Dear editor,

We wish to thank you, the two anonymous referees, and Dr H. Moosmüller for useful comments on the manuscript. We have carefully revised the text to improve the clarity of the reading.

In particular we have made a small change to the paper title from

"Spectral- and size-resolved mass absorption efficiency of mineral dust aerosols in the shortwave: a simulation chamber study"

to

"Spectral- and size-resolved mass absorption efficiency of mineral dust aerosols in the shortwave spectrum: a simulation chamber study"

The detailed answers to the two anonymous referees are presented

Anonymous Referee #1

The paper presents needed results on dust optical properties (mass absorption efficiency and absorption Angstrom exponent) for D<2.5um and D<10um particles, relating them to chemical composition. The samples analyzed are from 12 locations in northern Africa (5 samples), Namibia (1 sample), northern China (1 sample), the Middle East (2 samples), North America (1 sample), South America (1 sample) and Australia (1 sample). I have no substantial issues with the analysis or the paper. I recommend publication after addressing the minor points below:

1) In the Abstract and somewhere earlier in the paper (before the Results section) the location where the samples were collected from should be presented. Some information about sample collection is also needed – i.e.: a) Was only one sample collected at each location, or were multiple samples collected then combined? b) Were the samples collected from locations know to be preferential sources for atmospheric dust or was the collection location just random? The latter point is important, since it's know that atmospheric dust comes from preferential locations.

To address this point, the sentence in the result section has been rewritten as "The selection of these soils and sediments was made out of 137 individual top-soil samples collected in major arid and semi-arid regions worldwide and representing the mineralogical diversity of the soil composition at the global scale. As discussed in Di Biagio et al. (2017), this large sample set was reduced by a set of 19 samples

their availability in sufficient quantities for injection in the chamber. Because some of the experiments did not produce enough dust to perform good-quality optical measurements, , in this paper we present a set of twelve samples distributed worldwide but mostly in Northern and Western Africa (Libya, Algeria, Mali, Bodélé) and the Middle East (Saudi Arabia and Kuwait). Individual samples from the Gobi desert in Eastern Asia, the Namib Desert, the Strzelecki desert in Australia, the Patagonian deserts in South America, and the Sonoran Desert in Arizona have also been investigated."

The first sentence of the abstract has been changed as "This paper presents new laboratory measurements of the mass absorption efficiency (MAE) between 375 and 850 nm for twelve individual samples of mineral dust from different source areas worldwide and in two size classes".

2) Abstract, pg. 2, lines 33-34: "The size-independence of AAE suggests that, for a given size distribution, the possible variation of dust composition with size would not affect significantly the spectral behavior of shortwave absorption." Either that or the composition simply DIDN'T vary with size for this set of samples – so I'd reword this a bit.

This correction has been accepted

3) pg. 2, line 39: Need to spell out AAOD this first time that you use it.

This correction has been accepted

4) pg. 2, lines 40-41: "which is relevant to the development of remote sensing of light-absorption aerosols from space. " Not only from space! This approach has also been used extensively for AERONET (surface-based remote sensing) AAOD attribution.

The reviewer is right, the sentence has been rewritten as ", which is relevant to the development of remote sensing of light-absorption aerosols from space, and their assimilation in climate models."

5) pg. 3 line 56 contains a partial sentence ("In the solar spectrum (Boucher et al., 2013)").

This was reworded as "Albeit partially compensated by the radiative effect in the thermal infrared, the global mean radiative effect of mineral dust in the shortwave is negative both at the surface and the top of the atmosphere (TOA) and local warming of the atmosphere (Boucher et al., 2013)."

6) pg. 6, lines 162-163: “The total uncertainty, including the effects of photon counting and the deposit inhomogeneity, on the absorption coefficient measurement is estimated at 8%.” Is there a basis/reference for this?

The references to the papers of Petzold et al. (2004) and Massabo’ et al. (2013) have been added to the text.

7) pg. 7, line 170: It would be useful to give typical total masses and/or the fractional error/uncertainty in total aerosol gravimetric mass based on this error in filter mass.

This has been added in lines 173-175.

8) pg. 8, line 223 equation: When I read this my immediate question was: “How does this compare to the measure of total gravimetric mass?”. You answer this question (appropriately) in the results section but I still think it would be useful to add a note here pointing to the fact the in your results discussion you found this agreed well with the total gravimetric mass.

The following sentence has been added “As it will be explained in the result section (paragraph 3.1), the values of MCdust estimated from Equation 4 were found in excellent agreement with the measured gravimetric mass on the filters”

9) pg. 10: Do you have any estimate / sense of how well the dust suspended by this “shaking” compares to the dust lofted by winds?

Prior to any scientific analysis, we have dedicated a lot of energy to investigate the realism of our dust generation system, both in terms of the composition and the size distribution of the dust aerosols. These results are reported in two papers:

- Di Biagio, C., P. Formenti, S. A. Styler, E. Pangui, and J.-F. Doussin (2014), Laboratory chamber measurements of the longwave extinction spectra and complex refractive indices of African and Asian mineral dusts, Geophys. Res. Lett., 41, doi:10.1002/2014GL060213. – Figure 2 and discussion

But in particular in

- Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Caquineau, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Global scale variability of the mineral dust long-wave refractive index: a new dataset of in situ measurements for climate modeling and remote sensing, Atmos. Chem. Phys., 17, 1901-1929, doi:10.5194/acp-17-1901-2017, 2017

where we have dedicated two paragraphs (5.1. Atmospheric representativity: mineralogical composition and 5.2 Atmospheric representativity: size distribution) to show how the composition and the size distribution of the generated dust are well representative of those of real dust in the atmosphere, which makes the laboratory experiments well suited for studying the dust optical properties.

In order to stress this point further, without repeating results already presented in these two publications, we have added the following sentence in paragraph 3 “Di Biagio et al. (2014; 2017) have demonstrated the realism of the generation system concerning the composition and the size distribution of the generated dust with respect to the properties of mineral dust in the atmosphere”.

10) pg. 11, line 209: Mauritania is not listed as one of the sample site locations in Table 3. ?

For Mauritania, we only have chemical composition but not optical measurement results. The reference to this sample has been taken out of the paper.

11) pg. 11, lines 309-310 (an onward): The results here are said to agree well with that found for atmospheric aerosols in other studies, but the values in these studies is not given so this feels very handwaving and unconvincing. Are you referring to the values given in Table 5? If so, please refer directly to them. If not, the comparison here needs to be more quantitative (discuss numbers from the literature vs. what is found here).

Additional text and values have been added in lines 321-392 to address this point

12) pg. 13, lines 353-354: If you have results for Niger why not show them?

We do not have optical results for Niger

13) pg. 14, lines 387-388: MAE doesn’t vary linearly inversely with wavelength, it varies linearly inversely with the log of the wavelength (hence our ability to use the AAE relationship).

The reviewer is correct, this has been corrected as “and decrease in a linear way with the logarithm of the wavelength”

14) pg. 15, lines 429-434: A few things: a) “satisfactory” and “loose” are not quantitative terms, nor are they really appropriate for a scientific paper. What constitutes “satisfactory”? Best is to just give the correlations. b) The high correlation coefficients for PM2.5 are really driven by one high data point and so are probably not very robust.

These sentences have been reworded as “Regardless of the size fraction, the correlation between the MAE values and the percent mass of total elemental iron are higher at 375, 407 and 532 nm” and “At these wavelengths, linear correlations with the mass fraction of iron oxides are low in the $PM_{10.6}$ mass fraction ($R^2$ up to 0.38-0.62), but higher in the $PM_{2.5}$ fraction ($R^2$ up to 0.83-0.99)”

15) pg. 16, lines 455-456: How can fine-mode-only AAOD be GREATER than total aerosol AAOD?

Total and fine were inverted by mistake, this is now corrected

16) pg. 17, lines 487-488: As written this implies Solomon et al. varied SSA by 5%.

For, say, SSA of 0.9, that they varied SSA by 0.045. That is, the co-albedo (or absorption) was varied by 45% (0.1+/-0.045). This is the proper comparison to make to the variation in MAE that you calculate

The reviewer is right. The sentence has been corrected as “As a comparison, Solmon et al. (2008) showed that varying the single scattering albedo of mineral dust over western Africa by ± 5%, that is, varying the co-albedo (or absorption) by 45% (0.1± 0.045) could drastically change the climate response in the region.”

7) Overall: Some editing is needed for language throughout. Here I list some that stood out to me – all small stuff but editing would help readability:

pg. 3, line 67-68: “in the last ten years or so” (too casual for scientific writing)

removed

pg. 3, line 74: “A significant body of observations have been performed.”

Replaced by “A significant number of observations have quantified”

pg. 8, lines 214-215: “The linear deconvolution, performed the Athena IFEFFIT free-ware analysis program (Ravel and Newville, 2005), provided with the proportionality factors alpha_i representing the mass fraction of elemental iron to be assigned to the i-th standard mineral.” (I found this sentence nearly impossible to follow)

Replaced by “The linear deconvolution has been performed with the Athena IFEFFIT freeware analysis program (Ravel and Newville, 2005). This provided with the proportionality factors □i representing the mass fraction of elemental iron to be assigned to the i-th standard mineral.”

pg. 10, line 253: “the chamber was evacuated by to” (delete “by”)

Corrected

pg. 10, line 259: “dust particles produced was” –> “dust particles produced were”

The sentence was corrected as “The dust particles produced by the mechanical shaking, mimicking the saltation processing that soils experience when eroded by strong winds, were injected in the chamber by flushing the flask with N2 at 10 L min-1 for about 10-15 min, whilst continuing shaking the soil.”

pg. 10, lines 279 “dust on filter for” –> “dust on the filter for”

Corrected as “dust on the filter membranes for subsequent chemical analysis”

pg. 10, line 280: “by placing the loaded filter holders”: This reads as if you are placing LOADED FILTERS (vs holders with blank filters in them, which is what I assume you mean). Reword.

Corrected as “by placing the filter holders loaded with filter membranes”

pg. 11, line 206: “the origin of used dust samples”. I think this should be “the origin of our dust samples”, yes?

Corrected as “the origin of dust samples”

176

177 pg. 12, line 315: “Henceforth, and contrary to the soil samples”

178 pg. 12, lines 317-318: “could reflect more that of the parent sedimentary soil than not the other samples.”

179 These two sentences were rewritten as “This powder is likely to be injected in the chamber with little or
180 no size fractionation. Henceforth, the aerosol generated from it should have a closer composition to the
181 original powder than the other samples.

182

183 pg. 12, line 335: “An the exception”

184 Corrected as “Exceptions are”

185

186 pg. 13, lines 353-354: “As a matter of fact, the number fraction of particles in the size classes above
187 0.5 μm in diameter are different in the dust aerosol generated in the Alfaro et al. (2004) study with
188 respect to ours.”

189 Corrected as “As a matter of fact, the number fraction of particles in the size classes above 0.5 μm in
190 diameter is different in the dust aerosol generated in the Alfaro et al. (2004) study with respect to ours.”

191

192 pg. 13, line 364: “On the contrary” –> “In contrast

193 Corrected

194

195 pg. 13, line 365: “These differences could yield either to difference in the”

196 Corrected as “These differences could either be due to difference in the chemical composition and/or
197 in the total mass in the denominator of Equation 8.”

198

199 pg. 15, lines 415-417: “using a power-law function fit as from Equation 2, provides withthe values of”

200 Corrected as “using the power-law function fit (Equation 2)”

201

202 pg. 16, line 448: “The size-dependence, yielding significantly higher MAE values”

Corrected as “The size-dependence, yielding significantly higher MAE values in the fine fraction (PM2.5) than in the bulk (PM10.6) aerosol,”

pg. 17, line 470: “A closer look to observations” –> “A closer look at observations”

Corrected as “These differences could either be due to difference in the chemical composition and/or in the total mass in the denominator of Equation 8.”

pg. 17, line 478: “our estimated MAE average at” –> “our average MAE values are”

Corrected

pg. 17, line 490: “As Moosmuller et al.” –> “As in Moosmuller et al.”

Corrected

pg. 18, line 503-504: “pointing out to the” –> “pointing out the

Corrected

Anonymous Referee #2

GENERAL COMMENT

The manuscript presents important results from a carefully designed and conducted study on the light-absorbing properties of mineral dust from various origins. Dust samples collected at the different source regions have been re-suspended in an aerosol chamber and characterized with respect to microphysical, optical and chemical properties by state of the art methods. The study provides urgently needed knowledge on the multi-spectral light absorbing properties of mineral dust and for sure deserves publication in ACP. The manuscript is well structured, the methods are described in necessary detail and the referenced literature reflects the current state of knowledge. I recommend publication after the following minor revisions have been considered.

SPECIFIC REMARKS

1. In the experimental protocol section, the potential impact of gravitational settling on the re-suspended fraction of the dust samples is mentioned.

Given the instrumentation list, the size distributions of the airborne dust samples were monitored during the runs of the experiments. It appears obvious to control the change of the size distribution during the experiment time in the chamber. Since the mass concentration in the chamber decreased very rapidly after injection, whereas the chemical composition of the dust samples was determined from bulk samples, it would be important to know if the airborne fraction sampled for the determination of optical properties features the same chemical properties as the bulk samples. At least a discussion of this potential source of uncertainties should be presented, along with a plot showing the change of the size distributions during the experiment time. The current analysis starts from the assumption that the dust bulk properties represent also the properties of the sampled airborne fractions. However, is this really justified?

The reviewer is right when saying that the samples collected for the investigation of the chemical composition are time-integrated and henceforth might reproduce dust with varying size distributions. Examples of the time variability of the size distributions are provided by Di Biagio et al. (2017) - Figures 7 and 5S in the supplementary material. These figures show that, after the very strong initial depletion of particles larger than 10 μm in diameter (when no sampling on filters was performed), the number concentration decreases at a rate, which is almost independent of size, suggesting that no significant distortion of the particle size distribution occurs after the most significant removal at the beginning of the experiment.

We also would like to stress that our generation system allows to generate a dust aerosol from a soil, and that this dust aerosol is injected in the chamber, not the soil. Henceforth, when talking about ”bulk” we refer to the total aerosol fraction, sampled from the chamber without any size segregation other than that imposed by the cutoff the sampling lines. The fine fraction corresponds to the same dust aerosols, but sampled by an impactor with a 2.5 diameter cutoff.

2. In section 3.2, the variability of dust optical properties with particle size is discussed. The authors found no statistically significant size-dependence of the absorption Ångström exponent (AAE), whereas the absolute values of the mass absorption efficiencies (MAE) show large differeces between the PM2.5 and PM10 fractions with larger values for the fine mode fraction.

These findings imply that the relative chemical composition with respect to light-absorbing compounds does not change between the size fractions (similar AAE values), whereas the differences between the MAC values indicate that coarse mode particles contain more non-absorbing matter than fine mode particles (higher MAE values for smaller particles). This however, this is in contrast to the assumption that the chemical composition is uniformly distributed across the particle size distribution. Here, a detailed discussion is requested.

The reviewer's statement is in error as for a given mineral composition (given effective complex refractive index) the MAE depends strongly on size, decreasing with size at larger sizes. So having a smaller MAE for PM10 than for PM2.5, does not necessarily imply that PM10 contains less absorbing matter than PM2.5, but may just be due to the change in size distribution. This is shown by our calculations. An independent example of the size dependence is also given by Fig. 1 of Moosmuller et al. (2009).

Moosmüller, H., R. K. Chakrabarty, and W. P. Arnott (2009), Aerosol light absorption and its measurement: A review, Journal of Quantitative Spectroscopy and Radiative Transfer, 110(11), 844-878.

3. In section 4, it is discussed that the potential impact of light absorption by mineral dust may play an important role even after long range transport. Cited studies all refer to observations in China.

However, there is another detailed study on this effect available for the pollution plume of Dakar mixing with mineral dust which also includes the variation of the AAE during mixing

(Petzold et al., 2011). The authors may consider including this study.

This reference has been added to the manuscript.

MINOR COMMENTS

1. The list of references contains various references which are not cited in the manuscript. This should be checked, I found the following but there may be more: Anderson et al., 1998, Andrews et al., 2006, Arnott et al., 2005, Collaud Coen et al., 2010, Petzold et al., 2013.

Corrected

2. Line 56: The sentence seems to be incomplete.

The sentence was corrected as “Albeit partially compensated by the radiative effect in the thermal infrared, the global mean radiative effect of mineral dust in the shortwave is negative both at the surface and the top of the atmosphere (TOA) and local warming of the atmosphere (Boucher et al., 2013).”

3. Line 97 – 98: The basic unit of mass concentrations is gm-3. Using this unit, then the unit of the combined property MAE is m2 g-1 as stated. In its current version this link is not clearly visible.

4. Line 102: The sentence seems to be incomplete.

The sentence was removed

5. Line 163: A reference for the uncertainty of the MWAA is required.

This has been added

6. Line 912: Please check for correct reference, there is no reference Petzold et al. (2008) in the list of references.

Corrected

7. In Figure 4, regression lines may be shown as full line to improve their visibility.

Done

TYPOS

1. Line 108: It should read: “absorption Ångström exponent”.

Corrected

2. Line 138: Skip “with”.

Corrected

3. Line 165: It should read “deposited on a filter ...”.

Corrected

4. Line 245: It should read: “the uncertainty of values ...”.

Corrected

5. Line 253: Skip “by”.

Corrected

6. Line 338: It should read: “PM2.5 fraction”.

Corrected

7. Line 426 –427: I assume the Figures 4 are referenced here.

Corrected

REFERENCES

Petzold, A., Veira, A., Mund, S., Esselborn, M., Kiemle, C., Weinzierl, B., Hamburger, T., Ehret, G., Lieke, K., and Kandler, K.: Mixing of mineral dust with urban pollution aerosol over Dakar (Senegal): impact on dust physico-chemical and radiative properties, Tellus, 63B, 619-634, doi: 10.1111/j.1600-0889.2011.00547.x, 2011.

# Spectral- and size-resolved mass absorption efficiency of mineral dust aerosols in the shortwave spectrum: a simulation chamber study

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## Abstract

This paper presents new laboratory measurements of the mass absorption efficiency (MAE) between 375 and 850 nm for twelve individual samples of mineral dust from different source areas worldwide and ~~in of different origin~~ in two size classes: $PM_{10.6}$ (mass fraction of particles of aerodynamic diameter lower than 10.6 µm) and $PM_{2.5}$ (mass fraction of particles of aerodynamic diameter lower than 2.5 µm). ~~E~~The experiments ~~have been~~were performed in the CESAM simulation chamber using ~~generated~~ mineral dust generated from natural parent soils~~,~~ and included optical and gravimetric analyses.

The ~~R~~results show that the MAE values are lower for the $PM_{10.6}$ mass fraction (range 37-135 $10^{-3}$ $m^2$ $g^{-1}$ at 375 nm) than for the $PM_{2.5}$ (range 95-711 $10^{-3}$ $m^2$ $g^{-1}$ at 375 nm), and decrease with increasing wavelength as $\lambda^{-AAE}$, where the Angstrom Absorption Exponent (AAE) averages between 3.3-3.5, regardless of size. The size ~~-~~independence of AAE suggests that, for a given size distribution, the ~~possible variation of~~ dust composition did not vary with size for this set of samples~~with size would not affect significantly the spectral behavior of shortwave absorption~~. Because of its high atmospheric concentration, light ~~-~~absorption by mineral dust can be competitive ~~to~~ with black and brown carbon even during atmospheric transport over heavy polluted regions, when dust concentrations are significantly lower than at emission. The AAE values of mineral dust are higher than for black carbon (~1), but in the same range as light-absorbing organic (brown) carbon. As a result, depending on the environment, there can be some ambiguity in apportioning the aerosol absorption optical depth (AAOD) based on spectral dependence, which is relevant to the development of remote sensing of light-absorbing~~ption~~ aerosols ~~from space,~~ and their assimilation in climate models. We suggest that the sample-to-sample variability in our dataset of MAE values is related to regional differences ~~of~~ in the mineralogical composition of the parent soils. Particularly in the $PM_{2.5}$ fraction, we found a strong linear correlation between the dust light-absorption properties and elemental iron rather than the iron oxide fraction, which could ease the application and the validation of climate models that now start to include the representation of the dust composition, as well as for remote sensing of dust absorption in the UV-VIS spectral region.


## 1. Introduction

Mineral dust aerosols emitted by wind erosion of arid and semi-arid soils account for about 40% of the total emitted aerosol mass per year at the global scale (Knippertz and Stuut, 2014). The episodic but frequent transport of intense mineral dust plumes is visible from spaceborne sensors, as their high concentrations, combined with ~~to~~ their ability ~~of~~ to scatter~~ing~~ and absorb~~ing~~ solar and thermal radiation,

give r~~a~~ise to the highest registered values of aerosol optical depth (AOD) on Earth (Chiapello, 2014). The instantaneous radiative efficiency of dust particles, that is, their radiative effect per unit AOD, is of the order of ten~~th~~s to hundreds of W $m^{-2}$ $AOD^{-1}$ in the solar spectrum, and of the order ~~of order~~ of ten~~th~~s of W $m^{-2}$ $AOD^{-1}$ in the thermal infrared (e.g., Haywood et al., 2003; di Sarra et al., 2011; Slingo et al., 2006 and the compilation of Highwood and Ryder, 2014). ~~In the solar spectrum, (Boucher et al., 2013).~~ Albeit partially compensated by the radiative effect in the thermal infrared, the global mean radiative effect of mineral dust in the shortwave is negative both at the surface and the top of the atmosphere (TOA) and produces a local warming of the atmosphere (Boucher et al., 2013). ~~Many~~ There are ~~the consequences~~ numerous impacts of dust on ~~the~~ global and regional climate, ~~that~~ which ultimately feed back on wind speed and vegetation and therefore on dust emission (Tegen and Lacis, 1996; Solmon et al., 2008; Pérez et al., 2006; Miller et al., 2014). Dust particles perturb the surface air temperature through their radiative effect at TOA, can increase the atmospheric stability (e.g., Zhao et al. 2011) and might affect precipitation at the global and regional scale (Solmon et al., 2008; Xian, 2008; Vinoj et al., 2014; Miller et al., 2014 and references therein).

All models ~~show~~ indicate that the effect of mineral dust on climate has ~~a~~ great sensitivity to their shortwave absorption properties ~~of mineral dust~~ (Miller et al., 2004; Lau et al., 2009; Loeb and Su, 2010; Ming et al., 2010; Perlwitz and Miller, 2010). Absorption by mineral dust started receiving a great deal of interest ~~in the last ten years or so,~~ when spaceborne and ground-based remote sensing studies (Dubovik et al., 2002; Colarco et al., 2002; Sinyuk et al., 2003) suggested that mineral dust was less absorbing ~~that~~ than~~it~~ had been suggested ~~indicated~~ by in situ observations (e.g., Patterson et al., 1977; Haywood et al., 2001), particularly at wavelengths below 600 nm. Balkanski et al. (2007) showed that lowering the dust absorption properties to an extent that reconciles them both with the remote-sensing observations and the state-of-knowledge of the mineralogical composition, allowed calculating the clear-sky ~~dust~~ shortwave radiative effect of dust in agreement with satellite-based observations. A significant ~~body~~ number of observations has ~~ve been performed in quantify~~quantified~~ing~~ the shortwave light-absorbing properties of mineral dust, by direct measurements (Alfaro et al., 2004; Linke et al., 2006; Osborne et al., 2008; McConnell et al., 2008; Derimian et al., 2008; Yang et al., 2009; Müller et al., 2009; Petzold et al., 2009; Formenti et al., 2011; Moosmüller et al., 2012; Wagner et al., 2012; Ryder al., 2013a; Utry et al., 2015; Denjean et al., 2015c; 2016), and indirectly~~,~~ by quantifying the amount and the speciation of the light-absorbing compounds in mineral dust, principally iron oxides (Lafon et al., 2004; 2006;

Lazaro et al., 2008; Derimian et al., 2008; Zhang et al., 2008; Kandler et al., 2007; 2009; 2011; Formenti et al., 2014a; 2014b).

However, existing data are often limited to a single wavelength, which moreover ~~are~~ is not the same ~~identical~~ for all experiments. Also, frequently they do not represent the possible regional variability of the dust absorption, either because they are obtained from field measurements integrating the contributions of different source regions, or conversely, by laboratory investigations targeting samples from a limited number of locations. This might lead to biases in the data. Indeed, iron oxides in mineral dust, mostly in the form of hematite ($Fe_2O_3$) and goethite (Fe(O)OH), have specific absorption bands in the UV-VIS spectrum (Bédidi and Cervelle, 1993), and have a variable content depending on the soil mineralogy of the source regions (Journet et al., 2014).

~~Henceforth, i~~In this study, e~~E~~xperiments on twelve aerosol samples generated from natural parent top soils from various source regions worldwide ~~have been~~were conducted with a large atmospheric simulation chamber. ~~we~~ We present a new evaluation of the ultraviolet to near-infrared (375-850 nm) light-absorbing properties of mineral dust by ~~studying~~ investigating the size-segregated mass absorption efficiency (MAE, units of $m^2\ g^{-1}$) and its spectral dependence, ~~largely~~ widely used in climate models to calculate the direct radiative effect of aerosols. ~~Experiments on twelve aerosol samples generated from natural parent top soils from various source regions worldwide have been conducted with a large atmospheric simulation chamber.~~

## 2. Instruments and methods

At a given wavelength, λ, the mass absorption efficiency (MAE, units of $m^2\ g^{-1}$) is defined as the ratio of the aerosol light-absorption coefficient $b_{abs}(\lambda)$ (units of $m^{-1}$)~~,~~ and its mass concentration (in $\mu g\ m^{-3}$)

$$MAE(\lambda) = \frac{b_{abs}(\lambda)}{Mass\ Conc} \quad (1)$$

~~MAE values for mineral dust aerosol are expressed in.~~ MAE values for mineral dust aerosol are expressed in $10^{-3}\ m^2\ g^{-1}$.

The spectral dependence of the aerosol absorption coefficient $b_{abs}(\lambda)$ is described by the power-law relationship

$$b_{abs}(\lambda) \sim \lambda^{-AAE} \quad (2)$$

where the AAE is the Absorption Ångström ~~Absorption~~ Exponent, representing the negative slope of $b_{abs}(\lambda)$ in a log-log plot (Moosmüller et al., 2009)

$$AAE = -\frac{dln(b_{abs}(\lambda))}{dln(\lambda)} \quad (3)$$

## 2.1. The CESAM simulation chamber

The e~~E~~xperiments in this work have been performed in the 4.2 $m^3$ stainless-steel CESAM (French acronym for Experimental Multiphasic Atmospheric Simulation Chamber) simulation chamber (Wang et al., 2011). The CESAM chamber has been extensively used in recent years to simulate, at sub and supersaturated conditions, the formation and properties of aerosols at concentration levels comparable to those encountered in the atmosphere (Denjean et al., 2015a; 2015b; Br~~é~~égonzio-Rozier et al., 2015; 2016; Di Biagio et al., 2014; 2017~~6~~).

CESAM is a multi-instrumented platform, equipped with twelve circular flanges to support its analytical environment. Basic instrumentation comprises sensors to measure the temperature, pressure and relative humidity within the chamber (two manometers MKS Baratrons (MKS, 622A and MKS, 626A) and a HMP234 Vaisala® humidity and temperature sensor). The particle size distribution is routinely measured by a combination of (i) a scanning mobility particle sizer (SMPS, mobility diameter range 0.02–0.88 µm), composed of a Differential Mobility Analyzer (DMA, TSI Inc. Model 3080) and a Condensation Particle Counter (CPC, TSI Inc. Model 3772); (ii) a SkyGrimm optical particle counter (Grimm Inc., model 1.129, optical equivalent diameter range 0.25–32 µm); and (iii) a WELAS optical particle counter (PALAS, model 2000, optical equivalent diameter range 0.5–47 µm). Full details of operations and data treatment of the particle counters are provided in Di Biagio et al. (~~2016~~2017).

## 2.2. Filter sampling

Three filter samples per top soil sample were collected on different types of substrate based on the analysis to be performed. Sampling dedicated to the determination of the aerosol mass concentration by

gravimetric analysis and the measurement of the absorption coefficients by optical analysis was performed on 47-mm quartz membranes (Pall Tissuquartz™, 2500 QAT-UP). Two samples were collected in parallel. The first quartz membrane sample ("total") was collected without a dedicated size cut-off using an in-house built stainless steel sampler operated at 5 L $min^{-1}$. However, as detailed in Di Biagio et al. (~~2016~~2017), the length of the sampling line from the intake point in the chamber to the filter entrance was 50 cm, ~~yielding~~ resulting in ~~with~~ a 50% cut-off of the transmission efficiency at 10.6 µm ~~in~~ particle aerodynamic diameter. This fraction is therefore indicated as $PM_{10.6}$ in the ~~forthcoming~~ following discussion. The second quartz membrane sample was collected using a 4-stage DEKATI impactor operated at ~~the~~ a flow rate of 10 L $min^{-1}$ to select the aerosol fraction of particles with aerodynamic diameter smaller than 2.5 µm, indicated as $PM_{2.5}$ ~~here forth~~. Sampling for the analysis of the iron oxide content was performed on polycarbonate filters (47-mm Nuclepore, Whatman; pore size ~~of~~ 0.4 µm) using the same sample holder ~~than~~ as used for the total quartz filters, and therefore ~~referring~~ corresponding to the $PM_{10.6}$ mass fraction. Samples were collected at a flow rate of 6 L $min^{-1}$. All flow rates were monitored by a thermal mass flow meter (TSI Inc., model 4140). These samples were also used to determine the elemental composition (including Fe) and the fraction of iron oxides in the total mass.

### 2.3. The Multi-Wavelength Absorbance Analyzer (MWAA)

The aerosol absorption coefficient, $b_{abs}(\lambda)$, at 5 wavelengths ($\lambda$ = 375, 407, 532, 635, and 850 nm) was measured by *in situ* analysis of the quartz filter samples using the Multi-Wavelength Absorbance Analyzer (MWAA), described in detail in Massabò et al. (2013; 2015).

The MWAA performs a non-destructive scan of the quartz filters ~~on~~ at 64 different points, each ~ 1 $mm^2$ wide. It measures the light transmission through the filter as well as backscattering at two different angles (125° and 165°). This is necessary to constrain the multiple scattering effects occurring within the particle-filter system. The m~~M~~easurements are used as input ~~of~~ to a radiative transfer model (Hänel, 1987; 1994) as implemented by Petzold and Schönlinner (2004) for the Multi-Angle Absorption Photometry (MAAP) measurements. In this model, a two stream approximation is applied (Coakley and Chylek, 1975), in which the fractions of hemispherical backscattered radiation with respect to the total scattering for collimated and diffuse incident radiation are approximated on the basis of the Henyey-Greenstein scattering phase function (Hänel, 1987). This approximation assumes a wavelength-independent asymmetry parameter (g) set to 0.75, appropriate for mineral dust (Formenti et al., 2011; Ryder et al., 2013b).

The total uncertainty, including the effects of photon counting and the deposit inhomogeneity, on the absorption coefficient measurement is estimated at 8% (Petzold et al., 2004; Massabò et al., (2013).

### 2.4. Gravimetric analysis

The aerosol mass deposited on the filters (µg) was obtained by weighing the quartz filter before and after sampling, after a period of 48 hours of conditioning in a room with controlled atmospheric conditions (temperature, T ~ 20 ± 1 °C; relative humidity, RH ~ 50 ± 5%). Weighting is performed with an analytical balance (Sartorius model MC5, precision of 1 µg), and repeated three times to control the statistical variability of the measurement. Electrostatic effects are removed by exposing the filters, prior weighing, to a de-ionizer. The error on in the measured mass is estimated at 10 µg, including the repetition variability. The aerosol mass concentration (µg $m^{-3}$) is obtained by dividing the mass deposited on the filter to the total volume of sampled air ($m^3$) obtained from the mass flowmeter measurements (+-5%). The percent error on mass concentrations is estimated to 5%.

### 2.5. Dust composition measurements

#### 2.5.1. Elemental composition

Elemental concentrations for the major constituents of mineral dust (Na, Mg, Al, Si, P, S, Cl, K, Ca, Fe, Ti, Mn) were obtained by a Wavelength Dispersive X-ray fluorescence Fluorescence (WD-XRF) of the Nuclepore filters using a PW-2404 spectrometer by Panalytical. Excitation X-rays are produced by a Coolidge tube ($I_{max}$ = 125 mA, $V_{max}$ = 60 kV) with a Rh anode; the primary X-ray spectrum can be controlled by inserting filters (Al, at different thickness) between the anode and the sample. Each element was analyzed three times, with specific conditions (voltage, tube filter, collimator, analyzing crystal, and detector). Data collection was controlled by the SuperQ software provided with the instrument. The elemental mass thickness (µg $cm^{-2}$), that is, the analyzed elemental mass per unit surface, was obtained by comparing the elemental yields with a sensitivity curve measured in the same geometry on a set of certified mono- or bi-elemental thin layer standards by Micromatter Inc. The certified uncertainty of the standard deposit (± 5%) determines the lower limit of the uncertainty of the measured elemental concentrations, which ranges between 8% and 10% depending on the element considered element. Thanks to the uniformity of the aerosol deposit on the filters, the atmospheric elemental concentrations (µg $m^{-3}$) were calculated by multiplying the analyzed elemental mass thickness by the ratio between the collection and analyzed surfaces of each sample (41 and 22 mm, respectively), then dividinged by the total sampled volume ($m^3$). Finally, concentrations of light-weight elements (atomic

number Z < 19) were corrected for the underestimation induced by the self-absorption of the emitted soft X-rays inside aerosol particles according to Formenti et al. (~~2011~~2010).

Additional XRF analysis of the quartz filters was ~~has been~~ performed both in the $PM_{10.6}$ and the $PM_{2.5}$ fractions, ~~so~~ to verify the absence of biases between the experiments dedicated to the determination of particle composition ~~to~~ and those where the optical properties where measured.

### 2.6.2. Iron oxide content

The content and the mineralogical speciation of the iron oxides, also defined as free-iron, ~~that is~~i.e., the fraction of iron ~~which~~ that is not in the crystal lattice of silicates (Karickhoff and Bailey, 1973), was determined by XANES (X-ray absorption near-edge structure) in the Fe K-range ($K_\alpha$, 7112 eV) at the SAMBA (Spectroscopies Applied to Materials based on Absorption) beamline at the SOLEIL synchrotron facility in Saclay, France (Briois et al., 2011). The position and shape of the K pre-edge and edge peaks were analyzed as they depend on the oxidation state of iron and the atomic positions of the neighboring ions, mostly $O^+$ and $OH^-$.

As in Formenti et al. (2014b), samples were mounted in an external setup mode. A Si(220) double-crystal monochromator was used to produce a monochromatic X-ray beam, which was 3000 x 250 $\mu m^2$ in size at the focal point. The energy range was scanned from 6850 eV to 7800 eV at a step resolution varying between 0.2 eV in proximity to the Fe-K absorption edge (at 7112 eV) to 2 eV in the extended range. Samples were analyzed in fluorescence mode without prior preparation. One scan acquisition lasted approximately 30 minutes, and was repeated three times to improve the signal-to-noise ratio.

The same analytical protocol was applied to five standards of Fe(III)-bearing minerals (**Table 1**), including iron oxides (hematite, goethite) and silicates (illite, montmorillonite, nontronite). The standard spectra were used to deconvolute the dust sample spectra to quantify the mineralogical status of iron. The linear deconvolution was~~,~~ performed with the Athena IFEFFIT freeware analysis program (Ravel and Newville, 2005). This~~,~~ provided ~~with~~ the proportionality factors, $\alpha_i$~~,~~, representing the mass fraction of elemental iron to be assigned to the *i*-th standard mineral. In particular, the values of $\alpha_{hem}$ and $\alpha_{goe}$ represent the mass fractions of elemental iron that can be attributed to hematite and goethite, and $\alpha_{Fe\,ox}$ ($\alpha_{hem} + \alpha_{goe}$), the mass fraction of elemental iron that can be attributed to iron oxides.

### 2.6.3. Calculation of the iron oxide content

The measured elemental concentrations obtained by X-ray Fluorescence (XRF) are expressed in the form of elemental oxides and summed to estimate the total mineral dust mass concentration $MC_{dust}$ according to the equation from Lide (1992)

$$[MC_{dust}]=1.12\times\left\{\begin{matrix}1.658[\mathrm{Mg}]+1.889[\mathrm{Al}]+2.139[\mathrm{Si}]+1.399[\mathrm{Ca}]+1.668[\mathrm{Ti}]+1.582[\mathrm{Mn}]\\+(0.5\times1.286+0.5\times1.429+0.47\times1.204)[\mathrm{Fe}]\end{matrix}\right\} \quad (4)$$

The relative uncertainty ~~on~~ in $MC_{dust}$, estimated from the analytical error i~~o~~n the measured concentrations, does not exceed 6%. As it will be explained in the result section (paragraph 3.1), the values of $MC_{dust}$ estimated from Equation 4 were found in excellent agreement with the measured gravimetric mass on the filters.

The fractional mass ratio (in percent) of elemental iron ($MR_{Fe\%}$) with respect to the total dust mass concentration, $MC_{dust}$, is then calculated as

$$MR_{Fe\,\%} = \frac{[Fe]}{[MC_{Dust}]} \times 100 \quad (5)$$

The mass concentration of iron oxides or free-iron ($MC_{Fe\,ox}$), representing the fraction of elemental iron in the form of hematite and goethite ($Fe_2O_3$ and FeOOH, respectively), is equal to

$$MC_{Fe\,ox} = MC_{hem} + MC_{goe} \quad (6)$$

where $MC_{hem}$ and $MC_{goe}$ are the total masses of hematite and goethite. These can be calculated from the values $\alpha_{hem}$ and $\alpha_{goe}$ from XANES analysis, which represent the mass fractions of elemental iron attributed to hematite and goethite, as

$$MC_{hem} = \frac{\alpha_{hem}\, x\, [Fe]}{0.70} \quad (7.a)$$

$$MC_{goe} = \frac{\alpha_{goe}\, x\, [Fe]}{0.63} \quad (7.b)$$

where the values of 0.70 and 0.63 represent the mass molar fractions of Fe in hematite and goethite, respectively. The relative errors of~~n~~ $MC_{hem}$ and $MC_{goe}$ are obtained from the uncertainties ~~on~~ of the values of $\alpha_{hem}$ and $\alpha_{goe}$ from XANES analysis (less than 10%).

The mass ratio of iron oxides ($MR_{Fe\ ox\%}$) with respect to the total dust mass can then be calculated as

$$MR_{Fe\ ox\%} = MC_{Fe\ ox} \times MR_{Fe\ \%} \quad (8)$$

## 3. Experimental protocol

At the beginning of each experiment, the chamber was evacuated ~~by~~ to $10^{-4}$-$10^{-5}$ hPa. Then, the reactor was filled with a mixture of 80% $N_2$ and 20% $O_2$ at a pressure slightly exceeding the current atmospheric pressure, in order to avoid contamination from ambient air. The e~~E~~xperiments were conducted at ambient temperature and at a relative humidity <2%. As in Di Biagio et al. (2014; ~~2016~~2017), dust aerosols were generated by mechanical shaking of the parent soils, previously sieved to < 1000 µm and dried at 100 °C for about 1 h to remove any residual humidity. About 15 g of soil was placed in a Buchner flask and shaken for about 30 min at 100 Hz by means of a sieve shaker (Retsch AS200). The dust particles ~~produced w~~produced by the mechanical shaking, mimicking the saltation processing that soils experience when eroded by strong winds, ~~as~~ were ~~then~~ injected in the chamber by flushing the flask with $N_2$ at 10 L $min^{-1}$ for about 10-15 min, whilst continuing shaking the soil. Di Biagio et al. (2014; 2017) have demonstrated the realism of the generation system concerning the composition and the size distribution of the generated dust with respect to the properties of mineral dust in the atmosphere.

The ~~D~~dust ~~was injected for about 10-15 minutes, and left~~ remained suspended in the chamber for approximately 120 min thanks to the 4-wheel fan located in the bottom of the chamber body. Previous measurements at the top and bottom of the chamber showed that the fan ensures a homogeneous distribution of the dust starting approximately 10 minutes after the end of the injection (Di Biagio et al., 2014).

To compensate for the air extracted from the chamber by sampling, a particle-free flow of $N_2/O_2$, regulated in real time as a function of the total volume of sampled air, was re-injected in the chamber. To avoid excessive dilution the flow was limited to 20 L $min^{-1}$. Two experiments per soil type were conducted: a first experiment for sampling on the nuclepore polycarbonate filters (determination of the elemental composition and the iron oxide fraction) and *in situ* measurements of the infrared optical constants (Di Biagio et al., ~~2016~~2017), and a second experiment sampling on total quartz filter and impactor for the study of dust MAE presented in this paper.

**Figure 1** illustrates as typical example the time series of the aerosol mass concentration during the two experiments conducted for the Libyan sample. The comparison demonstrates the repeatability of the dust concentrations, both in absolute values and in temporal dynamics. It also shows that the mass concentrations decreased very rapidly by gravitational settling within the first 30 minutes of the experiment (see also the discussion in Di Biagio et al., ~~(2016~~2017~~)~~), after which concentrations only decrease by dilution. The filter sampling was started after this transient phase, and then continued through the end of the experiments, in order to collect enough dust on the filter membranes for subsequent ~~the~~ chemical analysis. Blank samples were collected before the start of the experiments by placing the ~~loaded filter~~ holders loaded with filter membranes in line with the chamber and by flushing them for a few seconds with air coming from the chamber.

At the end of each experimental series with a given soil sample, the chamber was manually cleaned in order to remove carry-over caused by resuspension of particles deposited to the walls. Background concentrations of aerosols in the chamber vary between 0.5 and 2.0 µg $m^{-3}$, i.e., a factor of 500 to 1000 below the operating conditions.

## ~~3~~4. Results and discussion

The geographical location of the soil collection sites is shown in **Figure 2**, ~~whereas~~ and the coordinates are summarized in **Table 2**. ~~As discussed in Di Biagio et al. (2016), the selection of these soils and sediments was governed by the need of representing the major arid and semi-arid regions worldwide, the need of taking into account the mineralogical diversity of the soil composition at the global scale, and finally by their availability in sufficient quantities for injection in the chamber. When doing so, we obtained a set of twelve samples distributed worldwide but mostly in Northern and Western Africa (Libya, Algeria, Mali, Bodélé) and the Middle East (Saudi Arabia and Kuwait). Individual samples from the Gobi desert in Eastern Asia, the Namib Desert, the Strzelecki desert in Australia, the Patagonian~~

~~deserts in South America, and the Sonoran Desert in Arizona have also been investigated.~~ The selection of these soils and sediments was made out of 137 individual top-soil samples collected in major arid and semi-arid regions worldwide and representing the mineralogical diversity of the soil composition at the global scale. As discussed in Di Biagio et al. (2017), this large sample set was reduced ~~by~~to a set of 19 samples representing the mineralogical diversity of the soil composition at the global scale and based on their availability in sufficient quantities for injection in the chamber. Because some of the experiments did not produce enough dust to perform good-quality optical measurements, ~~,~~ in this paper we present a set of twelve samples distributed worldwide but mostly from~~in~~ Northern and Western Africa (Libya, Algeria, Mali, Bodélé) and the Middle East (Saudi Arabia and Kuwait). Individual samples from the Gobi desert in Eastern Asia, the Namib Desert, the Strzelecki desert in Australia, the Patagonian deserts in South America, and the Sonoran Desert in Arizona ~~have~~were also ~~been~~ investigated.

### ~~3~~4.1. Elemental composition and iron oxide content

A total of 41 filters including 15 polycarbonate filters (12 samples and 3 blanks) and 25 quartz filters (12 for the total fraction, 10 for the fine fraction and 3 blanks) were collected for analysis.

The dust mass concentration found by gravimetric analysis varied between 50 μg $m^{-3}$ and 5 mg $m^{-3}$, in relatively good agreement with the dust mass concentrations, $MC_{dust}$ ~~-~~, from ~~(~~Equation 4~~)~~, based on ~~X-ray~~ Ray ~~fluorescence~~ Fluorescence XRF ~~analysis~~aAnalysis: the slope of the linear regression between the calculated and the gravimetric values of $MC_{dust}$ is 0.90 with $R^2 = 0.86$.

Di Biagio et al. (~~2016~~2017) showed that clays are the most abundant mineral phases, together with quartz and calcite, and that significant variability exists as function of the compositional heterogeneity of the parent soils. Here we use the Fe/Ca and Si/Al elemental ratios obtained from ~~X-ray Fluorescence~~XRF analysis to discriminate the origin of ~~used~~ dust samples. These ratios have been extensively used in the past to discriminate the origin of African dust samples collected in the field (Chiapello et al., 1997; Formenti et al., 2011; ~~Formenti et al.,~~ 2014a). The values obtained during our experiments are reported in **Table 3.** There is a very good correspondence between the values obtained for the Mali, Libya, ~~,~~ Algeria, ~~Mauritania~~ and (to a lesser extent) Morocco experiments to values found in environmental aerosol samples by Chiapello et al. (1997) and Formenti et al. (2011; 2014a). These authors indicate that dust from local erosion of Sahelian soils, such as from Mali, have Si/Al ratios in the range of 2-2.5 and Fe/Ca ratios in the range 3-20, depending on the time proximity to the erosion event. Dust from sources in the Sahara, such as Libya and Algeria, show Si/Al ratios in the range of 2-3 and Fe/Ca ratios in the range

0.7-3, whereas dust from Morocco has Si/Al ratios around 3 and Fe/Ca ratios around 0.4. The only major difference is observed for the Bodélé experiment, for which the Fe/Ca ratio is enriched by a factor of 6 with respect to the values of 1 found during to the field observations (Formenti et al., 2011; Formenti et al., 2014a). This could reflect the fact that the Bodélé aerosol in the chamber is generated from a sediment sample and not from a soil. As a matter of fact, the Bodélé sediment sample is constituted consists of by a very fine powder which becomes very easily airborne. Henceforth, and contrary to the soil samples, this This powder is likely to be injected in the chamber with little or no size fractionation. Henceforth, the aerosol generated from it . As a consequence, should have a closer the composition to the original powder of the aerosol collected in the chamber could reflect more that of the parent sedimentary soil than the not the other samples. On the other hand, Bristow et al. (2010) and Moskowitz et al. (2016) showed that the iron content and speciation of the Bodélé sediments is very heterogeneous at the source scale. For samples from areas other than non northern Africa n samples, the largest variability is observed for the Fe/Ca values, ranging from 0.1 to 8, whereas the Si/Al ratio varied only between 2.5 and 4.8. In this case, values are available in the literature for comparison (e.g., Cornille et al., 1990; Reid et al., 1994; Eltayeb et al., 2001; Lafon et al., 2006; Shen et al., 2007; Radhi et al., 2010; 2011; Formenti et al., 2011; 2014a; Scheuvens et al., 2013, and references within). Values in the $PM_{2.5}$ fraction are very consistent with those obtained in the $PM_{10.6}$: their linear correlation has a slope of 1.03 (± 0.05) and a $R^2$ equal to 0.97, suggesting that the elemental composition is relatively size -independent.

The mass fraction of total Fe ($MC_{Fe\%}$ from Equation 5), also reported in **Table 3**, ranged from 2.8 (Namibia) to 7.3% (Australia), values found for the Namibia and the Australia samples, respectively. This These are in the range is in good agreement with of values reported in the literature, taking into account that differences might be also due to the method (direct measurement/calculation) and/or the size fraction over which the total dust mass concentration is estimated (Chiapello et al., 1997; Reid et al., 1994; 2003; Derimian et al., 2008; Formenti et al., 2001; 2011; 2014a; Scheuvens et al., 2013). The agreement of $MC_{Fe\%}$ values obtained by the XRF analysis of polycarbonate filters (Equation 5) and those obtained from the XRF analysis of the quartz filters, normalized to the measured gravimetric mass is well within 10% (that is, the percent error of each estimate). An the e Exceptions are the samples of from Bodélé and Algeria, for which the values obtained from the analysis of the quartz filters are significantly lower than those obtained from the nuclepore filters (3.1% versus 4.1% for Bodélé and 4.3% versus 6.8% for Algeria). We treat that as an additional source of error in the rest of the analysis, and add it to the total uncertainty. In the $PM_{2.5}$ fraction, the content of iron is more variable, ranging from 4.4% (Morocco) to

33.6% (Mali), showing a size dependence. A word of caution on this conclusion ~~as~~ is that the two estimates are not necessarily consistent in the way that the total dust mass is estimated (from Equation 4 for the $PM_{10.6}$ fraction and by gravimetric weighing for~~on~~ the $PM_{2.5}$).

Finally, between 11 and 47% of iron in the samples can be attributed to iron oxides, in variable proportions between hematite and goethite. The iron oxide fraction of total Fe in this study is ~~on~~ at the lower end of the range (36-72%) estimated for field dust samples of Saharan/Sahelian origin (Formenti et al. 2014b). The highest value of Formenti et al. (2014b), obtained for a sample of locally-emitted dust collected at the Banizoumbou station in the African Sahel, is anyhow in excellent agreement with the value of 62% obtained for an experiment (not shown here) using a soil collected in the same area. Likewise, the proportions between hematite and goethite (not shown) are reproduced, showing that goethite is more abundant than hematite. The mass fraction of iron oxides ($MR_{Fe\ ox\%}$), estimated from Equation 8 and shown in Table 3, ranges between 0.7% (Kuwait) ~~to~~ and 3.6% (Australia), which is in the range of available field estimates (Formenti et al., 2014a; Moskowitz et al., 2016). For China, our value of $MR_{Fe\ ox\%}$ is lower by almost a factor of 3 ~~in comparison with~~compared to that obtained on ~~the same~~ dust of the same origin ~~sample~~ by Alfaro et al. (2004) (0.9% against 2.8%), whereas on a sample from Niger (~~however~~ not considered in this study) our estimates and that by Alfaro et al. (2004) agree perfectly ~~agree~~ (5.8%). A possible underestimate of the iron oxide fraction for samples other than those from the Sahara-Sahel area could be due to the fact that - opposite to the experience of Formenti et al. (2014b) - the linear deconvolutions of the XANES spectra were not always satisfactory (see Figure S1 in the supplementary). This resulted in a significant residual between the observed and fitted XANES spectra. ~~Indeed~~In fact, the mineralogical reference for hematite is obtained from a soil from Niger (Table 1) and might not be fully suitable for representing aerosols of different origins. Additional differences could arise from differences in the size distributions of the generated aerosol. As a matter of fact, the number fraction of particles in the size classes above 0.5 µm in diameter is ~~are~~ different in the dust aerosol generated in the Alfaro et al. (2004) study ~~with respect~~ compared to ours. In the study by Alfaro et al. (2004), the number fraction of particles is lowest in the 0.5-0.7 size class and highest between 1 and 5 µm. ~~On the contrary~~In contrast, in our study the number fraction is lowest in the 1-2 µm size range and highest between 0.5 and 0.7 µm. These differences could ~~yield~~ either be due to differences in the chemical composition and/or ~~to a difference~~ in the total mass in the denominator of Equation 8.

### ~~3~~4.2. Spectral and size ~~-~~variability of the mass absorption efficiency

The spectral mass absorption efficiencies (MAE) at 375, 407, 532, 635, and 850 nm for the $PM_{10.6}$ and the $PM_{2.5}$ dust fractions are summarized in **Table 4** and displayed in **Figure 3**. Regardless of particle size, the MAE values decrease with increasing wavelength (almost one order of magnitude between 375 and 850 nm), and display a larger variability at shorter wavelengths. The MAE values for the $PM_{10.6}$ range from 37 (± 3) $10^{-3}$ $m^2$ $g^{-1}$ to 135 (± 11) $10^{-3}$ $m^2$ $g^{-1}$ at 375 nm, and from 1.3 (± 0.1) $10^{-3}$ $m^2$ $g^{-1}$ to 15 (± 1) $10^{-3}$ $m^2$ $g^{-1}$ at 850 nm. Maxima are found for the Australia and Algeria samples, whereas the minima are for Bodélé and Namibia, respectively at 375 and 850 nm. In the $PM_{2.5}$ fraction, the MAE values range from 95 (± 8) $10^{-3}$ $m^2$ $g^{-1}$ to 711 (± 70) $10^{-3}$ $m^2$ $g^{-1}$ at 375 nm, and from 3.2 (± 0.3) $10^{-3}$ $m^2$ $g^{-1}$ to 36 (± 3) $10^{-3}$ $m^2$ $g^{-1}$ at 850 nm. Maxima at both 375 and 850 nm are found for the Morocco sample, whereas the minima are for Algeria and Namibia, respectively. The MAE values for mineral dust resulting from this work are relatively in good agreement with the estimates available in the literature (Alfaro et al., 2004; Linke et al., 2006; Yang et al., 2009; Denjean et al., 2016), reported in **Table 5**. For the China Ulah Buhn sample, Alfaro et al. (2004) reported 69.1 $10^{-3}$ and 9.8 $10^{-3}$ $m^2$ $g^{-1}$ at 325 and 660 nm, respectively. The former is lower than the value of 99 $10^{-3}$ $m^2$ $g^{-1}$ that we obtain by extrapolating our measurement at 375 nm. Likewise, our values for the Morocco sample are higher than reported by Linke et al. (2006) at 266 and 660 nm. Conversely, the agreement with the estimates of Yang et al. (2009) for mineral dust locally re-suspended in Xianghe, near Beijing (China) is very good at all wavelengths between 375 and 880 nm. As expected, the MAE values for mineral dust resulting from this work are almost one order of magnitude smaller than for other absorbing aerosols. For black carbon, MAE values are in the range of 6.5–7.5 $m^2$ $g^{-1}$ at 850 nm (Bond and Bergstrom, 2006; Massabò et al., 2016), and vary decrease in a linear way with the logarithm of the inversely with wavelength. For brown carbon, the reported MAE range between 2.3–7.0 $m^2$ $g^{-1}$ at 350 nm (Chen and Bond, 2010; Kirchstetter et al., 2004; Massabò et al., 2016), 0.05–1.2 $m^2$ $g^{-1}$ at 440 nm (Wang et al., 2016) and 0.08–0.72 $m^2$ $g^{-1}$ at 550 nm (Chen and Bond, 2010).

The analysis of **Table 4** indicates that, at every wavelength, the MAE values in the $PM_{2.5}$ fraction are equal or higher than those for $PM_{10.6}$. The $PM_{2.5}/PM_{10.6}$ MAE ratios reach values of 6 for the Mali sample, but are mostly in the range 1.5-3 for the remaining other aerosols. The vValues decrease with wavelength up to 635 nm, whereas at 850 nm they have values comparable to those at 375 nm. The observed size -dependence of the MAE values is consistent with the expected behavior of light absorption of particles in the Mie and geometric optical regimes that concern are relevant for the two size fractions. Light -absorption of particles of sizes smaller or equivalent to the wavelength is proportional to their

bulk volume, whereas for larger particles absorption occurs on their surface only (Bohren and Huffmann, 1983). On the other hand, the size-resolved measurements of Lafon et al. (2006) show that the proportion (by volume) of iron oxides might be higher in the coarse than in the fine fraction, which would counteract the size-dependence behavior of MAE. To validate the observations, we calculated the spectrally-resolved MAE values in the two size fractions using the Mie code for homogeneous spherical particles (Bohren and Huffmann, 1983) and the number size distribution estimated by ~~(~~Di Biagio et al~~., ~~2016~~(2017)~~ and averaged over the duration of filter sampling. We estimated the dust complex refractive index as a volume-weighted average of a non-absorbing dust fraction having the refractive index of kaolinite, the dominant mineral in our samples (see Di Biagio et al., ~~2016~~2017), from Egan et Hilgeman (1979) and an absorbing fraction estimated from the mass fraction of iron oxides and having the refractive index of hematite (Bedidi and Cervelle, 1993). The r~~R~~esults of this calculation indicate that the observed size-dependent behavior is well reproduced at all wavelengths, even in the basic hypothesis that the mineralogical composition does not change with size. The only exception is 850 nm, where at times, $PM_{2.5}/PM_{10.6}$ MAE ratio is much higher than expected theoretically. We attribute that to the relatively high uncertainty affecting the absorbance measurements at this wavelength, where the signal-to-noise ratio is low. Indeed, the two sets of values (MAE in the $PM_{2.5}$ fraction and MAE in the $PM_{10.6}$ fraction) are not statistically different according to a two-pair t-test (0.01 and 0.05 level of confidence), confirming that any attempt of differentiation of the size ~~-~~dependence at this wavelength would require a stronger optical signal.

The analysis of the spectral dependence, using ~~a~~ the power-law function fit ~~as (from~~ Equation 2), provides ~~with~~ the values of the Angstrom Absorption Exponent (AAE), also reported in **Table 4**. Contrary to the MAE values, there is no statistically significant size ~~-~~dependence of the AAE values, ranging from 2.5 (± 0.2) to 4.1 (± 0.3), with an average of 3.3 (± 0.7), for the $PM_{10.6}$ size fraction and between 2.6 (± 0.2) and 5.1 (± 0.4), with an average of 3.5 (± 0.8), for the $PM_{2.5}$ fraction. Our values are in the range of those published in the ~~open~~ literature (Fialho et al., 2005; Linke et al., 2006; Müller et al., 2009; Petzold et al., 2009; Yang et al., 2009; Weinzierl et al., 2011; Moosmüller et al., 2012; Denjean et al., 2016), shown in **Table 5**. AAE values close to 1.0 are found for urban aerosols where fossil fuel~~s~~ combustion is dominant, while AAE values for brown carbon (BrC) from incomplete combustion are in the range 3.5-4.2 (Yang et al., 2009; Chen et al., 2015; Massabò et al., 2016).

Finally, **Figure 4** shows correlations between the MAE values in the $PM_{10.6}$ fraction (Figure ~~3~~4.a) and in the $PM_{2.5}$ fraction (Figure ~~3~~4.b) and the estimated percent mass fraction of iron and iron oxides

($MC_{Fe\%}$ and $MC_{Fe\ ox\%}$), respectively. Regardless of the size fraction, t~~T~~he correlation between the MAE values and the percent mass of total elemental iron are ~~satisfactory. Higher~~ higher ~~correlations are obtained~~ at 375, 407 and 532 nm ~~, and in the $PM_{2.5}$ fraction, where a linear correlation with $R^2$ up to 0.94 are obtained~~. Best correlations are obtained when forcing the intercept to zero, indicating that elemental iron fully accounts for the measured absorption. At these wavelengths, linear correlations with the mass fraction of iron oxides are ~~loose~~ low in the $PM_{10.6}$ mass fraction ($R^2$ up to 0.38-0.62), but ~~again satisfactory~~higher in the $PM_{2.5}$ fraction ($R^2$ up to 0.83-0.99), where, ~~whoever~~however, one should keep in mind that they have been established only indirectly by considering the ratio of iron oxides to elemental iron independent of size. At 660 and 850 nm, little or no robust correlations are ~~is~~ obtained, often based on very few data points and with very low MAE values. It is noteworthy that, in both size fractions, the linear correlation yields a non-zero intercept ~~is obtained~~, indicating a contribution from minerals other ~~minerals but~~than iron oxides ~~account~~ to ~~for~~ the measured absorption.

## 45. Conclusive remarks

In this paper, we report~~ed~~ new laboratory measurements of the shortwave mass absorption efficiency (MAE) of mineral dust of different origins and as a function of size and wavelength in the 375-850 nm range. ~~Results~~ Our results ~~have been~~were obtained in the CESAM simulation chamber using ~~generated~~ mineral dust generated from natural parent soils, in combination with ~~and~~ optical and gravimetric analysis on extracted samples.

Our results can be summarized as follows: at 375 nm, the MAE values are lower for the $PM_{10.6}$ mass fraction (range 37-135 $10^{-3}$ $m^2\ g^{-1}$) than for the $PM_{2.5}$ fraction (range 95-711 $10^{-3}$ $m^2\ g^{-1}$), and vary opposite to wavelength as $\lambda^{-AAE}$, where AAE (Angstrom Absorption Exponent) averages between 3.3-3.5 regardless of size fraction. These results deserve some ~~conclusive~~ concluding comments:

- The size ~~-~~dependence, ~~yielding~~ characterized by significantly higher MAE values in the fine fraction ($PM_{2.5}$) than ~~for the~~ in the bulk ($PM_{10.6}$) aerosol, indicates that light ~~-~~absorption by mineral dust can be important even during atmospheric transport over heavily polluted regions, ~~when~~ where dust concentrations are significantly lower than at emission. This can be shown by comparing the aerosol absorption optical depth (AAOD) at 440 nm for China, a well-known mixing region of mineral dust and pollution (e.g., Yang et al., 2009; Laskin et al., 2014; Wang et al., 2013), as well as offshore western Africa where large urban centers are downwind of dust transport areas (Petzold et al., 2011). Laskin et al. (2014) reports that the average AAOD in China

~~the area~~ is of the order of 0.1~~,~~ for carbonaceous absorbing aerosols (sum of black and brown carbon; Andreae and Gelencsér, 2006). This is lower or comparable to the AAOD of 0.17 and 0.11 at 407 nm (~~fine and~~ total and fine mass fractions, respectively) that we ~~obtain~~ derive by a simple calculation (AAOD = MAE x $MC_{dust}$ x H), ~~where~~ from MAE ~~are the~~ values estimated in this study, ~~(~~, $MC_{dust}$~~,~~, the dust mass concentrations typically observed in ~~the area~~urban ~~area of~~ Beijing during dust storms (Sun et al., ~~2001~~2005), and $H$, a scale height factor of 1 km~~)~~.

- The spectral variability of the dust MAE values, represented by the AAE parameter, is equal in the $PM_{2.5}$ and $PM_{10.6}$ mass fractions. This suggests that, for a given size distribution, the possible variation of dust composition with size does not affect in a significant way the spectral behavior of the absorption properties. Our average value for AAE is 3.3 ± 0.7, higher than for black carbon, but in the same range ~~than~~ as light-absorbing organic (brown) carbon. As a result, depending on the environment, there can be some ambiguity in apportioning the AAOD based on spectral dependence. Bahadur et al. (2012) and Chung et al. (2012) couple the AAE and the spectral dependence of the total AOD (and/or its scattering fraction only) to overcome this problem. Still, Bahadur et al. (2012) show that there is an overlap in the scatterplots of the spectral dependence of the scattering and absorption fractions of the AOD based on an analysis of ground-based remote sensing data for mineral dust, urban, and non-urban fossil fuel over California. A closer look should be taken at ~~to~~ observations in mixing areas where biomass burning aerosols may have different chemical composition and/or mineral dust has heavy loadings ~~should be given~~ in order to generalize the clear separation observed in the spectral dependences of mineral dust and biomass burning (Bahadur et al., 2012). This aspect is relevant to the development of remote sensing retrievals of light~~-~~ absorption by aerosols from space, and their assimilation in climate models (Torres et al., 2007; Buchard et al., 2015; Hammer et al., 2016).

- There is an important sample-to-sample variability in our dataset of MAE values for mineral dust aerosols. At 532 nm, our ~~estimated~~ average MAE values ~~average to~~are ~~at~~ 34 ± 14 $m^2$ $g^{-1}$ and 78 ± 70 $m^2$ $g^{-1}$ in the $PM_{10.6}$ and $PM_{2.5}$ mass fractions, respectively. Figure 3, showing the correlation with the estimated mass fraction of elemental iron and iron oxides, suggests that this variability could be related to the regional differences of the mineralogical composition of the parent soils. These observations lead to ~~different considerations~~ further conclusions. To start with, our study reinforces the need for regionally-resolved representation of the light ~~-~~absorption properties of

mineral dust in order to improve the representation of its effect on climate. As a matter of fact, the natural variability of the absorption properties that we obtain from our study is in the range 50-100%, even when we limit ourselves to smaller spatial scales, for example those ~~of~~ from north Africa (samples from Libya, Algeria, Mali and Bodélé). ~~This is far above the ± 5% sensitivity factor used by Solmon et al. (2008) to vary the single scattering albedo (as a proxy of absorption) of mineral dust over western Africa, and to show how this could drastically change the climate response in the region.~~ As a comparison, Solmon et al. (2008) showed that varying the single scattering albedo of mineral dust over western Africa by ± 5%, that is, varying the co-albedo (or absorption) by 45% (0.1± 0.045) could drastically change the climate response in the region.

The question is then "how to represent this regional variability?" ~~As~~ Like Moosmüller et al. (2012) and Engelbrecht et al. (2016), we found that elemental iron is a very good proxy for the MAE, especially in the $PM_{2.5}$ fraction, where iron-bearing absorbing minerals (hematite, goethite, illite, smectite clays) ~~would be~~are more concentrated. In the coarse fraction, Ca-rich minerals, quartz, and feldspars could also play a role, and that could result in the observed lower~~ed~~ correlation (although adding a term proportional to elemental Ca does not ~~ameliorate~~ improve the correlation ~~result~~ in the present study). The correlation of the spectral MAE values with the iron oxide fraction is satisfactory but rather noisy, also owing to some uncertainty in the quantification of iron oxides from X-Ray ~~Absorption~~ absorption measurements. In this case, the intercept is significantly different from zero, again indicating that a small but ~~clear~~ distinct fraction of absorption is due to minerals other than iron oxides. There are contrasting results on this topic: Alfaro et al. (2004) found an excellent correlation between MAE and the iron oxide content, whereas Klaver et al. (2011) found that the single scattering albedo (representing the capacity of an aerosol population to absorb light ~~with respect~~in relation to extinction) was almost independent on the mass fraction of iron oxides. Moosmüller et al. (2012) disagreed, pointing out ~~to~~ the uncertainty in the correction procedure of the measurement of absorption by Klaver et al. (2011). As a matter of fact, Klaver et al. (2011) and Alfaro et al. (2004) used the same correction procedure. It is more likely that the lack of correlation found in Klaver et al. (2011) is due to the fact that ~~other~~ minerals other than iron oxides contribute to absorption, in particular at their working wavelength (567 nm), where the absorption efficiency of iron oxides starts to weaken. Clearly, the linear correlation between elemental iron in mineral dust and its light-absorption properties

could ease the application and validation of climate models that are now starting to include ~~including~~ the representation of the mineralogy (Perlwitz et al., 2015a; 2015b; Scanza et al., 2015). Also, ~~they~~ this would facilitate detecting source regions based on remote sensing of dust absorption in the UV-VIS spectral region (e.g., Hsu et al., 2004). However, such a quantitative relationship cannot be uniquely determined from these studies, including the present one, which use different ways of estimating elemental iron, iron oxides, and the total dust mass. A more robust estimate should be obtained from~~by~~ ~~estimating~~ the imaginary parts of the complex refractive indices associated ~~to~~ with the~~se~~ measurements of absorption, and their dependence on the mineralogical composition.

## Author contributions

L. Caponi, P. Formenti, D. Massabò, P. Prati, C. Di Biagio, and J. F. Doussin designed the chamber experiments and discussed the results. L. Caponi and C. Di Biagio ~~realized~~ conducted the experiments with contributions by M. Cazaunau, E. Pangui, P. Formenti, and J.F. Doussin. L. Caponi, D. Massabò and P. Formenti performed the full data analysis with contributions by C. Di Biagio, P. Prati and J.F. Doussin. L. Caponi, P. Formenti and S. Chevaillier performed the XRF measurements. P. Formenti and G. Landrot performed the XAS measurements. D. Massabò performed the MWAA and the gravimetric measurements. M. O. Andreae, K. Kandler, T. Saeed, S. Piketh, D. Seibert, and E. Williams collected the soil samples used for experiments. L. Caponi, P. Formenti, D. Massabò and P. Prati wrote the manuscript with comments from all co-authors.

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## Table captions

**Table 1.** Characteristics of the standards used for the quantification of the iron oxides in the XAS analysis.

**Table 2.** ~~Summary~~ Geographical ~~of~~ information on the soil samples used in this work.

**Table 3.** Chemical characterisation of the dust aerosols in $PM_{10.6}$ and $PM_{2.5}$ (in parentheses) size fractions. Columns 3 and 4 ~~report~~ give the Si/Al and Fe/Ca elemental ratios obtained from X-Ray Fluorescence analysis. The uncertainty of~~n~~ each individual value is estimated to be 10%. Column 5 ~~reports~~ shows $MR_{Fe\%}$, the fractional mass of elemental iron with respect to the total dust mass concentration (uncertainty 10%). Column 5 reports $MR_{Fe\%}$, the mass fraction of iron oxides with respect to the total dust mass concentration (uncertainty 15%). For $PM_{2.5}$ the determination of the Si/Al ratio is impossible due to the composition of the filter ~~medium~~ membranes (quartz).

**Table 4.** Mass absorption efficiency (MAE, $10^{-3}$ $m^2$ $g^{-1}$) and Ångström Absorption Exponent (AAE) in the $PM_{10.6}$ and $PM_{2.5}$ size fractions. Absolute errors are in brackets.

**Table 5.** Mass absorption efficiency (MAE, $10^{-3}$ $m^2$ $g^{-1}$) and Ångström Absorption Exponent (AAE) ~~of~~ from the literature data discussed in the paper

## Figure captions

**Figure 1.** Time series of aerosol mass concentration in the chamber for ~~the~~ two companion experiments (Libyan dust ~~sample~~). Experiment 1 (top panel) was dedicated to the determination of the chemical composition (including iron oxides) by sampling on polycarbonate filters. Experiment 2 (bottom panel) was dedicated to the determination of the absorption optical properties by sampling on quartz filters.

**Figure 2.** Locations (red stars) of the soil and sediment samples used to generate dust aerosols.

**Figure 3.** Spectral dependence of the MAE values for the samples investigated in this study in the $PM_{10.6}$ (left) and in the $PM_{2.5}$ (right) mass fractions.

**Figure 4.** Illustration of the links between the MAE values and the dust chemical composition found in this study. Left column, from top to bottom: linear regression between the MAE values ~~between~~ in the range from 375 to~~and~~ 850 nm and the fraction of elemental iron ~~with respect to~~ relative to the total dust mass ($MR_{Fe\%}$) in the $PM_{10.6}$ fraction; Middle column: same as left column but ~~respect to~~for the mass

fraction of iron oxides relative to the total dust mass ($MR_{Fe\ ox\%}$) in the $PM_{10.6}$ size fraction; Right column: same as left column but in the $PM_{2.5}$ size fraction.

**Table 1.** Characteristics of the standards used for the quantification of the iron oxides in the XAS analysis.

| Standard | ~~Stoechiometric~~Stoichiometric Formula | Origin |
|---|---|---|
| Illite of Puy | $(Si_{3.55}Al_{0.45})(Al_{1.27}Fe_{0.36}Mg_{0.44})O_{10}(OH)_2(Ca_{0.01}Na_{0.01}K_{0.53}X(I)_{0.12})$ | Puy, France |
| Goethite | FeO OH | Minnesota |
| Hematite | $Fe_2O_3$ | Niger |
| Montmorillonite | $(Na,Ca)_{0,3}(Al,Mg)_2Si_4O10(OH)_2 \cdot n(H_2O)$ | Wyoming |
| Nontronite | $Na_{0.3}Fe_2(Si,Al)_4O10(OH)_2 \cdot nH2O$ | Pennsylvania |

**Table 2.** Geographical information on the soil samples used in this work.

| Geographical area | Sample | Desert area | Geographical coordinates |
|---|---|---|---|
| Sahara | Morocco | East of Ksar Sahli | 31.97°N, 3.28°W |
| | Libya | Sebha | 27.01°N, 14.50°E |
| | Algeria | Ti-n-Tekraouit | 23.95°N, 5.47°E |
| Sahel | Mali | Dar el Beida | 17.62°N, 4.29°W |
| | Bodélé | Bodélé depression | 17.23°N, 19.03°E |
| Middle East | Saudi Arabia | Nefud | 27.49°N, 41.98°E |
| | Kuwait | Kuwaiti | 29.42°N, 47.69°E |
| Southern Africa | Namibia | Namib | 21.24°S, 14.99°E |
| Eastern Asia | China | Gobi | 39.43°N, 105.67°E |
| North America | Arizona | Sonoran | 33.15 °N, 112.08°W |
| South America | Patagonia | Patagonia | 50.26°S, 71.50°W |
| Australia | Australia | Strzelecki | 31.33°S, 140.33°E |

**Table 3.** Chemical characterisation of the dust aerosols in $PM_{10.6}$ and $PM_{2.5}$ (in parentheses) size fractions. Columns 3 and 4 give the Si/Al and Fe/Ca elemental ratios obtained from X-Ray Fluorescence analysis. The uncertainty of each individual value is estimated to be 10%. Column 5 shows $MR_{Fe\%}$, the fractional mass of elemental iron with respect to the total dust mass concentration (uncertainty 10%). Column 5 reports $MR_{Fe\%}$, the mass fraction of iron oxides with respect to the total dust mass concentration (uncertainty 15%). For $PM_{2.5}$ the determination of the Si/Al ratio is impossible due to the composition of the filter- membranes (quartz)

~~Chemical characterisation of the dust aerosols in $PM_{10.6}$ and $PM_{2.5}$ (in parentheses) size fractions. Columns 3 and 4 report the Si/Al and Fe/Ca elemental ratios obtained from X-Ray Fluorescence analysis. The uncertainty on each individual value is estimated to be 10%. Column 5 reports $MR_{Fe\%}$, the fractional mass of elemental iron with respect to the total dust mass concentration (uncertainty 10%). Column 5 reports $MR_{Fe\text{-}ox\%}$, the mass fraction of iron oxides with respect to the total dust mass concentration (uncertainty 15%). For $PM_{2.5}$ the determination of the Si/Al ratio is impossible due to the composition of the filter medium.~~

| Geographical area | Sample | Si/Al | Fe/Ca | $MC_{Fe\%}$ | $MC_{Fe\text{-}ox\%}$ |
|---|---|---|---|---|---|
| Sahara | Morocco | 3.12 (---) | 0.24 (0.28) | 3.6 (4.4) | 1.4 (1.8) |
| | Libya | 2.11 (---) | 1.19 (1.12) | 5.2 (5.6) | 3.1 (3.4) |
| | Algeria | 2.51 (---) | 3.14 (4.19) | 6.6 (5.4) | 2.7 (2.2) |
| Sahel | Mali | 3.03 (---) | 2.99 (3.67) | 6.6 (33.6) | 3.7 (18.7) |
| | Bodélé | 5.65 (---) | 12.35 (----) | 4.1 (----) | 0.7 (----) |
| Middle East | Saudi Arabia | 2.95 (---) | 0.29 (0.27) | 3.8 (5.1) | 2.6 (3.5) |
| | Kuwait | 3.15 (---) | 0.89 (1.0) | 5.0 (13.6) | 1.5 (4.2) |
| Southern Africa | Namibia | 3.41 (---) | 0.11 (0.10) | 2.4 (6.9) | 1.1 (3.1) |
| Eastern Asia | China | 2.68 (---) | 0.77 (0.71) | 5.8 (13.6) | 0.9 (2.5) |
| North America | Arizona | 3.30 (---) | 0.95 (----) | 5.3 (----) | 1.5 (----) |
| South America | Patagonia | 4.80 (---) | 4.68 (4.64) | 5.1 (----) | 1.5 (---) |
| Australia | Australia | 2.65 (---) | 5.46 (4.86) | 7.2 (11.8) | 3.6 (5.9) |

**Table 4.** Mass absorption efficiency (MAE, $10^{-3}$ $m^2$ $g^{-1}$) and Ångström Absorption Exponent (AAE) in the $PM_{10.6}$ and $PM_{2.5}$ size fractions. Absolute errors are in brackets.

| | | **$PM_{10.6}$** | | | | | |
|---|---|---|---|---|---|---|---|
| **Geographical area** | **Sample** | **375 nm** | **407 nm** | **532 nm** | **635 nm** | **850 nm** | **AAE** |
| Sahara | Morocco | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) |
| | Libya | 89 (11) | 75 (9) | 30 (5) | --- (---) | --- (---) | 3.2 (0.3) |
| | Algeria | 99 (10) | 80 (10) | 46 (7) | 16 (3) | 15 (3) | 2.5 (0.3) |
| Sahel | Mali | --- (---) | 103 (18) | 46 (12) | --- (---) | --- (---) | --- (---) |
| | Bodélé | 37 (4) | 25 (3) | 13 (2) | 6 (1) | 3 (1) | 3.3 (0.3) |
| Middle East | Saudi Arabia | 90 (9) | 79 (8) | 28 (3) | 6 (1) | 4 (1) | 4.1 (0.4) |
| | Kuwait | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) | 2.8 (0.3) |
| Southern Africa | Namibia | 52 (7) | 49 (7) | 13 (3) | 5 (2) | 1 (2) | 4.7 (0.5) |
| Eastern Asia | China | 65 (8) | 58 (7) | 32 (4) | 8 (2) | 7 (2) | 3 (0.3) |
| North America | Arizona | 130 (15) | 99 (12) | 47 (7) | 21 (4) | 13 (4) | 3.1 (0.3) |
| South America | Patagonia | 102 (11) | 80 (9) | 29 (4) | 17 (2) | 10 (2) | 2.9 (0.3) |
| Australia | Australia | 135 (15) | 121 (13) | 55 (7) | 26 (4) | 14 (3) | 2.9 (0.3) |

| | | $PM_{2.5}$ | | | | | |
|---|---|---|---|---|---|---|---|
| **Geographical area** | **Sample** | **375 nm** | **407 nm** | **532 nm** | **635 nm** | **850 nm** | **AAE** |
| Sahara | Morocco | 107 (13) | 88 (11) | 34 (6) | 14 (3) | 15 (4) | 2.6 (0.3) |
| | Libya | 132(17) | 103 (14) | 33 (7) | --- (---) | --- (---) | 4.1 (0.4) |
| | Algeria | 95(8) | 71 (11) | 37 (7) | 12 (5) | 12 (5) | 2.8 (0.3) |
| Sahel | Mali | 711 (141) | 621 (124) | 227 (78) | --- (---) | --- (---) | 3.4 (0.3) |
| | Bodelé | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) |
| Middle East | Saudi Arabia | 153 (18) | 127 (15) | 42 (7) | 8 (4) | 6 (4) | 4.5 (0.5) |
| | Kuwait | 270 (100) | 324 (96) | --- (---) | 54 (52) | --- (---) | 3.4 (0.3) |
| Southern Africa | Namibia | 147 (36) | 131 (32) | 31 (21) | 6 (16) | 3 (15) | 5.1 (0.5) |
| Eastern Asia | China | 201 (30) | 176 (26) | 89 (17) | 14 (10) | 23 (10) | 3.2 (0.3) |
| North America | Arizona | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) |
| South America | Patagonia | --- (---) | --- (---) | --- (---) | --- (---) | --- (---) | 2.9 (0.3) |
| Australia | Australia | 335 (39) | 288 (33) | 130 (19) | 57 (11) | 36 (9) | 2.9 (0.3) |

**Table 5.** Mass absorption efficiency (MAE, $10^{-3}$ $m^2$ $g^{-1}$) and Ångström Absorption Exponent (AAE) from the ~~of~~ literature data discussed in the paper

| Geographical area | Sample | 266 nm | 325 nm | 428 nm | 532 nm | 660 nm | 880 nm | 1064 nm | AAE |
|---|---|---|---|---|---|---|---|---|---|
| Sahara | Morocco* | | | | | | | | 2.25–5.13 |
| | Morocco, $PM_{2.5}$£ | | | | | | | | 2.0–6.5 |
| | Morocco, submicron# | 1100 | | | 60 | | | 30 | 4.2 |
| | Egypt, submicron# | 810 | | | 20 | | | | 5.3 |
| | Tunisia$ | | 83 | | | 11 | | | |
| | Saharan, transported$^{\mu}$ | | | | | | | | 2.9 ± 0.2 |
| | Saharan, transported ($PM_{10}$)% | | | 37 | 27%% | 15%%% | | | 2.9 |
| | Saharan, transported ($PM_1$)% | | | 60 | 40%% | 30%%% | | | 2.0 |
| Sahel | Niger$ | | 124 | | | 19 | | | |
| Eastern Asia | China$ | | 69 | | | 10 | | | |
| | China& | | 87&& | 50&&& | 27&&&& | 13 | 1 | | 3.8 |
| Arabian Peninsula, N/NE Africa, Central Asia | Various locations@ | | | | | | | | 2.5-3.9 |

* Müller et al. (2008)
£ Petzold et al. (~~2008~~2009)
# Linke et al. (2006)
$ Alfaro et al. (2004)
$^{\mu}$ Fialho et al. (2005)
% Denjean et al. (2016); %% at 528 nm, %%% at 652 nm
& Yang et al. (2009); && at 375 nm, &&& at 470 nm, &&&& at 590 nm
@ Mossmüller et al. (2012)

**Figure 1.** Time series of aerosol mass concentration in the chamber for two companion experiments (Libyan dust). ~~Time series of aerosol mass concentration in the chamber for the two companion experiments (Libya sample)~~. Experiment 1 (top panel) was dedicated to the determination of the chemical composition (including iron oxides) by sampling on polycarbonate filters. Experiment 2 (bottom panel) was dedicated to the determination of the absorption optical properties by sampling on quartz filters.

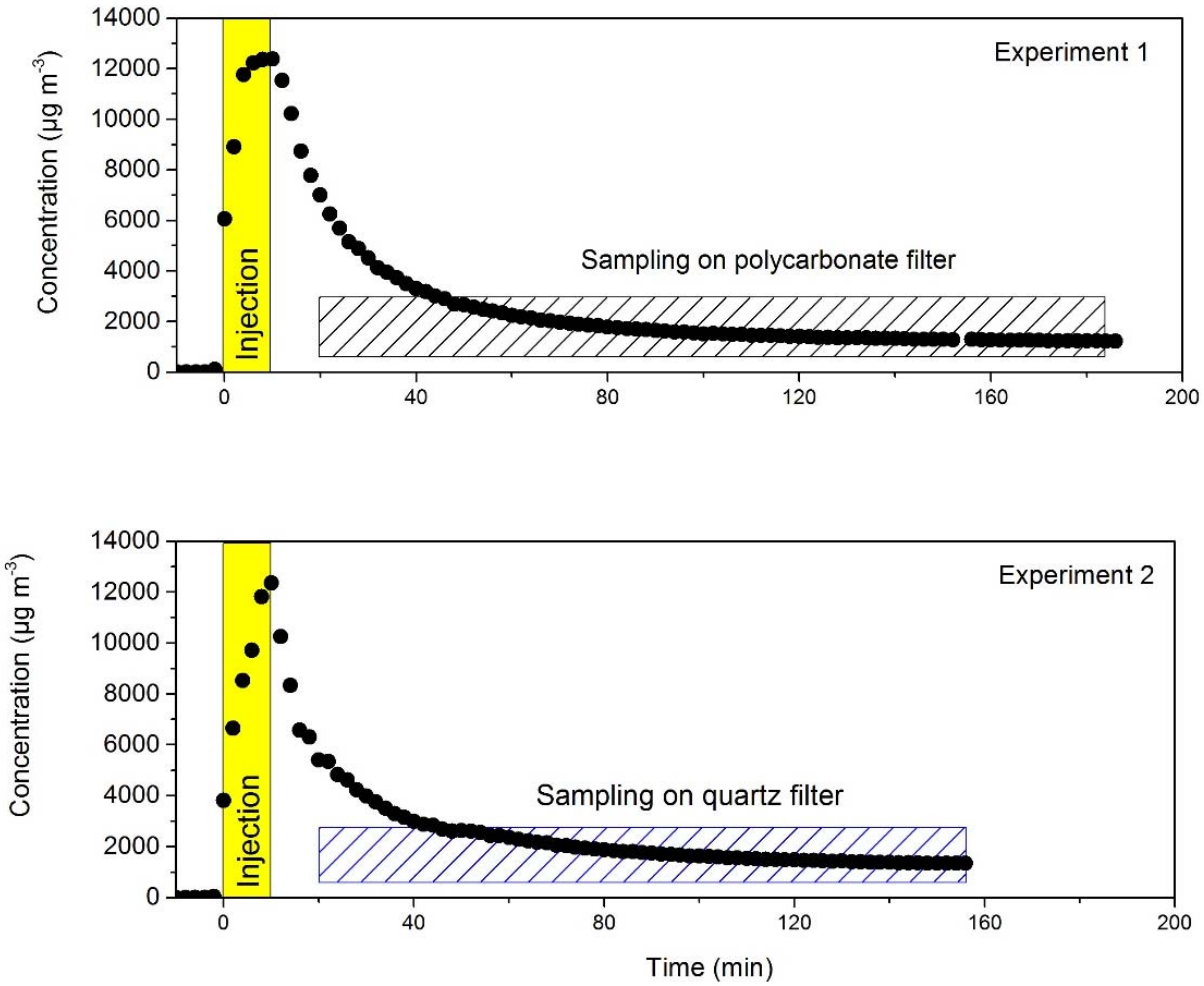


**Figure 2.** Locations (red stars) of the soil and sediment samples used to generate dust aerosols.

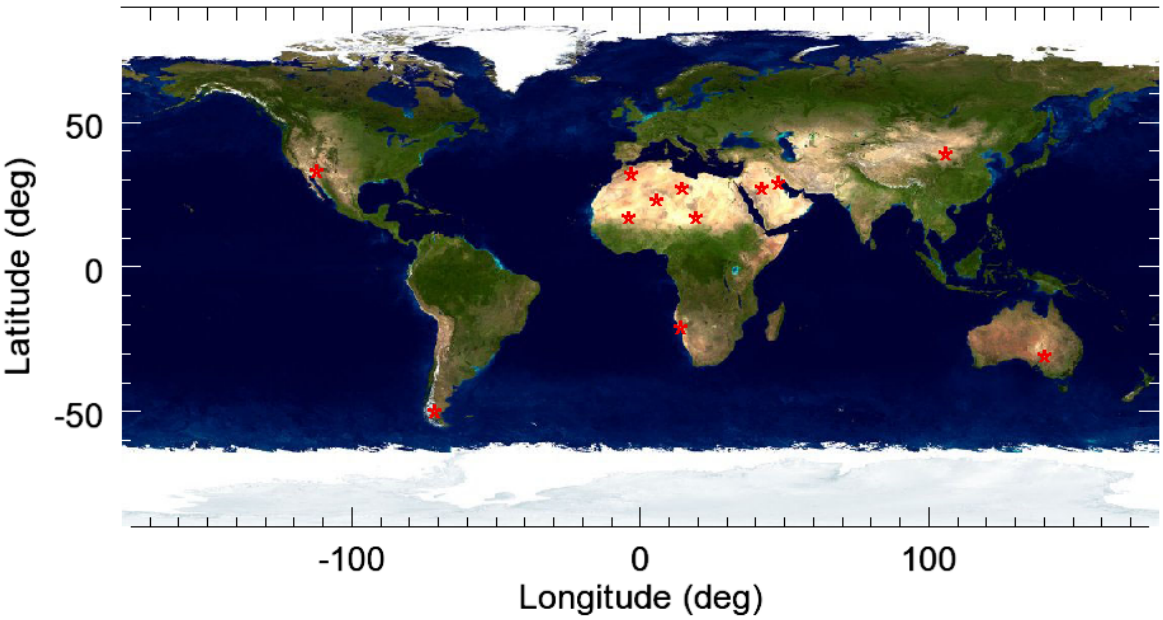


**Figure 3.** Spectral dependence of the MAE values for the samples investigated in this study in the $PM_{10.6}$ (left) and in the $PM_{2.5}$ (right) mass fractions.

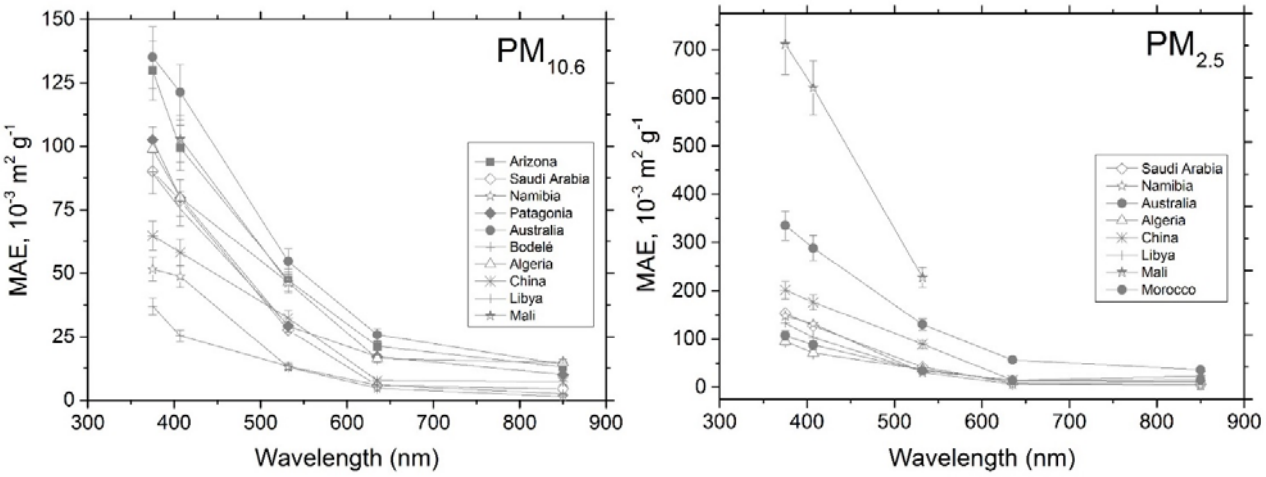
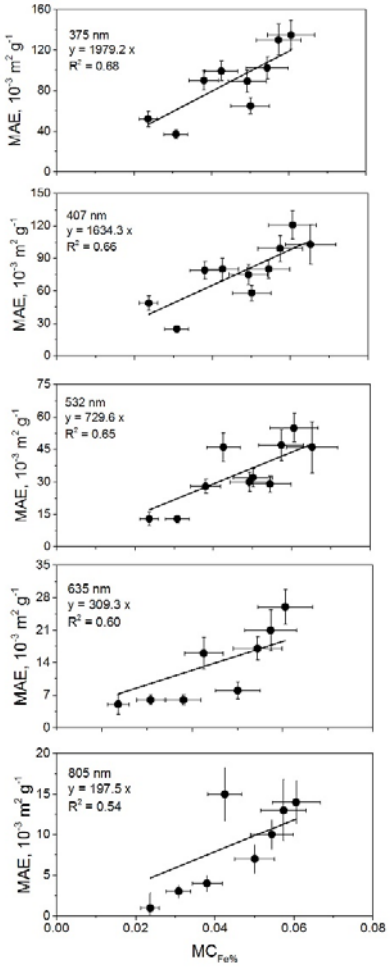
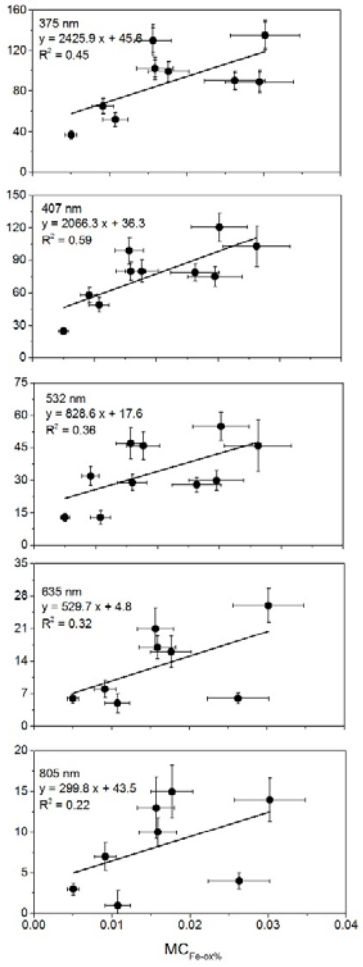
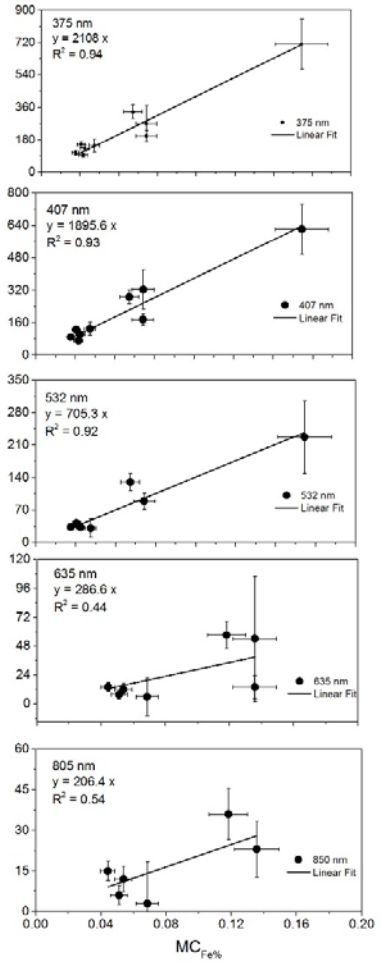


**Figure 4.** Illustration of the links between the MAE values and the dust chemical composition found in this study. Left column, from top to bottom: linear regression between the MAE values in the range from 375 to 850 nm and the fraction of elemental iron relative to the total dust mass ($MR_{Fe\%}$) in the $PM_{10.6}$ fraction; Middle column: same as left column but for the mass fraction of iron oxides relative to the total dust mass ($MR_{Fe\ ox\%}$) in the $PM_{10.6}$ size fraction; Right column: same as left column but in the $PM_{2.5}$ size fraction.

~~Illustration of the links between the MAE values and the dust chemical composition found in this study. Left column, from top to bottom: MAE values between 375 and 850 nm versus the fraction of elemental iron with respect to the total dust mass ($MR_{Fe\%}$) in the $PM_{10.6}$ fraction; Middle column: same as left column but versus the mass fraction of iron oxides to the total dust mass ($MR_{Fe\ ox\%}$) in the $PM_{10.6}$ size fraction; Right column: same as left column but in the $PM_{2.5}$ size fraction. The linear regression lines between MAE and $MR_{Fe\%}$ and MAE and $MR_{Fe\ Ox\%}$ are reported in each plot.~~

| $PM_{10.6}$ size fraction | | $PM_{2.5}$ size fraction |
|---|---|---|
| Spectral MAE values vs $MC_{Fe\%}$ | Spectral MAE values vs $MC_{Fe\text{-}ox\%}$ | Spectral MAE values vs $MC_{Fe\%}$ |
|  |  |  |