fit in these cases.

645   We also investigated the dependence of the spectral k and SSA on the mass concentration of other

646   minerals, such as clays, calcite, quartz, and feldspars, and also on the mass concentration of different

647 elements. We found that there is no statistically significant correlation between k or SSA and the mass
648 concentration of any of these compounds (not shown), with R^2 values between 0.002 and 0.46 at the
649 different wavelengths for all cases.

650 These results therefore clearly show that iron, particularly in the form of iron oxides (hematite + goe-
651 thite), is the main driver of dust shortwave absorption. Measuring only the hematite mass fraction to
652 estimate the dust absorption, as it is sometimes done, is therefore not sufficient.

653 **4.5 Imaginary refractive index and SSA versus dust coarse size fraction**

654 The dependence of the spectral k and SSA on the dust coarse fraction is investigated by relating it to
655 the $D_{eff,coarse}$ calculated from the size distribution data behind the SW instruments inlets. The k_{10-min} and
656 SSA_{10-min} at 370, 520, and 950 nm versus $D_{eff,coarse}$ are shown in Fig. 10 for all experimental data, which
657 we separated into three classes based on their iron oxide content ($MC_{Fe-ox\%} \leq 1.5\%$, $1.5\% < MC_{Fe-ox\%}$
658 $< 3\%$, $MC_{Fe-ox\%} \geq 3\%$). Figure 10 shows that even if the correlation is not very strong ($R^2 < 0.54$), there
659 is a clearly decreasing tendency for the SSA_{10-min} with increasing $D_{eff,coarse}$, particularly at 370 and 520
660 nm for strongly absorbing samples with iron oxide content larger than 3%. The SSA_{10-min} is mostly
661 independent of changes of $D_{eff,coarse}$ at 950 nm. Conversely, k_{10-min} has a very poor correlation with
662 $D_{eff,coarse}$ ($R^2 < 0.35$) and thus does not depend on size. Similar results were also obtained for the real
663 part (not shown).

664 These results confirm previous observations (Sokolik and Toon, 1999; McConnell et al., 2008, 2010;
665 Ryder et al., 2013a; 2013b) that the refractive index is independent of size. This suggests that size–
666 dependent mineralogical composition is not sufficient to affect k (in the limit of our measurement and
667 retrieval procedure precision). It is worth mentioning that only few past studies evidenced a dependence
668 of k on the size distribution of the dust aerosols (i.e., Kandler et al., 2009, 2011; Otto et al., 2009) maybe
669 because the refractive index was retrieved in these studies from mixing rules based on the estimated
670 size–dependent mineralogical composition.

671 Differently from k, the SSA increases as the coarse dust size fraction decreases. This is due to the fact
672 that absorption efficiency for a single particle (Q_{abs}) increases with particle diameter while the scattering
673 efficiency (Q_{sca}) decreases. Ryder et al. (2013a) also showed that the dependence of SSA on size is
674 linear, but important only when the coarse fraction is high (if particles larger than about 3 µm in diameter
675 are present), otherwise the SSA depends mainly on composition, also in agreement with more recent
676 field observations by Ryder et al. (2018).

677 **5. Summary**

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

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

691 Some other laboratory studies have been performed in the past to investigate the shortwave SSA of
692 dust from different sources worldwide and its dependence on composition (Linke et al., 2006; Moosmül-
693 ler et al., 2012; Engelbrecht et al., 2016). Conversely, for the refractive index there exists to our
694 knowledge only one other chamber study (Wagner et al., 2012), that retrieved the imaginary part k
695 between 305 and 955 nm for dust aerosols from a limited number of source areas in Africa (Burkina
696 Faso, Egypt and Morocco). As a matter of fact, our work provides the first consistent simulation chamber
697 study of the complex refractive index of global dust.

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

699 1. The spectral k and SSA retrieved in this study vary from sample to sample within the same region
700   but also from a region to another. For k, values vary between 0.0011–0.0088 at 370 nm, 0.0006 to
701   0.0048 at 520 nm, and 0.0003–0.002 at 950 nm. For SSA, values vary from 0.70 to 0.96 at 370 nm,
702   0.85 to 0.98 at 520 nm, and from 0.95 at 0.99 at 950 nm. In contrast, n is wavelength–independent
703   and almost uniform for the different sources, with values between 1.48 and 1.55. Values for n and
704   SSA fall within the range of published literature estimates, while for k we obtain a much narrower
705   range of variability than the ensemble of literature results, as illustrated in Fig. 8. In particular, we
706   found lower values of k compared to most of the literature values currently used in climate models,
707   such as Volz et al. (1972), Patterson et al. (1977), and the OPAC database (Hess et al., 1998;
708   Koepke et al., 2015). In their study, Miller et al. (2014) state that the values of Dubovik et al. (2002)
709   from AERONET, Patterson et al. (1977) for far–travelled dust, and OPAC probably bracket the
710   global solar absorption by dust. In contrast, our results indicate that dust absorption is lower than
711   previously thought, and its average is close to the values reported by Dubovik et al. (2002) from
712   AERONET observations and Balkanski et al. (2007) for a dust with a 1.5% volume fraction of hem-
713   atite. Our range of variability of an order of magnitude for k and between 4% and 30% for the
714   spectral SSA is actually large enough to change the sign of the global dust direct effect at the TOA
715   (Miller et al., 2004), as well as its regional implications (e.g., Solmon et al., 2008; Jin et al., 2016),
716   and has to be taken into account in climate modelling.
717 2. The documented changes in k and SSA also impact remote sensing retrievals. To give an example,
718   following Gasteiger et al. (2011), our observed variability of about 10% for the SSA at 532 nm would
719   translate to about 40% variability in the retrieved extinction profiles and optical depths from lidar
720   observations for dust from varying sources.
721 3. The sample–to–sample variability observed in this study is mostly related to the iron oxide and
722   elemental iron content in dust. At each investigated wavelength the magnitude of k and SSA is

723   linearly correlated to the mass concentration of total iron oxides, hematite, goethite, and total ele-
724   mental iron. Small variations of these compounds translate into large variations of k and SSA.

725 4. We also investigated the dependence of k and SSA on the size distribution of dust. While k is
726   independent of size (suggesting that a constant value can be used along transport), below 600 nm
727   the SSA linearly decreases for increasing $D_{eff,coarse}$ for strongly absorbing samples with more than
728   3% iron oxide content. The investigated range of $D_{eff,coarse}$ is within about 2 and 4 µm, and thus
729   comparable to values obtained along a transport path over the Atlantic Ocean for dust during about
730   2 to 6 days following emission (Denjean et al., 2016a).

731 5. The observations of points 3 and 4 suggest that while it is sufficient to know the content of iron
732   oxide (or elemental iron) in dust to predict its spectral k, which means that only one tracer is needed
733   in models to parametrize its regional and global variability, for the spectral SSA both composition
734   and size distribution are required.

6. Concluding remarks

737 Based on our results, we recommend that dust simulations, as well as remote sensing retrievals, use
738 source–dependent values of the spectral SW refractive index and SSA instead of generic values. We
739 propose, as a first step, a set of regionally–averaged n, k, and SSA values to represent dust from each
740 of the eight regions analysed here as well as a global average value from the ensemble of our data
741 (Tables 4 and 5). Furthermore, the relationships found between k and SSA and the iron oxides or ele-
742 mental iron content in dust open the perspective to establish predictive rules to estimate the spectrally–
743 resolved SW absorption of dust based on composition. We recommend the use of iron oxide content
744 rather than iron content as it is better correlated with k and SSA. The relationship found in this study,
745 nonetheless, refer to the bulk composition of the dust aerosols and to a size range typical of 2 to 6 days
746 of transport in the atmosphere. As demonstrated in C17 for the mass extinction efficiency, the relation-
747 ships linking the dust absorption to iron content vary as a function of the analysed size fraction due to
748 the fact that iron bearing minerals are more concentrated in the clay fraction (<2.0 µm) than in the
749 coarsest fraction of the dust (Kandler et al., 2009; C17). Further investigation should be therefore ad-
750 dressed to evaluate the dependence of the spectral k and SSA versus iron content as a function of the
751 size distribution of the particles, in particular extending to a wider range of $D_{eff,coarse}$ compared to the
752 one investigated in the present study. This will allow to determine if the k and SSA versus iron relation-
753 ships change or not in different phases of the aerosol lifetime, so if it is valid close to source areas
754 (when the coarsest fraction is dominant, i.e. $D_{eff,coarse}$ up to 15 µm, Ryder et al. (2013b)), and in long–
755 range transport conditions (when most of the coarse particle fraction above few µm has settled out (i.e.,
756 $D_{eff,coarse}$ of 2–3 µm or lower, Denjean et al. (2016b)).

757 We point out, however, that the use of mineralogy to estimate k and SSA based on linear relationships,
758 as obtained in our study, requires that the model–predicted dust composition accurately reflects that of
759 the natural atmospheric aerosols. To this aim, realistic soil mineralogy databases and accurate model-
760 ling of the soil to aerosol size fractionation need to be developed in model schemes. In this sense we
761 mention the EMIT project (Earth Surface Mineral Dust Source Investigation) as a potential near–future
762 source of high resolution surface mineralogy data for arid and semi–arid regions based on imaging

spectroscopy satellite data (Green et al., 2018). Also, a realistic representation of the size distribution, in particular the coarse mode fraction of dust and its retention during atmospheric transport, should be provided in models given its importance in affecting the SSA, as shown in this study and previously reported in other papers (Ryder et al., 2013a, 2013b, 2018; Kok et al., 2017).

Our study focuses on the dust spectral optical properties between 370 and 950 nm. Further work is required to extend the range of spectral refractive index and SSA data to wavelengths lower than 370 nm or higher than 950 nm given that these data are often required in Global Circulation Models and Numerical Weather Prediction models.

We do not provide any quantification of the uncertainty associated with the assumption of spherical particles in our study, even if we acknowledge the potential role of non–sphericity in affecting our data treatment and results. Additional work is foreseen to better investigate the shape of our generated dust and the impact of non–sphericity on retrieved spectral refractive indices and SSA.

Finally, this study had the objective to investigate the variability of the dust SW optical properties at the global scale linked 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 variability over these regions is comparable to the one 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 be therefore devoted to better characterize the variability of dust spectral optical properties at the regional and sub–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 and will be compiled together with aerosol properties from other studies within the Library of Advanced Data Products (LADP) of the EUROCHAMP datacenter (https://data.eurochamp.org). The CESAM data used in this study are immediately available upon request to the contact author and will also soon be made available through the Database of Atmospheric Simulation Chamber Studies (DASCS) of the EUROCHAMP datacenter (https://data.eurochamp.org /)).

Code availability

The following IDL routines were used in the analysis: mpfitexy.pro (available at https://github.com/williamsmj/mpfitexy) was used to linearly fit data taking into account uncertainties on both x and y; mie_single.pro (available at http://www.atm.ox.ac.uk/code/mie/mie_single.html) was used for optical calculations using Mie theory; mpcurvefit.pro (available at http://cow.physics.wisc.edu/~craigm/idl/idl.html) 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. S. Nowak performed the XRD measurements. C. Di Biagio and P. Formenti wrote the manuscript with comments from all co–authors.

Competing interests

The authors declare that they have no conflict of interest.

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	Parameter	Time resolution	Relative uncertainty	Uncertainty calculation	Comments
Optical SW	Scattering coefficient at 450, 550, and 700 nm, β_{sca} (λ)	*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 β_{sca} (λ) 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_{abs} (λ)	*10–min data*	22–30% at 370 nm 23–87% at 950 nm	Error propagation formula[1] on Eq. (2) considering the uncertainties on $\beta_{ATT}(\lambda)$ from 10–min fitting procedure (error propagation formula[1] on Eq. 1, ~20%), and uncertainties on $\alpha(\lambda)$ (1%), $\beta_{sca}(\lambda)$ (5–12%), C_{ref} (10%), and R (1–10%).	
	Extinction coefficient, β_{ext} (λ) = β_{sca} (λ) + β_{abs} (λ)	*10–min data*	~25%	Sum of β_{sca} (λ) and β_{abs} (λ) uncertainties	
	Single Scattering Albedo, SSA (λ) = β_{sca} (λ) / (β_{sca} (λ) + β_{abs} (λ))	*10–min data*	9–12%	Error propagation formula[1] considering single uncertainties on $\beta_{sca}(\lambda)$ and $\beta_{abs}(\lambda)$.	
	Single Scattering Albedo, SSA (λ) = $(1+1/m(\lambda))^{-1}$	*Experiment averaged*	1–12% at 370 nm 1–3% at 950 nm	Error propagation formula[1] on Eq. (6) considering the uncertainty on m(λ), i.e., the slope of the linear fit between β_{sca} (λ) and β_{abs} (λ) over the whole duration of each experiment.	
	Complex refractive 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[1] 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 in their overlapping range (see DB17)

	SkyGrimm geometrical diameter (D_g)	–	<15.2%	Standard deviation of the D_g values obtained for different refractive indices values used in the optical to geometrical conversion	The conversion of optical to geometrical diameters for the SkyGrimm and the WELAS was performed by taking into account the visible complex refractive index of dust aerosols. Optical calculations were computed at the SkyGrimm operating wavelength (0.655 µm) and over the spectral range of the WELAS (0.35 to 0.7 µm) using Mie theory for spherical particles by fixing n at 1.47, 1.50, and 1.53, and by varying k in steps of 0.001 between 0.001 and 0.005. Then D_g is set at the mean ± 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.
	WELAS geometrical diameter (D_g)	–	<7%	The same as for the SkyGrimm	
	$(dN/dlogD)_{SWoptics}$	*10–min data*	~20–90%	Error propagation formula[1] considering the $dN/dlogD_g$ st. dev. over 10–min and the uncertainty on particle loss function along sampling tubes $L(D_g)$ (~50% at 2 µm, ~10% at 8 µm)	The uncertainty of $L(D_g)$ 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)
	$D_{eff,fine}$	*10–min data*	<5%	Deviation obtained by repeating the calculations by using the size distribution ± its uncertainty.	
	$D_{eff,coarse}$	*10–min data*	5–40%		
Mineralogical composition	Elemental iron mass concentration ($MC_{Fe\%}$)	*Experiment averaged*	10%	Uncertainties calculated as discussed in DB17 and C17	
	Iron oxides mass concentration ($MC_{Fe–ox\%}$)	*Experiment averaged*	15%		
	Goethite mass concentration ($MC_{Goet\%}$)	*Experiment averaged*	<10%		
	Hematite mass concentration ($MC_{Hem\%}$)	*Experiment averaged*	<10%		

1192   [1] $\sigma_f = \sqrt{\sum_{i=1}^{n}\left(\dfrac{\partial f}{\partial x_i}\sigma_{x_i}\right)^2}$

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1203 **Table 2.** Summary of information on the soil samples and sediments used in this study.
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Geographical area	Sample	Coordinates	Desert area
Northern Africa – Sahara	Tunisia	33.02°N, 10.67°E	Maouna
	Morocco	31.97°N, 3.28°W	east of Ksar Sahli
	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
Sahel	Niger	13.52°N, 2.63°E	Banizoumbou
	Mali	17.62°N, 4.29°W	Dar el Beida
	Bodélé	17.23°N, 19.03°E	Bodélé depression
Eastern Africa and the Middle East	Ethiopia	7.50°N, 38.65°E	Lake Shala National Park
	Saudi Arabia	27.49°N, 41.98°E	Nefud
	Kuwait	29.42°N, 47.69°E	Kuwaiti
Eastern Asia	Gobi	39.43°N, 105.67°E	Gobi
	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
	Patagonia	50.26°S, 71.50°W	Patagonia
Southern Africa	Namib–1	21.24°S, 14.99°E	Namib
	Namib–2	19.00°S, 13.00°E	Namib
Australia	Australia	31.33°S, 140.33°E	Strzelecki

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1208 **Table 3**. Chemical characterization of the dust aerosols in the $PM_{10.6}$ size fraction. Column 3 shows
1209 $MC_{Fe\%}$, the fractional mass of elemental iron with respect to the total dust mass concentration (±10%
1210 relative uncertainty), and column 4 reports $MC_{Fe-ox\%}$, the mass fraction of iron oxides with respect to the
1211 total dust mass concentration (±15% relative uncertainty) and its speciation in hematite $MC_{Hem\%}$ and
1212 goethite $MC_{Goeth\%}$ (<±10% relative uncertainty). The iron oxide measurements were not made on the
1213 Taklimakan sample. Mean values and standard deviations based on single sample data are reported
1214 for the full dataset.

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Geographical area	Sample	$MC_{Fe\%}$	$MC_{Fe-ox\%}$	$MC_{Hem\%}$	$MC_{Goeth\%}$
Northern Africa – Sahara	Tunisia	4.1	2.2	1.2	1.1
	Morocco	3.6	1.4	0.4	1.0
	Libya	5.2	3.1	0.9	2.2
	Algeria	6.6	2.7	1.4	1.4
	Mauritania	8.1	3.3	3.3	0.0
Sahel	Niger	6.1	5.8	2.3	3.5
	Mali	6.6	3.7	2.0	1.7
	Bodélé	4.1	0.7	0.7	0.0
Eastern Africa and the Middle East	Ethiopia	6.8	2.0	2.0	0.0
	Saudi Arabia	3.8	2.6	1.8	0.8
	Kuwait	5.0	1.5	1.5	0.0
Eastern Asia	Gobi	4.8	0.9	0.9	0.0
	Taklimakan	5.8	–	–	–
North America	Arizona	5.3	1.5	1.5	0.0
South America	Atacama	4.7	1.6	1.6	0.0
	Patagonia	5.1	1.5	0.9	0.6
Southern Africa	Namib–1	2.4	1.1	0.8	0.3
	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)**

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 (n_{SW}) is indicated.

Sample/Region	n_{SW} 037–0.95 µm	σ_{nSW} 037–0.95 µm	k 0.37 µm	0.47 µm	0.52 µm	0.59 µm	0.66 µm	0.88 µm	0.95 µm	σ_k 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 Africa – Sahara (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 Africa 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 America (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 America (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 Africa (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	**1.52**		**0.0026**	**0.0019**	**0.0014**	**0.0008**	**0.0006**	**0.0007**	**0.0006**							
Full dataset 10% percentile	**1.49**		**0.0012**	**0.0009**	**0.0007**	**0.0005**	**0.0004**	**0.0004**	**0.0004**							
Full dataset 90% percentile	**1.54**		**0.0061**	**0.0044**	**0.0035**	**0.0023**	**0.0019**	**0.0015**	**0.0015**							

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

Sample/Region	SSA							σ_{SSA}						
	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	0.85	0.90	0.93	0.95	0.95	0.97	0.97	0.03	0.02	0.02	0.01	0.01	0.01	0.01
Morocco	0.92	0.95	0.96	0.98	0.98	0.98	0.99	0.01	0.01	0.01	0.00	0.00	0.00	0.00
Lybia	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
Algeria	0.87	0.92	0.94	0.97	0.97	0.98	0.98	0.02	0.01	0.01	0.00	0.00	0.00	0.00
Mauritania	0.85	0.90	0.94	0.96	0.97	0.98	0.98	0.02	0.01	0.01	0.01	0.01	0.00	0.00
Northern Africa – Sahara (mean and st. dev.)	**0.88**	**0.92**	**0.94**	**0.97**	**0.97**	**0.98**	**0.98**	**0.03**	**0.02**	**0.02**	**0.01**	**0.01**	**0.01**	**0.01**
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	0.75	0.85	0.89	0.93	0.95	0.96	0.96	0.04	0.03	0.02	0.02	0.02	0.01	0.01
Bodélé	0.96	0.98	0.98	0.99	0.99	0.99	0.99	0.04	0.02	0.02	0.01	0.01	0.01	0.01
Sahel (mean and st. dev.)	**0.81**	**0.89**	**0.92**	**0.94**	**0.95**	**0.96**	**0.97**	**0.13**	**0.07**	**0.05**	**0.04**	**0.04**	**0.03**	**0.02**
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 Arabia	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
Patagonia	0.88	0.91	0.94	0.96	0.97	0.98	0.98	0.02	0.02	0.01	0.01	0.01	0.00	0.01
South America (mean and st. dev.)	**0.89**	**0.92**	**0.94**	**0.96**	**0.97**	**0.98**	**0.98**	**0.00**	**0.01**	**0.00**	**0.00**	**0.00**	**0.00**	**0.00**
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 dataset (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 dataset median	**0.88**	**0.92**	**0.94**	**0.96**	**0.97**	**0.98**	**0.98**							
Full dataset 10% percentile	**0.74**	**0.84**	**0.88**	**0.92**	**0.94**	**0.96**	**0.96**							
Full dataset 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 χ^2_{red} is the reduced chi–square of the fit.

	$k = a\,MC_{Fe-ox\%} + b$			$SSA = a\,MC_{Fe-ox\%} + b$		
Wavelength (nm)	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}
370	$(11.9 \pm 2.4)\,10^{-4}$	$(2.4 \pm 4.6)\,10^{-4}$	0.88 ; 0.6	$(-5.8 \pm 0.8)\,10^{-2}$	(1.00 ± 0.02)	0.83 ; 1.7
470	$(9.0 \pm 1.7)\,10^{-4}$	$(1.7 \pm 3.2)\,10^{-4}$	0.89 ; 0.8	$(-3.8 \pm 0.6)\,10^{-2}$	(1.00 ± 0.01)	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 ± 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 ± 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 ± 0.00)	0.75 ; 2.2
880	$(3.4 \pm 0.6)\,10^{-4}$	$(0.6 \pm 1.2)\,10^{-4}$	0.79 ; 1.0	$(-0.76 \pm 0.16)\,10^{-2}$	(1.00 ± 0.00)	0.79 ; 1.4
950	$(3.2 \pm 0.6)\,10^{-4}$	$(0.6 \pm 1.0)\,10^{-4}$	0.77 ; 1.1	$(-0.62 \pm 0.13)\,10^{-2}$	(0.99 ± 0.00)	0.78 ; 1.1

	$k = a\,MC_{Hem\%} + b$			$SSA = a\,MC_{Hem\%} + b$		
Wavelength (nm)	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}
370	$(9.7 \pm 2.7)\,10^{-4}$	$(2.7 \pm 4.0)\,10^{-4}$	0.67 ; 1.9	$(-4.4 \pm 0.6)\,10^{-2}$	(0.95 ± 0.01)	0.73 ; 3.5
470	$(8.3 \pm 1.9)\,10^{-4}$	$(1.9 \pm 2.7)\,10^{-4}$	0.72 ; 1.9	$(-3.0 \pm 0.4)\,10^{-2}$	(0.97 ± 0.01)	0.76 ; 3.2
520	$(6.9 \pm 1.5)\,10^{-4}$	$(1.5 \pm 2.0)\,10^{-4}$	0.74 ; 2.0	$(-2.2 \pm 0.3)\,10^{-2}$	(0.98 ± 0.00)	0.78 ; 3.3
590	$(3.7 \pm 0.8)\,10^{-4}$	$(0.9 \pm 1.2)\,10^{-4}$	0.61 ; 2.1	$(-1.3 \pm 0.2)\,10^{-2}$	(0.99 ± 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 ± 0.00)	0.62 ; 2.5
880	$(2.9 \pm 0.7)\,10^{-4}$	$(0.7 \pm 1.1)\,10^{-4}$	0.43 ; 2.1	$(-0.6 \pm 0.1)\,10^{-2}$	(0.99 ± 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 ± 0.00)	0.49 ; 1.7

	$k = a\,MC_{Goeth\%} + b$			$SSA = a\,MC_{Goeth\%} + b$		
Wavelength (nm)	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}
370	$(9.0 \pm 2.5)\,10^{-4}$	$(2.5 \pm 2.2)\,10^{-4}$	0.47 ; 1.8	$(-13.4 \pm 6.9)\,10^{-3}$	(0.90 ± 0.01)	0.32 ; 6.8
470	$(5.5 \pm 1.7)\,10^{-4}$	$(1.7 \pm 1.5)\,10^{-4}$	0.43 ; 2.3	$(-8.3 \pm 4.7)\,10^{-3}$	(0.94 ± 0.00)	0.21 ; 6.2
520	$(3.4 \pm 1.1)\,10^{-4}$	$(1.1 \pm 1.2)\,10^{-4}$	0.41 ; 2.5	$(-4.9 \pm 3.2)\,10^{-3}$	(0.96 ± 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 ± 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 ± 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	$(-1.1 \pm 1.4)\,10^{-3}$	(0.98 ± 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	$(-2.1 \pm 1.4)\,10^{-3}$	(0.98 ± 0.00)	0.54 ; 2.6

|	$k = a\,MC_{Fe\%} + b$			$SSA = a\,MC_{Fe\%} + b$		
Wavelength (nm)	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}	$a \pm \sigma a$	$b \pm \sigma b$	R^2 ; χ^2_{red}
370	$(6.0 \pm 1.4)\,10^{-4}$	$(1.4 \pm 0.7)\,10^{-4}$	0.60 ; 1.5	$(-2.7 \pm 0.4)\,10^{-2}$	(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	$(3.9 \pm 0.8)\,10^{-4}$	$(0.8 \pm 3.9)\,10^{-4}$	0.65 ; 1.6	$(-1.3 \pm 0.2)\,10^{-2}$	(1.01 ± 0.01)	0.72 ; 2.9
590	$(2.5 \pm 0.5)\,10^{-4}$	$(0.5 \pm 2.4)\,10^{-4}$	0.56 ; 1.7	$(-0.8 \pm 0.1)\,10^{-2}$	(1.01 ± 0.01)	0.70 ; 2.4
660	$(2.0 \pm 0.4)\,10^{-4}$	$(0.4 \pm 1.7)\,10^{-4}$	0.48 ; 1.9	$(-0.5 \pm 0.1)\,10^{-2}$	(1.00 ± 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 ± 0.00)	0.54 ; 1.6
950	$(1.4 \pm 0.3)\,10^{-4}$	$(0.3 \pm 1.4)\,10^{-4}$	0.45 ; 2.0	$(-0.3 \pm 0.1)\,10^{-2}$	(1.00 ± 0.00)	0.49 ; 1.5

1241 **Figure 1.** Flowchart illustrating the procedure for data treatment and retrieval of physical and chemical
1242 (size, composition) and spectral optical properties (single scattering albedo, SSA, and complex refrac-
1243 tive index) of mineral dust aerosols. In red we mention the different corrections performed and the
1244 values adopted in the calculations.

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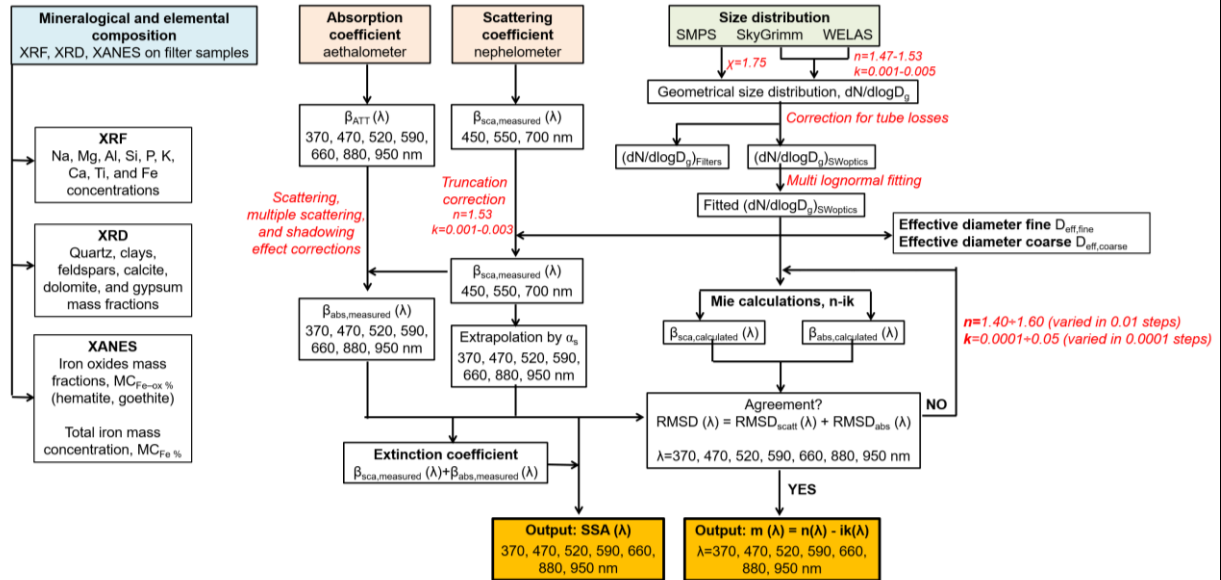
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1249 **Figure 2.** Box and whisker plot showing the full variability of hematite and goethite mass fractions in

1250 the soils for the clay–sized (<2 μm diameter) and silt–sized (<60 μm diameter) fractions as retrieved

1251 from the global soil mineralogical database by Journet et al. (2014). The box and whisker plot include

1252 data for the nine desert source areas depicted in Ginoux et al. (2012) and DB17 (Northern Africa, Sahel,

1253 Eastern Africa and the Middle East, Central Asia, Eastern Asia, North America, South America, South-

1254 ern Africa, and Australia). Dots indicate hematite and goethite content in clay–sized and silt–sized soils

1255 (always from Journet et al.) extracted in correspondence to the geographical coordinates where the

1256 nineteen soils used in the CESAM experiments were collected. The Journet et al. database assumes

1257 that the iron oxides in the silt fraction consist only of goethite.

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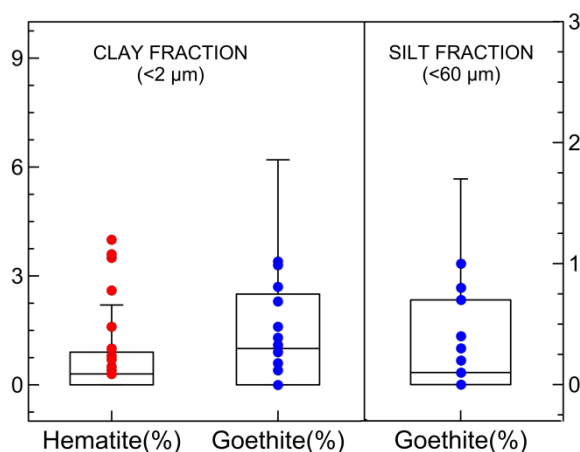
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Figure 3. Top panel: time series of the aerosol mass concentration (cross symbols) and effective fine ($D_{eff,fine}$, open dots) and coarse diameter ($D_{eff,coarse}$, open squares) measured inside the CESAM chamber (red symbols) and at the input of the SW instruments (black symbols) for one experiment (Morocco dust). Bottom panel: time series of the scattering β_{sca} and absorption β_{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.
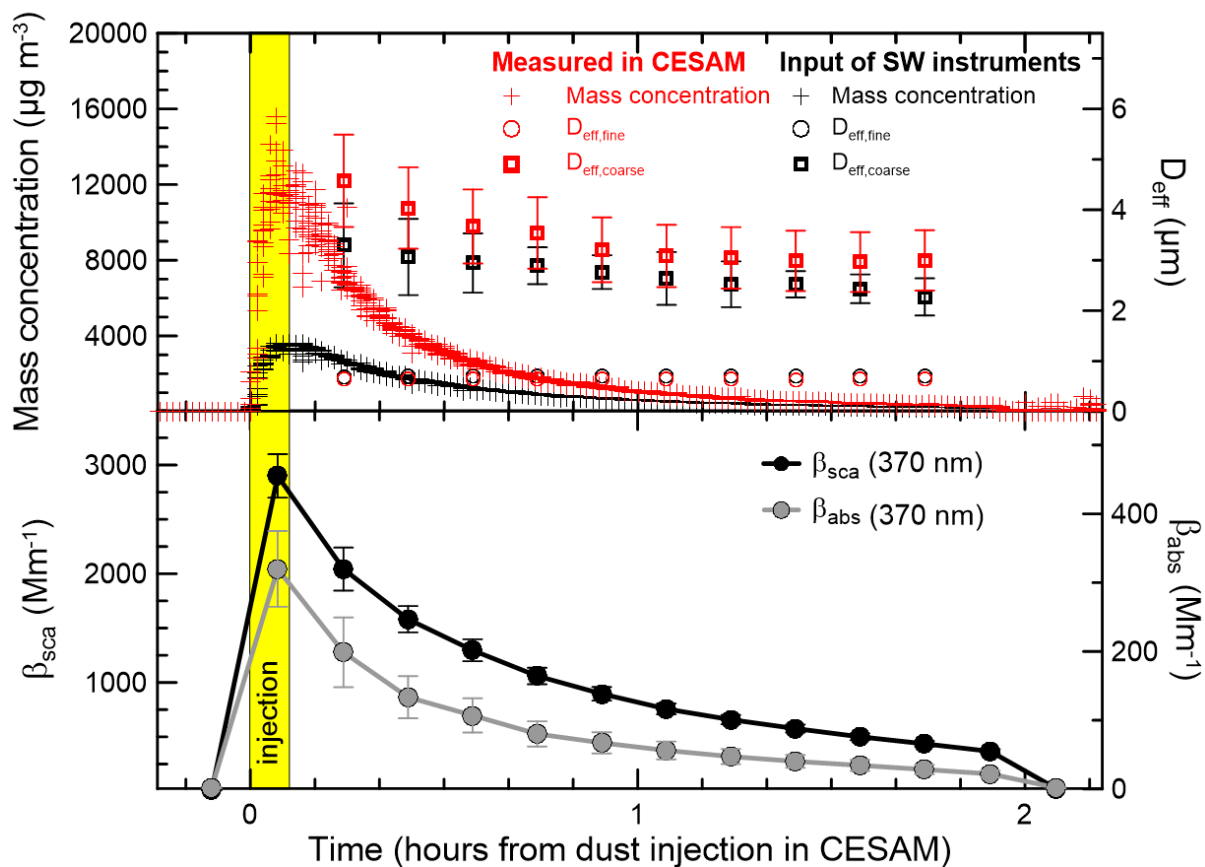
Figure 4. Comparison of dust size distributions sensed by the SW optical instruments (behind the SW instruments inlet $(dV/dlogD_g)_{SWoptics}$), with field data for long–range transported dust. The thick black line represents the mean value of $(dV/dlogD_g)_{SWoptics}$ at the peak of the dust injection in CESAM for experiments with the different samples. The grey shaded area indicates the range of $(dV/dlogD_g)_{SWoptics}$ 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–ATTACK campaigns in Puerto Rico, Caraibes), Müller et al. (2011), Chen et al. (2011) and Ryder et al. (2018) (SAMUM2, NAMMA, and AER–D campaigns in Cape Verde, eastern Atlantic), and Weinzierl et al. (2017) (SALTRACE campaign, data from Barbados). For comparison, data taken close to the source in Niger from Formenti et al. (2011) during the AMMA campaign are also shown. SAL stands for Saharan Air Layer. All data are reported as volume size distributions normalised at the maximum.
(The different acronyms spell out as: AER–D= AERosol Properties – Dust; AMMA = African Monsoon Multidisciplinary Analysis; CLARE= Cooperative LBA Airborne Regional Experiment; DUST–ATTACK+ Dust Aging and TransporT from Africa to the Caribbean; NAMMA = NASA African Monsoon Multidisciplinary Analysis; PRIDE = Puerto Rico Dust Experiment; SALTRACE= Saharan Aerosol Long–range Transport and Aerosol–Cloud–Interaction Experiment; SAMUM = Saharan Mineral Dust Experiment).

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1346 **Figure 6.** Real (*n*) and imaginary (*k*) parts of the dust complex refractive index at seven wavelengths
1347 between 370 and 950 nm obtained for the 19 aerosol samples analyzed in this study. Data correspond
1348 to the time average of the 10 min values obtained between the peak of the injection and 120 min later.
1349 The error bar corresponds to the absolute uncertainty in *n* and *k*, estimated to be <8% for n and between
1350 13 and 75 % for *k*.

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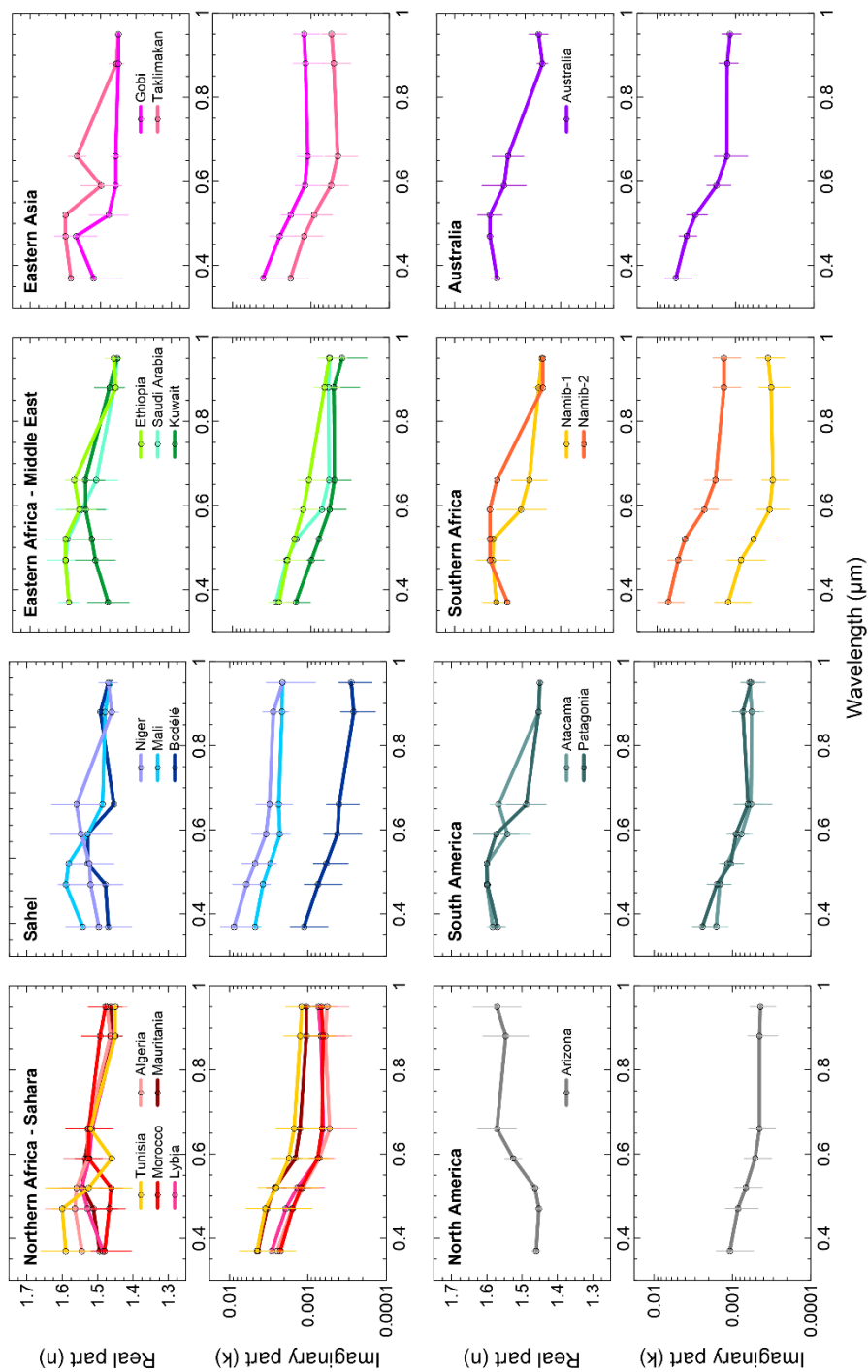
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Figure 7. Single scattering albedo (SSA) at seven wavelengths between 370 and 950 nm obtained for the 19 aerosol samples analyzed in this study. Data correspond for each sample (with the exception of Tunisia and Namib–2, see Sect. 3.1) to the fit of the 10 min values of β_{sca} versus β_{abs}, and the uncertainty is between 1% and 12% at 370 nm and between 1% and 3% at 950 nm.
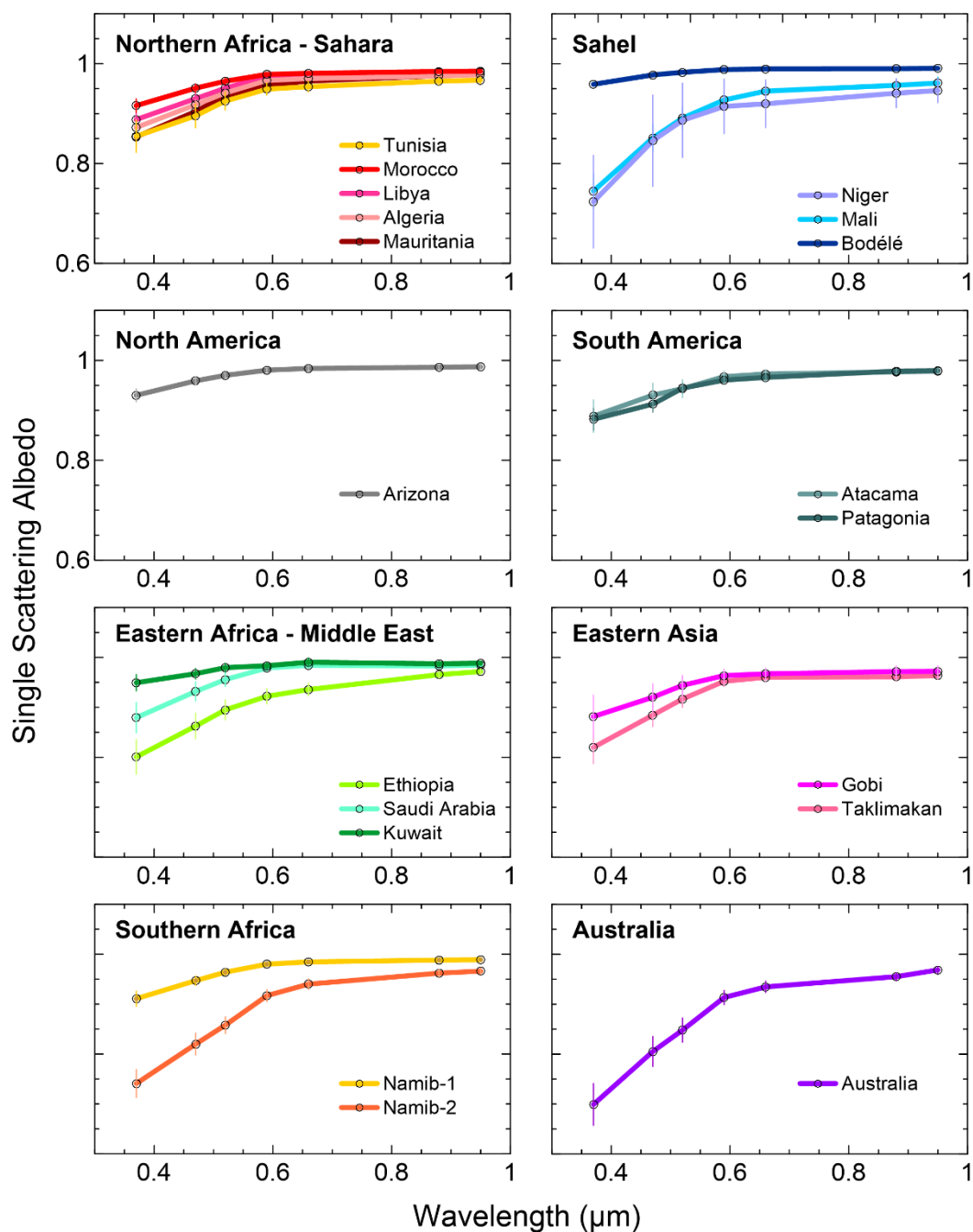
Figure 8. Comparison of results obtained in this study with literature–compiled values of the (a) dust real and imaginary parts of the refractive index (n, k) and (b) single scattering albedo (SSA) in the SW spectral range. The regions in grey indicate the full range of variability obtained in this study, and the black thick lines are the means of n, k and SSA obtained for the different aerosol samples. Literature values include estimates from ground–based and aircraft observations during field campaigns, laboratory studies, AERONET inversions, and estimates from dust mineralogical composition. Data are in some cases for the full dust size distribution, while in other only the fine fraction below about 2 μm is represented (identified with *).

The main provenance of the dust and datasets from the literature is provided in the following: Volz et al. (1972) is data for rainout dust collected in Germany; Patterson et al. (1977) is Saharan dust; Hess 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 Arabia AERONET stations; Haywood et al. (2003) is dust from Mauritania; Sinyuk et al. (2003) is data from Cape Verde, Dakar, and Burkina Faso; Clarke et al. (2004) is Asian dust offshore of China, Japan, and Korea; Linke et al. (2006)–A is dust from Cairo; Linke et al. (2006)–B is dust from Morocco; Balkanski et al (2007) is calculated from mineralogical composition assuming a 1.5% hematite mass fraction in dust; Todd et al. (2007) is from Bodélé; Osborne et al. (2008) is from Niger; Otto et al. (2009), Petzold et al. (2009), Schladitz et al. (2009), and Muller et al. (2010, 2011) is dust originated mostly in Morocco; McConnell et al. (2008, 2010) is dust from Niger/Senegal; Chen et al. (2011) is dust from Western 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 Layer, –B is from Bodélé/Sudan, and –C is a Sahelian uplift episode; Johnson et al. (2011) is dust from Western Sahara; Moosmüller et al. (2012) analysed samples from Middle East, Mali and Spain, and here we report the average of their obtained values; Wagner et al. (2012) obtained k values for several samples from Burkina Faso, Cairo and the SAMUM campaign and here we report the values for the maximum of their spectral k (Burkina Faso) and the minimum (Cairo); Ryder et al. (2013) is dust from Western Sahara and Mauritania and we report in both k and SSA plots the average of their observations; Engelbrecht et al. (2016) analysed many dust samples from all over the world, here we report their estimated minimum and maximum of the dust SSA that are –A from California and –B from the Etosha Pan in Namibia; Stegmann and Yang (2017) modelled the refractive index of dust based on assumed mineralogical compositions typical for Northern and Southern Sahara and Western and Eastern Asia dust, and here we report the average of their results for both n and k. Uncertainties in the field observations have been omitted for the sake of clarity. The legend identifies the line styles used in the plots.

(The different acronyms spell out as (see also the caption of Fig. 4): AERONET = Aerosol Robotic Network; OPAC = Optical Properties of Aerosols and Clouds; SHADE = Saharan Dust Experiment; BODEX = The Bodélé Dust Experiment; DABEX = Dust and Biomass Experiment; SAMUM1 and SAMUM2 refers to the two SAMUM campaigns in Morocco and Cape Verde, respectively, SAMUM = Saharan Mineral Dust Experiment; DODO = Dust Outflow and Deposition to the Ocean; ACE–Asia =

Asian Pacific Regional Aerosol Characterization Experiment; GERBILS = Geostationary Earth Radia-

1405 *tion Budget Intercomparison of Longwave and Shortwave radiation).*

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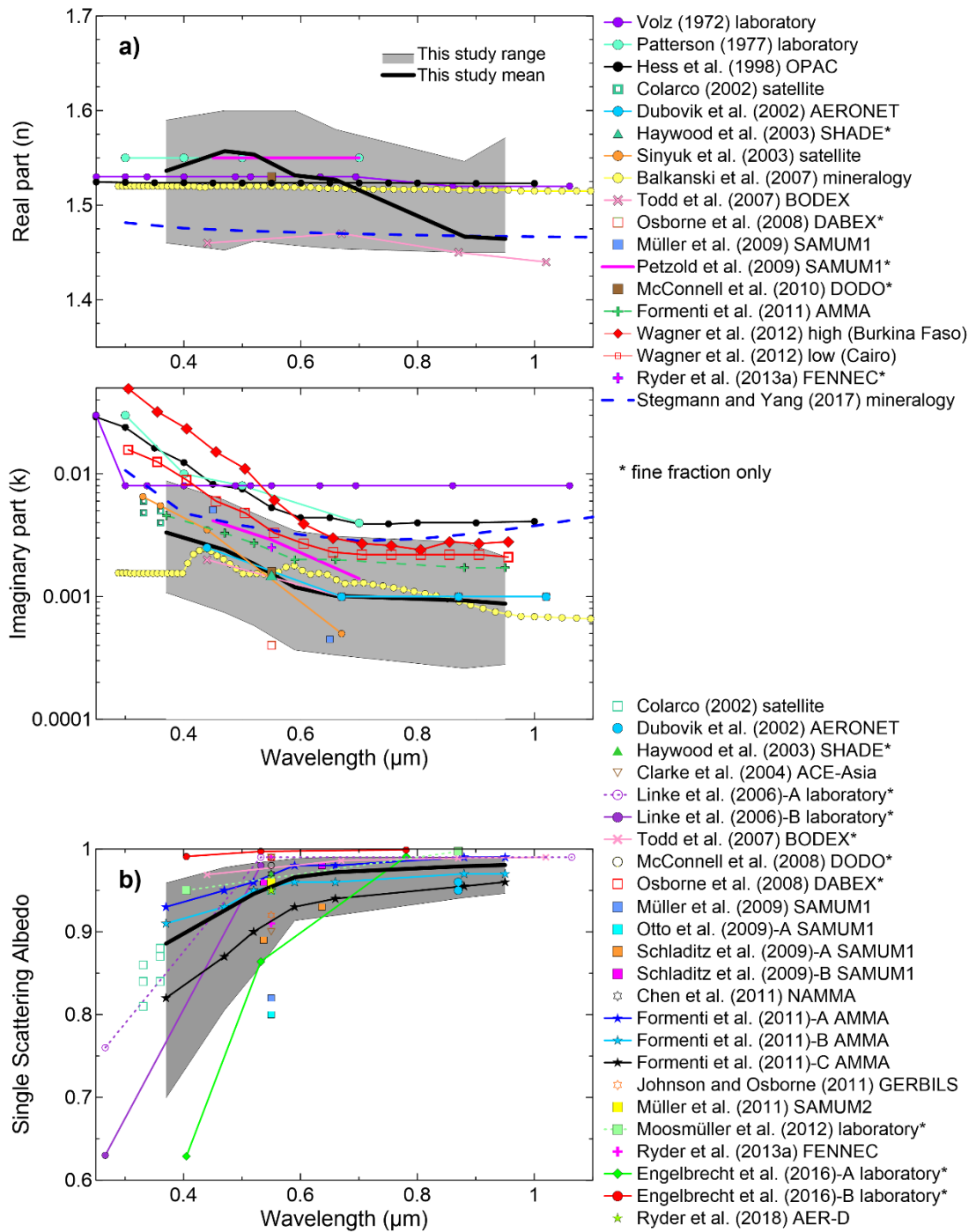
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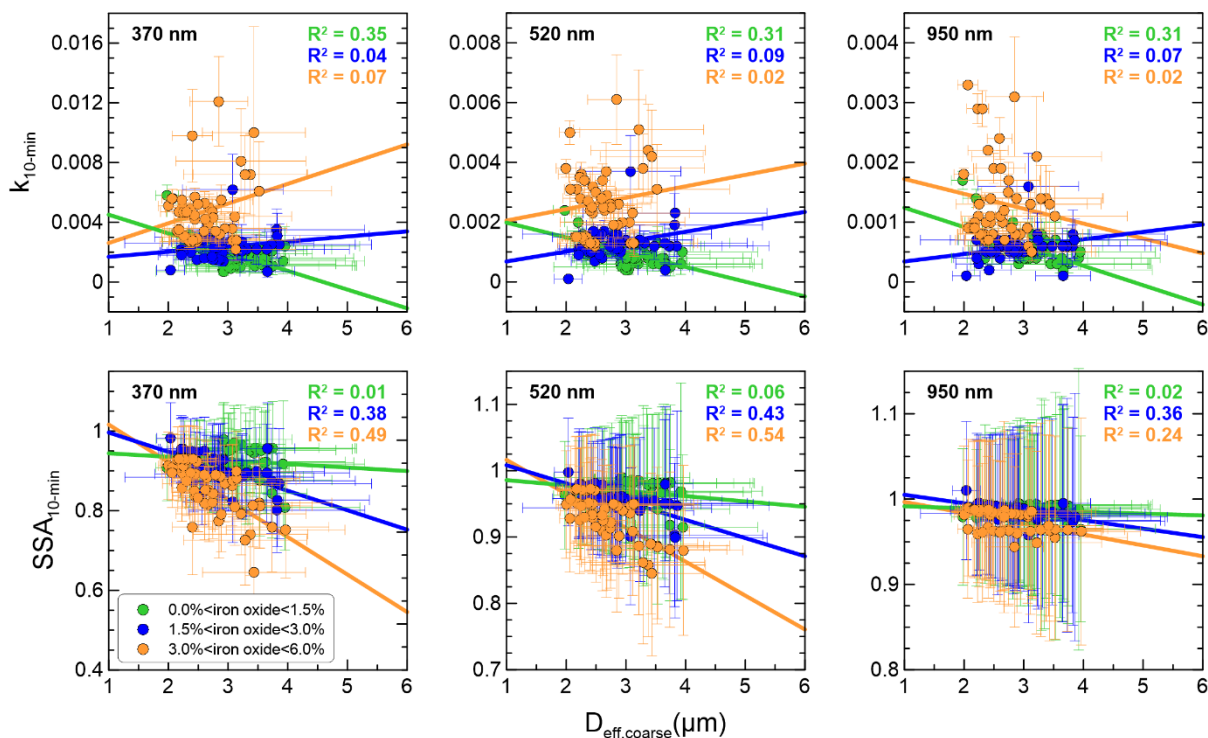
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Figure 9. Experiment–averaged imaginary part of the refractive index (k, top panels) and single scattering albedo (SSA, bottom panels) at 370, 520, and 950 nm versus the mass concentration of iron oxides (MC$_{Fe–ox\%}$), hematite (MC$_{Hem\%}$), goethite (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.
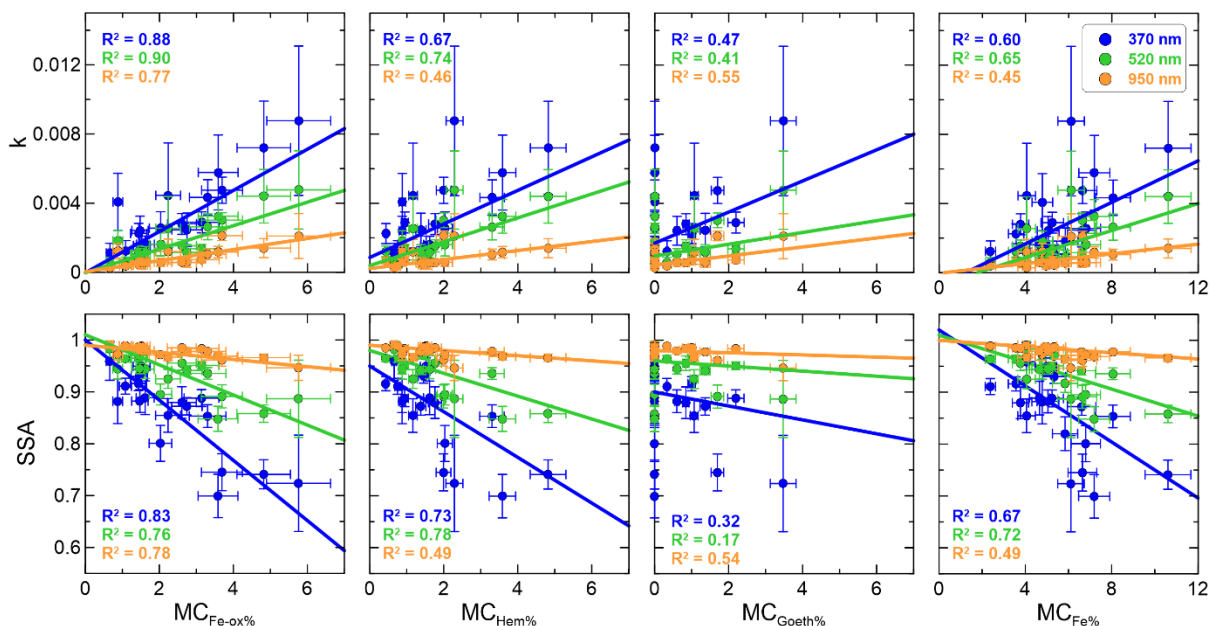
Figure 10. 10–min averaged imaginary refractive index (k_{10-min}, top panels) and single scattering albedo (SSA$_{10-min}$, bottom panels) at 370, 520, and 950 nm versus effective coarse diameter ($D_{eff,coarse}$) estimated at the input of the SW instruments. Data were classified in three classes based 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.