

We thank the anonymous referee #2 for his/her valuable comments to the manuscript and the constructive suggestions for improving the presentation quality. We have tried and addressed these comments at the best of our possibilities. Below, we explain how the comments and suggestions are addressed (responses in blue color).

Referee #2

I first would like to apologize with authors and the editor for the delay in submitting this review. This manuscript present a sensitivity study of the optical properties of dust mixtures including iron oxides to the values of their refractive index, to their state of mixing within the dust matrix and to the shape. The topic is surely of interest as iron oxides control the light-absorbing properties of mineral dust which ultimately control their direct and semi-direct radiative effect. Pointing out to the discrepancies in the current state of knowledge is surely beneficial in promoting new research in the area.

Nonetheless, the paper presents various weaknesses that must be addressed before considering publication. In particular, the paper presents a very large number of references. However, there is a great confusion and a great deal in simplification in their use, leading to misleading discussion and conclusions. The authors should pay more attention in separating research results that 1/ dealt with dust in the aerosol phase to those that dealt with dust in soils 2/ dealt with standard material (synthetic minerals, proxies.) to those that dealt with dust in the aerosol phase.

We agree to the referee that a very large number of references are presented in this paper, and some of them had been cited unreasonable and even in error. We have rechecked all references and revised them. We admit that we just want to fix the upper limit for iron oxides in dust aerosol. However, we know very little to how many iron oxides within dust in the aerosol phase are originated from the iron oxides within dust in soil. As the iron oxides emitted from the soil dust, and thus, we did not considering the specific forms of iron oxides in our original manuscript. After reading the suggestion from referee #2, we fully accept this good constructive suggestion and reorganized the related paragraphs to make our revised manuscript more understandable to readers, and make note of the changes we have made to the discussion paper.

Some specific comments:

- a) **Size distribution (paragraph 2.2): a model of size distribution is proposed. The authors do not consider a coarse mode, whereas, by their own admission, mineral dust is characterized by one. As a matter of fact, the coarse mode of mineral dust can contribute to lower the single scattering albedo of dust. What is the competition with the value of the complex refractive index of the iron oxides?**

The editor and both two reviewer have pointed out the setting of size distribution is not well justified. We are agreed to the 3 modes or more modes for dust size distribution. In this study, we want to fix the optical effects of iron oxides in dust aerosol, and thus we want to weaken the effects of size distribution. With reference to

the OPAC, We simply choose one mode of dust to calculate the optical properties. Although this choose may be ideal with compare to the field observations and recent knowledge on size distribution of dust (Osborne et al., 2008; Mahowald et al., 2014). More recently, a modified size distribution with only one mode following brittle fragmentation theory had been published by Kok et al. (2011), but we are not agree to this theory and we are agree to the old multi-modes for dust size distribution. More detailed formula proving, please see http://editor.copernicus.org/index.php?_mdl=msover_md&_jrl=10&_lcm=oc108lcm109w&_acm=get_comm_sup_file&_ms=28358&c=85559&salt=1619220327230502394. Although previous studies on mineral dust size distributions have often attributed three to five lognormal modes, the median radii, magnitudes and standard deviations of the modes vary from study to study owing to the time-varying nature of dust particles and the source regions. In a word, one mode size distribution was chosen to simplified calculation, to compare with Sokolik et al (1999) and to help better understanding for different readers.

With considering the advices of (2) and (3), we reviewed the reported values by several dust campaigns and AERONET observations, and then compared them with the calculated SSA curve in Figure 4b.

What is the competition of coarse size for dust with the value of the complex refractive index of the iron oxides? Figure 4b is shown that the SSAs were mostly ranged in 0.95-0.99 from the observation during different campaigns, but much lower (0.944-0.95) for the AERONET which cannot exclude the presence of black carbon with higher absorbing. Our calculated result could consistent with the higher part (0.97-0.99) of measured SSAs, but higher than the lower part (0.95-0.97) due to the effect of coarse particles during different dust campaigns. It is hard to compare the calculated the SSAs with measured values by considering the varied median radii, magnitudes and standard deviations of the modes, and even illustrated them in the same figure.

Mahowald N, Albani S, Kok J F, et al. The size distribution of desert dust aerosols and its impact on the Earth system[J]. *Aeolian Research*, 2014, 15: 53-71.

Kok J F. A scaling theory for the size distribution of emitted dust aerosols suggests climate models underestimate the size of the global dust cycle[J]. *Proceedings of the National Academy of Sciences*, 2011, 108(3): 1016-1021.

b) Goethite/hematite ratios: the authors have to distinguish between quantitative (such as those presented in Formenti et al. 2014 using X-ray absorption) and qualitative measurements such as those presented in Formenti et al. 2008, Lafon et al. (2006) and Klaver et al (2011) using diffuse reflectance spectroscopy.

Thanks for the suggestion. The speciation of iron oxides in dust aerosol could be identified by X-ray absorption (XAS) in the Fe K range: XANES (X-ray absorption near-edge structure) and EXAFS (extended X-ray absorption fine structure). XAS spectroscopy is based on the analysis of the position and shape of the K pre-edge and edge peaks, depending on the oxidation state of iron but also on the atomic position of the neighbouring atoms, mostly O⁺ and OH⁻. The XANES

analysis can identify also other Fe-minerals depending on the inflection points of their absorption spectra, such as magnetite. The position of the inflection points is very similar for goethite, hematite and magnetite; the relative proportions of the peaks in the absorption spectra are different. It should not be forgotten that, as discussed by Lazaro et al. (2008), magnetite accounts for less than 7% of the total iron in aerosol dust samples. This is of the same order of magnitude that the errors of the fit taking into account the differences between the minerals in their standard and in their real forms. The quantification of the Fe status was based on the analysis of five standards of Fe(III)-bearing minerals which can be found in mineral dust (Formenti et al., 2014b). The final obtained result is quantitative; the ratio of goethite/hematite is also quantitative.

DRS use the characteristic bands of hematite and goethite in the visible absorption spectrum at around 565 nm and 435 nm, respectively. Moreover, by taking the second derivative of the diffuse reflectance signal, it is possible to determine quantitatively a relative mass fraction of hematite and goethite (Lafon et al., 2006). Thus, the reported results from diffuse reflectance spectroscopy are also quantitatively relative ratios for goethite/hematite. So we compiled the results from XAS with DRS in Table 3 of our manuscript.

Lazaro, F. J., et al., The speciation of iron in desert dust collected in Gran Canaria (Canary Islands): Combined chemical, magnetic and optical analysis, *Atmospheric Environment* 42 (2008) 8987–8996

Lafon, S., Sokolik, I., Rajot, J., Caquineau, S., and Gaudichet, A.: Characterization of iron oxides in mineral dust aerosols: Implications for light absorption, *J. Geophys. Res.*, 111, D21207, doi:10.1029/2005JD007016, 2006.

c) Abundance of iron and iron oxides (paragraph 3.2.2) c.1/Page 15, lines 20 on. Not all the references you mention used XRD to quantify the iron oxides c.2/Page 16, line 9. Formenti et al. 2014 did NOT use diffuse reflectance spectroscopy to obtain the hematite-to-goethite ratio.

Response to C.1:

We carefully recheck the references, and found that:

Bernabé et al. (2005) had used XRD to identify the mineralogy of deposited atmospheric dust and dust aerosol, and reported that hematite presented only within the sedimentable dust particles but without reported on the abundance of hematite. Thus, we delete this reference from our citation.

All other cited references had used XRD to quantify the iron oxides, and we simply introduced as following:

Shi et al. (2005) quantified the abundance of hematite (0.9% in mass) in PM10 sample that collected during an Asian dust storm.

Linke et al. (2006) reported that no detectable iron oxides phases in Cairo samples and ~0.2 wt-% of goethite in the Agadez sample and 0.6±0.1 wt-% of hematite in the Morocco sample.

Kandler et al. (2009) reported their quantified results (1 wt%) for hematite in Morocco dust sample based on XRD with RIR method.

Lawrence et al. (2010) illustrated the abundance (0.1-0.8 wt%) of goethite in deposited dust of Colorado sample based on XRD with the RockJock software.

Klaver et al. (2011) measured the mass content (0.6-1.8 wt %) of iron oxides in transported dust samples collected by aircraft during GERBILS campaign and reported their detailed results in

Table V of their research paper.

Wagner et al. (2012) shown that the iron oxide content (hematite and/or goethite) does not exceed 1 wt % in entrained dust aerosol from soil dust samples collected during SAMUM-1 campaign. In contrast, the Burkina Faso sample collected during AMMA campaign shown a very high content of iron oxides (15 wt %) with a hematite to goethite ratio of 4:1. All samples were analyzed by XRD with the RIR method.

Finally, we add the new XRD analysis of Formenti et al. (2014b).

Formenti, P., Caquineau, S., Desboeufs, K., Klaver, A., Chevaillier, S., Journet, E., and Rajot, J.: Mapping the physic-chemical properties of mineral dust in western Africa: mineralogical composition, *Atmos. Chem. Phys.*, 14, 10663–10686, doi:10.5194/acp-14-10663-2014, 2014b.

Response to C.2:

We are sorry for this obviously error ciation, and this reference had been deleted in revised manuscript.

Furthermore, Formenti et al. (2014) in line 4 of page 5635 is also error cited due to our editing for the list of references. The correct reference is:

Formenti, P., Caquineau, S., Desboeufs, K., Klaver, A., Chevaillier, S., Journet, E., and Rajot, J.: Mapping the physic-chemical properties of mineral dust in western Africa: mineralogical composition, *Atmos. Chem. Phys.*, 14, 10663–10686, doi:10.5194/acp-14-10663-2014, 2014b.

Following is our revised manuscript in PDF format.

What's the real role of iron-oxides in the optical properties of dust aerosols?

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Abstract:

Iron oxides compounds constitute an important component of mineral dust aerosol. Several previous studies have shown that these minerals are strong absorbers at visible wavelengths and thus that they play a critical role in the overall climate perturbation caused by dust aerosol. When compiling a database of complex refractive indices of possible mineral species of iron-oxides to study their optical properties, we found that uniformly continuous optical constants for a single type of iron-oxides in the wavelength range between 0.2 μm and 50 μm is very scarce and that the use of hematite to represent all molecular or mineral iron-oxides types is a popular hypothesis. However, the crucial problem is that three continuous datasets for complex refractive indices of hematite are employed in climate models, but there are significant differences between them. Thus, the real role of iron-oxides in the optical properties of dust aerosols becomes a key scientific question, and we address this problem by considering different refractive indices, size distributions, and more logical weight fractions and mixing states of hematite. Based on the microscopic observations, a semi-external mixture that employs an external mixture between Fe-aggregates and other minerals and partly internal mixing between iron-oxides and aluminosilicate particles is advised as the optimal approximation. The simulations demonstrate that hematite with a spectral refractive indices from Longtin et al. (1988) shows approximately equal absorbing capacity to the mineral illite over the whole wavelength region from 0.55 μm to 2.5 μm , and only enhances the optical absorption of aerosol mixture at $\lambda < 0.55 \mu\text{m}$. Using the dataset from Query (1985) may overestimate the optical absorption of hematite at both visible

34 and near-infrared wavelengths. More laboratory measurements of the refractive index of
35 iron-oxides, especially for hematite and goethite in the visible spectrum, should therefore be taken
36 into account when assessing the effect of mineral dust on climate forcing.

37

38 **1. Introduction**

39 Iron oxides in dusts are now identified as being important component for a number of climatic,
40 environmental and biological processes. Over the past decade, iron-oxide minerals have been
41 shown to be able to strongly absorb solar radiation (Tegen et al., 1997; Sokolik and Toon, 1999;
42 Lafon et al., 2004; Qin and Mitchell, 2009; Redmond et al., 2010), and thus have a direct impact
43 on the Earth's radiation balance (Balkanski et al., 2007; Scanza et al., 2015; Smith and Grainger,
44 2014). Based on the ability to absorb acidic gases and water vapor (Baltrusaitis et al., 2007;
45 Wijenayaka et al., 2012; Song and Boily, 2013), iron oxides also contribute to heterogeneous
46 reactions and cloud processes (Shi et al., 2011; Dupart et al., 2012), further influencing the
47 radiation balance. In particular, layers of dust on snow and ice cover accelerate the melting of
48 snow and ice by diminishing the surface albedo (Painter et al., 2010; Ginot et al., 2014) and the
49 heat-absorbing properties of iron oxides in these dust layers can add to this effect (Kaspari et al.,
50 2014; Reynolds et al., 2013; Dang and Hegg, 2014). Moreover, the deposited iron-bearing dust
51 aerosols provide critical nutrients to marine and terrestrial ecosystems, which associated with
52 consequential important drawdown of atmospheric carbon dioxide (Jickells et al., 2005; Shao et al.,
53 2011; Nickovic et al., 2013). However, these effects can lead to either positive or negative net
54 radiative perturbation depending mostly on the underlying surface albedo, vertical profile (optical
55 depth and height of dust layer), particle size distribution and mineralogy (Liao and Seinfeld, 1998;
56 Calquin et al., 1999). This large uncertainty results from our limited knowledge of the physical,
57 chemical and optical properties of atmospheric iron oxides on various space and time scales
58 (Tegen et al., 1997; Sokolik et al., 2001; Formenti et al., 2011).

59 The element iron can be found among numerous mineralogical species, such as feldspars,
60 clays (e.g. illite, smectite, chlorite and biotite), iron-oxides, iron-hydroxides and so on. A useful
61 mineralogical classification frequently used in soil science distinguishes two categories of iron: (i)
62 "structural iron" (in either the Fe(II) or Fe(III) oxidation states), trapped in the crystal lattice of
63 aluminosilicate minerals; and (ii) iron (in the Fe(III) oxidation state), in the form of discrete oxide

64 or hydroxide particles (Lafon et al., 2004). According to the classical terminology of soil scientists
65 (Sumner, 1963; Anderson and Jenne, 1970; Angel and Vincent, 1978), the latter kind of iron will
66 be referred to as free-iron and its corresponding oxides and hydroxides as iron-oxides. Ten of 16
67 known iron oxides, hydroxides and oxide-hydroxides are known occur in nature, with goethite,
68 hematite and magnetite being the most abundant as rock-forming minerals; ferrihydrite,
69 maghemite and lepidocrocite being intermediately abundant in many locations; and wüstite,
70 akaganéite, ferroxhyte, and bernalite being the least abundant (Cornell and Schwertmann, 2006;
71 Guo and Barnard, 2013). Dust aerosols from arid and semi-arid regions typically contain goethite,
72 hematite, ferrihydrite and magnetite, and based on the mass contribution, hematite and goethite
73 are the major components of free-iron in the atmospheric dust aerosols (Schroth et al., 2009; Shi et
74 al., 2012; Takahashi et al., 2013). Hematite (Fe_2O_3) is very common in hot, dry soils and imparts a
75 red color to its sediments. Goethite ($\alpha\text{-FeOOH}$) is a common weathered product in soils and
76 loesses. It occurs in moist, acidic soils (Schwertmann, 1993), and is brown to yellow in color.

77 Sokolik and Toon (1999) found that hematite is an especially strong absorber at ultra-violet
78 (UV) and visible wavelengths, and it can also enhance the absorption of clay minerals and quartz
79 through the formation of aggregates. Derimian et al. (2008) mentioned that iron oxides (primarily
80 hematite and goethite) only affect the optical absorbing ability of aeolian dust at short wavelengths
81 (the blue spectral region). Since the importance of hematite relative to other dust mineral
82 components was discussed more fully in the study of Sokolik and Toon (1999), most subsequent
83 modeling studies have assumed the iron-oxides in dust aerosols to be in the form of hematite. An
84 opposite viewpoint was put forward, however, by Balkanski et al. (2007) who have argued that the
85 optical absorption of dust with hematite at visible wavelengths might be lower than previous
86 thought. The main reason is that internal mixing rule calculations with a hematite content of 1.5
87 volume % (or 2.8 weight %) was supposed to be representative for median dust absorption and
88 was, as well, consistent with the AERONET measurements.

89 However, recent measurements on dust samples from east Asia, northern Africa and western
90 Africa (Lafon et al., 2006; Formenti et al., 2008; Reynolds et al., 2014) indicated that goethite was
91 presented in higher concentration than hematite (i.e. the ratio of goethite to hematite is about 7:3, a
92 result that will be detail introduced in Section 3.2.2). For dust in snow, goethite is also the
93 dominant ferric oxide that detected by reflectance spectroscopy and thus appears to be the main

94 iron-oxide control on absorption of solar radiation (Reynolds et al., 2013). More recently hematite
95 and goethite has been taken into account interactively in global climate simulations due to the
96 availability of global mineralogical distribution maps (Nickovic et al., 2012; Journet et al., 2014).

97 Furthermore, Müller et al. (2009) have measured the spectral imaginary refractive indices of
98 hematite over the wavelengths from 590 nm to 790 nm during laboratory experiments, but these
99 derived values are much lower than the data that employed in Sokolik and Toon (1999). The
100 theoretical simulations of optical scattering of hematite and goethite at 470, 550, and 660 nm by
101 Meland et al. (2011) also found that differences are apparent for hematite in both the phase
102 function and polarization results at 660 nm where the imaginary indices from different references
103 differ. Additionally, two studies that refer to optical properties of hematite in dust samples have
104 also argued that the imaginary values of hematite refractive index in Sokolik and Toon (1999) are
105 more than a factor of 2 larger than those reported by Bedidi and Cervelle (1993) and Longtin et al.
106 (1988) at wavelengths below 600 nm (Moosmüller et al., 2012; Wagner et al., 2012).

107 Thus, what is the real role of iron-oxides in determining the overall impact of the optical
108 properties of dust aerosols? This study will focus on investigating this important scientific
109 question by considering heterogeneous optical refractive indices, mixing states and more logical
110 abundance of iron-oxides.

111

112 **2. Method and simulation**

113 **2.1 Complex refractive index**

114 The complex refractive index (optical constant) is the most basic and significant parameter
115 for calculating the optical properties of aerosols, but values for the optical constants of hematite
116 in the wavelength range 0.2 to 50 μm are scarce in published references. Table 1 compiles
117 information about the complex refractive indices of the major constituents of free-iron that we
118 have found in the published literature. Sokolik and Toon (1999) employed the refractive indices of
119 hematite from Querry et al. (1978), but Querry et al. (1978) mainly studied the optical constants of
120 limestone and, as far as we can discover, do not contain any work on hematite at all. Since the
121 publication of Sokolik and Toon (1999), studies (Höller et al., 2003; Alfaro et al., 2004; Muñoz et
122 al., 2006; Mishra and Tripathi, 2008; Otto et al., 2009; Meland et al., 2011; Wagner et al., 2012)
123 have misquoted the data as Querry et al. (1978) or Querry (1987) when modeling the optical

124 properties of hematite. Actually, Querry (1987) is entitled “Optical constants of minerals and other
125 materials from the millimeter to the UV” and gives optical constants of 29 materials, but still
126 without referring to hematite. After careful searching, we have found that Querry (1985) has
127 tabulated values for the refractive index ($m = n + ki$) of hematite. Moreover, Longtin et al.
128 (1988) also reported spectral refractive indices for hematite from earlier measurements by Steyer
129 (1974), Onari et al. (1977), Galuza et al. (1979) and Kerker et al. (1979). This dataset also has
130 been used in modeling the contribution of hematite to the optical properties of atmospheric dust
131 aerosols in recent studies (Klaver et al., 2011a; Köhler et al., 2011; Hansell et al., 2011).
132 Recently, the third unpublished continuous refractive indices of hematite (named TA2005 in Table
133 1) from the Aerosol Refractive Index Archive (ARIA) of Oxford University have been employed
134 in the Community Atmosphere Model (Scanza et al., 2015). Because this work has not been
135 peer-reviewed and because TA2005 and QE1985 show a similar wavelength dependency we pay
136 special attention to the differences of refractive indices between QE1985 and LG1988 in this paper.
137 Any errors due to uncertainties in the optical constants would be directly reflected in the
138 calculated results referred to above.

139
140
141

Table 1. Summary of the published complex refractive indices for major constituents of free-iron at different wavelengths (with their references).

| Iron-oxide species | Wavelength (μm) | Reference source | Abbreviation |
|--------------------|------------------------------|--------------------------------|--------------|
| Hematite | 8.3-50 | Popova et al. (1973) | PV1973 |
| Hematite | 1.0-333 | Onari et al. (1977) | OA1977 |
| Hematite | 0.25-0.7 | Shettle and Fenn, (1979) | SF1979 |
| Hematite | 0.35-0.65 | Hsu and Matijevic, (1985) | HM1985 |
| Hematite | 0.21-90 | Querry (1985) | QE1985 |
| Hematite | 0.3-300 | Longtin et al. (1988) | LG1988 |
| Hematite | 0.2-0.7 | Gillespie and Lindberg, (1992) | GL1992 |
| Hematite | 0.2-4.5 | Krekov (1992) | KE1992 |
| Hematite | 0.35-0.75 | Bedidi and Cerville (1993) | BC1993 |
| Hematite | 0.1-1000 | Triaud (2005)* | TA2005 |
| Hematite | 5-50 | Marra et al. (2005) | MR2005 |
| Hematite | 5-2000 | Glotch and Rossman, (2009) | GR2009 |
| Hematite | 0.59-0.79 | Müller et al. (2009) | ML2009 |
| Magnetite | 0.21-55 | Querry (1985) | QE1985 |
| Magnetite | 5-50 | Mukai (1989) | MK1989 |
| Magnetite | 0.25-0.7 | Gillespie and Lindberg, (1992) | GL1992 |
| Magnetite | 0.1-1000 | Amaury et al. (2002) | AU2002 |
| Magnetite | 5-100 | Glotch and Rossman, (2009) | GR2009 |

| | | | |
|----------|-----------|------------------------------|--------|
| Goethite | 0.45-0.75 | Bedidi and Cerville, (1993) | BC1993 |
| Goethite | 8-50 | Glotch and Roman, (2009) | GR2009 |
| Wüstite | 0.2-500 | Henning (1995) | HN1995 |
| Wüstite | 10-500 | Henning and Mutschke, (1997) | HN1997 |

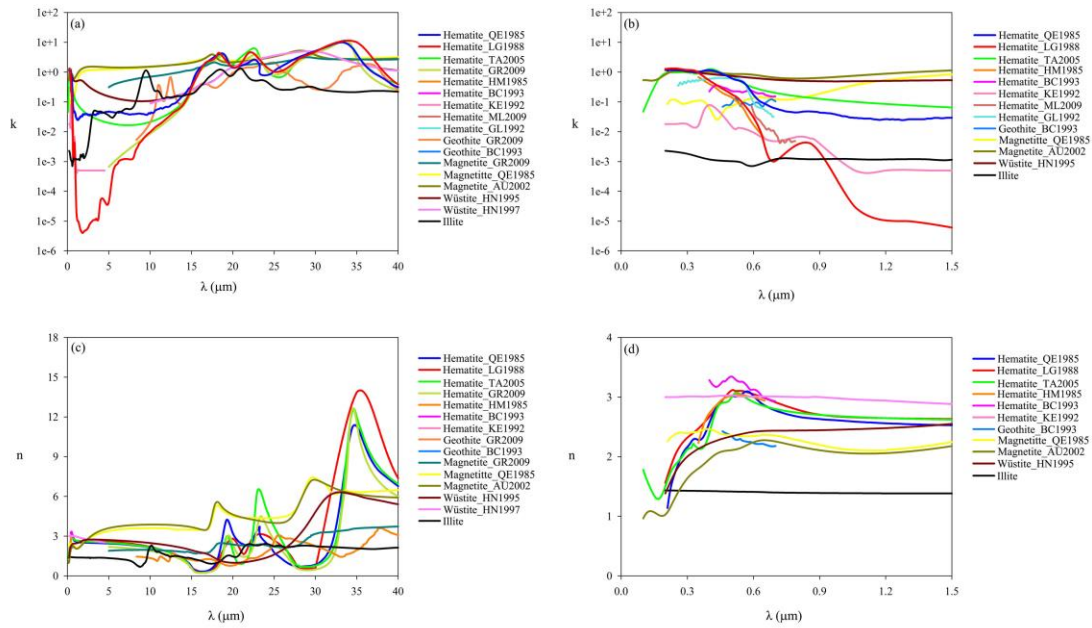
142 * The data was taken from <http://www.atm.ox.ac.uk/project/RI/hematite.html>

143 Bedidi and Cerville (1993) also presented refractive indices for hematite and goethite
 144 derived from reflectance measurements at wavelengths of 350-750 nm. Glotch and Rogers (2007)
 145 reported the optical constants of hematite, goethite and magnetite over the infrared (IR)
 146 wavelengths, and Hsu and Matijević (1985) also measured the refractive indices of hematite in the
 147 wavelengths of 350-650 nm.

148 Hematite is a uniaxial crystal which crystallizes in the trigonal system, whose optic axis
 149 corresponds to the crystallographic c -axis. Perpendicular to the c -axis are two radial a -axes. The
 150 dielectric constants of bulk hematite must therefore be measured for two principal polarizations of
 151 the incident light, namely one with the electric vector in any direction perpendicular to the c -axis
 152 (the so-called ordinary ray or – O ray) and the other with the electric vector along the crystalline
 153 c -axis (the extraordinary ray or – E ray). In this work we have calculated the average refractive
 154 indices for anisotropic hematite from all references in Table 1. The formula used is adopted from
 155 Longtin et al. (1988) and is:

$$156 \quad m_{avg} = (2n_{E-ray} / 3 + n_{O-ray} / 3) + (2k_{E-ray} / 3 + k_{O-ray} / 3)i.$$

157 The reported values for the complex refractive index from the sources in Table 1 are
 158 markedly different, especially the imaginary part which controls the optical absorption. In order to
 159 visually demonstrate the variation of optical constants from different references, values of the real
 160 (n) and logarithmic values of imaginary parts (k) are shown in Figure 1.



161
 162 **Fig. 1.** Spectral distribution of the imaginary and real parts of the complex refractive index for different iron oxides
 163 from Table 1 at wavelengths of 0 to 40 μm (Panels (a) and (c)) and expanded for wavelengths of 0 to 1.5 μm
 164 (Panels (b) and (d)).

165

166 For the real part of the refractive index for iron-oxides, there is a reasonable agreement
 167 between the hematite and magnetite datasets from the different references (Figure 1c). Because the
 168 real refractive index of hematite shows large fluctuations at wavelengths longer than 18 μm due to
 169 anisotropic refraction, the agreement between the different datasets decreases at these wavelengths.
 170 For goethite we are aware of only two sets of optical constants: one at visible wavelengths from
 171 Bedidi and Cervelle (1993) and the other at IR wavelengths from Glotch and Rogers (2007), but
 172 the wavelength gap between these two datasets hampers continuity. Unfortunately, Meland et al.
 173 (2011) have checked the former dataset for goethite using simulations according to Mie and
 174 T-Matrix theories and show that it may be in error. Nevertheless, we can see that goethite has
 175 optical constants similar to hematite. The real refractive index of hematite is larger than that of
 176 magnetite at wavelengths less than 2 μm , but is smaller between 2 and 33 μm (Figures 1c&d).

177 For the imaginary part of the refractive index of iron-oxides, hematite and goethite have
 178 different optical properties at short wavelengths, both in terms of magnitude and spectral
 179 dependence (Bedidi and Cervelle, 1993). Between 460 and 700 nm the imaginary part of the
 180 complex refractive index (representing absorption) of goethite is up to 3 times smaller than that of
 181 hematite. As a consequence, the proportions of hematite and goethite in mineral dust can

182 potentially change the magnitude and the spectral dependence of shortwave absorption of mineral
 183 dust. However, the limited and discontinuous refractive indices of goethite have constrained the
 184 evaluation of the effects of specific compositions of goethite and hematite to dust optical
 185 properties and solar radiation balance over broader wavelength ranges.

186 From Figure 1a, we clearly see that the k values for hematite from QE1985 and from
 187 LG1988 show significant differences for wavelengths between 650 nm and 15 μm . These
 188 differences are present at visible wavelengths and disappear at ultraviolet wavelengths, but the two
 189 datasets have similar trends at UV and visible wavelengths (Figure 1b). Note that the hematite
 190 optical constants vary dramatically across the visible wavelengths. In particular, the imaginary
 191 part of the index for hematite shows a sharp decrease with increasing wavelength in the red. As a
 192 consequence there is a large variability in the imaginary refractive index values for hematite taken
 193 from different published references, particularly at 2 μm where the values from different sources
 194 differ by a factor of 8600. Thus, this study firstly focuses on what will be the result if these
 195 heterogeneous optical constants of hematite are used as input for the calculation of radiation
 196 transfer models.

197

198 **2.2 Particle size distribution**

199 Size distribution is another important factor that affects the optical properties of particles.
 200 Because Sokolik and Toon (1999) has employed the refractive index dataset for hematite from
 201 QE1985 to calculate the radiative properties, we adopt here the same particle size distribution but
 202 with the refractive index dataset for hematite from LG1988 to compare our results with Sokolik
 203 and Toon (1999). The lognormal distribution is applied to dust aerosols:

$$204 \quad n_n(\ln r) \equiv \frac{dN}{d \ln r} = \frac{N_0}{\sqrt{2\pi} \ln \sigma} \exp \left[-\frac{1}{2} \left(\frac{\ln r - \ln r_0}{\ln \sigma} \right)^2 \right],$$

205 where r_0 is the median radius, σ is the geometric standard deviation, and N_0 is the total
 206 particle number density of the component in particles per cubic centimeter.

207 The optical properties of minerals are calculated on the assumption that they have one size
 208 mode but varying median radius. The particle size modes are selected as $r_0 = 0.5$ and $0.7 \mu\text{m}$, and
 209 $\sigma = 2.0$. The size mode with median radius $r_0 = 0.5 \mu\text{m}$ is believed to be representative of the

210 particle size distribution of the long-lived, long-distance-transport mode of airborne dust
211 (Patterson and Gillette, 1977; Arimoto et al., 1997). The larger r_0 is representative of a particle
212 size mode which occurs near the dust source (Gomes and Gillette, 1993). In reality, the size
213 distribution of dust aerosols can have one or several modes, characterized by a specific
214 composition (Mahowald et al., 2013).

215

216 **2.3 Theoretical simulations**

217 Images from scanning electron microscopy (SEM) reveal non-spherical, irregular and
218 compact shapes of the dust particles (Figure 3), but Otto et al. (2009) and Klaver et al. (2011b)
219 have shown that spherical/non-spherical differences only influence the single scattering albedo by
220 less than 1%. Meland et al. (2011) have also shown that moderate departures from spherical shape
221 are relatively unimportant in determining the scattering matrix for particles with high refractive
222 index values, such as hematite. Therefore, we expect the aerosol asphericity to have a negligible
223 impact on our calculated results of optical properties and subsequent calculations using the Mie
224 theory (which assumes a spherical morphology for the dust particles).

225 There are several different computer codes that can be used to compute optical properties
226 for a lognormal particle size distribution. The theoretical light scattering simulations in this paper
227 have used the MieTab software. MieTab uses a FORTRAN code with continued fraction
228 modification produced by W. J. Lentz from the Mie code originally developed by Dave and Center
229 (1968). This modified code can be obtained from
230 <http://diogenes.iwt.uni-bremen.de/vt/laser/codes/ddave.zip>. In order to validate the accuracy of
231 MieTab, we firstly compared it with a double precision Lorenz-Mie scattering code and a double
232 precision T-Matrix code for a lognormal particle size distribution from Mishchenko et al. (2002).
233 The double precision Lorenz-Mie and T-Matrix codes are available from
234 http://www.giss.nasa.gov/staff/mmishchenko/t_matrix.html.

235 In addition to the wavelength dependent optical constants and the size distribution, the
236 T-Matrix theory also requires assumptions about the particle shape. In this work we use an aspect
237 ratio of 1.000001 to represent a spherical particle shape, because use of an aspect ratio exactly
238 equal to 1 causes computational overflow in some cases. The calculated results from the three
239 codes at different wavelengths and complex refractive indices for the same size distribution are

240 listed in Table 2. The good agreement of the results from the three codes demonstrates that the
 241 possibility of computational error affecting the interpretation of the calculated optical properties of
 242 iron-oxides can be neglected.

243

244 **Table 2.** Comparison of simulated optical properties between MieTab, Lorenz-Mie and T-matrix methods.

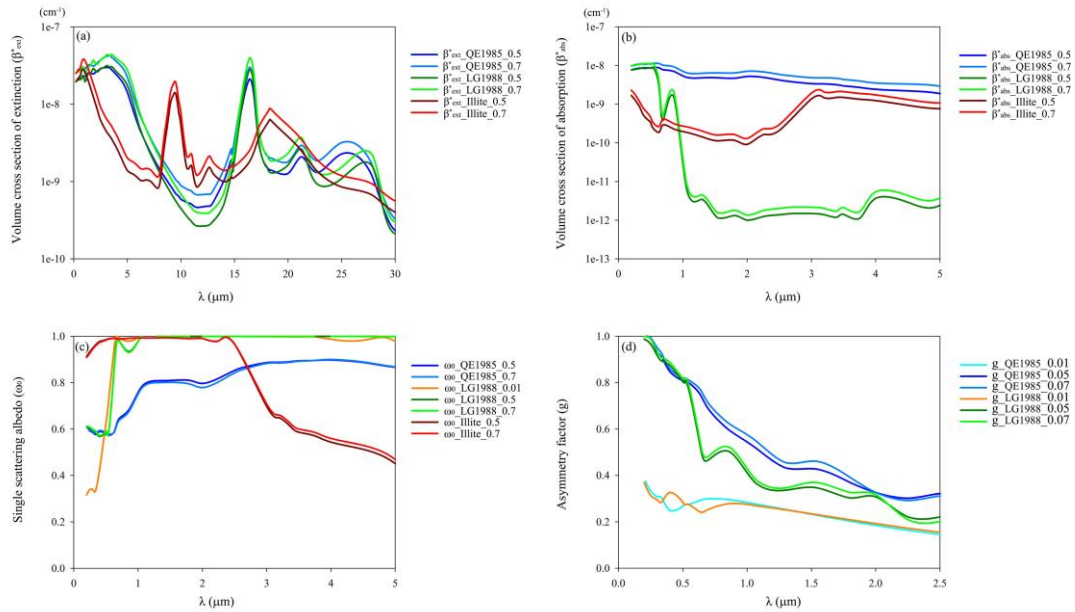
| Wavelength | 550 nm | | | 633 nm | | | 1060 nm | | |
|------------|-------------------|----------|------------|--------------------|----------|------------|-------------------|----------|------------|
| | n=3.102, k=0.0925 | | | n=3.007, k=0.00974 | | | n=2.66, k=0.00003 | | |
| $m=n+ki$ | | | | | | | | | |
| Code type | MieTab | T-matrix | Lorenz-Mie | MieTab | T-matrix | Lorenz-Mie | MieTab | T-matrix | Lorenz-Mie |
| Q_{ext} | 2.0950 | 2.1332 | 2.1637 | 2.1440 | 2.1917 | 2.2585 | 2.3250 | 2.5042 | 2.0226 |
| Q_{sca} | 1.2640 | 1.3284 | 1.3233 | 1.8500 | 1.9048 | 1.9587 | 2.3240 | 2.5033 | 2.0216 |
| Q_{abs} | 0.8310 | 0.8048 | 0.8404 | 0.2940 | 0.2870 | 0.2998 | 0.0010 | 0.0009 | 0.0010 |
| ω_0 | 0.6033 | 0.6227 | 0.6116 | 0.8629 | 0.8691 | 0.8673 | 0.9996 | 0.9996 | 0.9995 |

245

246 3. Results and discussion

247 3.1 Basic optical properties

248 We focus here on modeling the spectral optical properties of iron-oxides which are needed
 249 for climate modeling: the volume extinction coefficient β_{ext} (which is the sum of the scattering
 250 coefficient β_{sca} and the absorption coefficient β_{abs}), the single scattering albedo ω_0 , and the
 251 asymmetry parameter g (a cosine weighted integral of the scattering phase function). This set of
 252 parameters allows the calculation of radiation forcing in most climate models. Figure 2 shows
 253 calculated optical parameters for hematite (with complex refractive indices from QE1985 and
 254 LG1988) and illite with varying median radius at solar and infrared wavelengths. The volume total
 255 extinction coefficients β_{ext} have been normalized as β_{ext}^* for particle number concentration $N = 1$
 256 cm^{-3} .



257

258 **Fig. 2.** The calculated spectral optical properties for hematite, goethite and illite with different complex refractive
 259 index and size distribution. (a) Volume cross section of extinction, (b) Volume cross section of absorption, (c)
 260 Single scattering albedo and (d) Asymmetry factor

261

262 Figure 2a and Figure 2b demonstrate how the normalized spectral extinction coefficient and
 263 the normalized spectral absorption coefficient vary due to the differences in the refractive indices
 264 and median radius of the minerals. As shown in Figure 2a, β_{ext}^* for hematite has a spectrum which
 265 is clearly distinguishable from that for illite at UV, visible and IR wavelengths. One point should
 266 be noted: hematite has a lower normalized spectral extinction coefficient than illite at wavelengths
 267 less than 1.3 μm , which means that hematite has a weaker optical extinction capacity than illite at
 268 these wavelengths. In the IR region, the spectral features of hematite in β_{ext}^* show large differences
 269 in volatility, and mimic the features in the refractive index of hematite. The magnitude of β_{ext}^*
 270 depends on the parameters of the particle size distribution.

271 Figure 2b shows the equivalent normalized spectral absorption coefficient for hematite from
 272 QE1985 and LG1988 at wavelengths less than 5 μm . The normalized absorption coefficient of
 273 hematite from QE1985 is about 100 times larger than that for illite at both visible and near-IR
 274 wavelengths, but the normalized absorption coefficient for hematite from LG1988 has larger
 275 values than those for illite at wavelengths less than 1 μm and about 100 times smaller values than

276 those for illite at wavelengths between 1 μm and 5 μm . If we adopt the complex refractive indices
277 of QE1985, the calculated absorption coefficient of hematite indicates that hematite is an
278 especially strong absorber at UV and visible wavelengths. Conversely, the calculated absorption
279 coefficient of hematite using LG1988 data suggests that hematite is an important aerosol
280 component only for short-wavelength absorption. Considering the whole region from 0.2 μm to 5
281 μm , hematite with complex refractive indices from LG1988 has an approximately equal absorbing
282 capacity to that of illite.

283 Figure 2c illustrates the single scattering albedo of hematite and illite at wavelengths between
284 0.2 μm and 5 μm for different particle size distributions. The single scattering albedo of hematite
285 is about 0.6 for wavelengths $\lambda < 0.55 \mu\text{m}$ and varies little from $r_0 = 0.7 \mu\text{m}$ to $r_0 = 0.5 \mu\text{m}$. In
286 contrast, illite has ω_0 in the range from about 0.9 to 1 for $\lambda < 0.55 \mu\text{m}$, showing strong spectral
287 dependence at short wavelengths. For $\lambda > 0.55 \mu\text{m}$, illite has ω_0 of about 0.99 for $0.55 < \lambda < 2.0$
288 μm and this gradually reduces to about 0.5 for $2.0 < \lambda < 5.0 \mu\text{m}$, while hematite shows large
289 differences of ω_0 depending on the source of the refractive indices. The single scattering albedo
290 decreases to about 0.35 at UV wavelengths for hematite nanoparticles (which are always observed
291 as aggregates with other clay mineral particles) with $r_0 = 0.01 \mu\text{m}$ and $\sigma = 2.0$, but it rapidly
292 increases to nearly 1 at wavelengths $\lambda > 0.7 \mu\text{m}$.

293 Figure 2d compares the asymmetry parameter of hematite with refractive indices from
294 QE1985 and LG1988 at UV and visible wavelengths with size modes of $r_0 = 0.7, 0.5,$ and $0.01 \mu\text{m}$.
295 For $r_0 = 0.5$ and $0.7 \mu\text{m}$, hematite from QE1985 has $g = 0.3 - 0.99$, g decreasing as λ increases.
296 The magnitudes of g from LG1988 are in the range from 0.2 to 0.99 with a few fluctuations.
297 For $r_0 = 0.01 \mu\text{m}$, both datasets put g in the range from about 0.15 to 0.38. Thus, the magnitude of
298 g depends significantly on the particle size distribution.

299

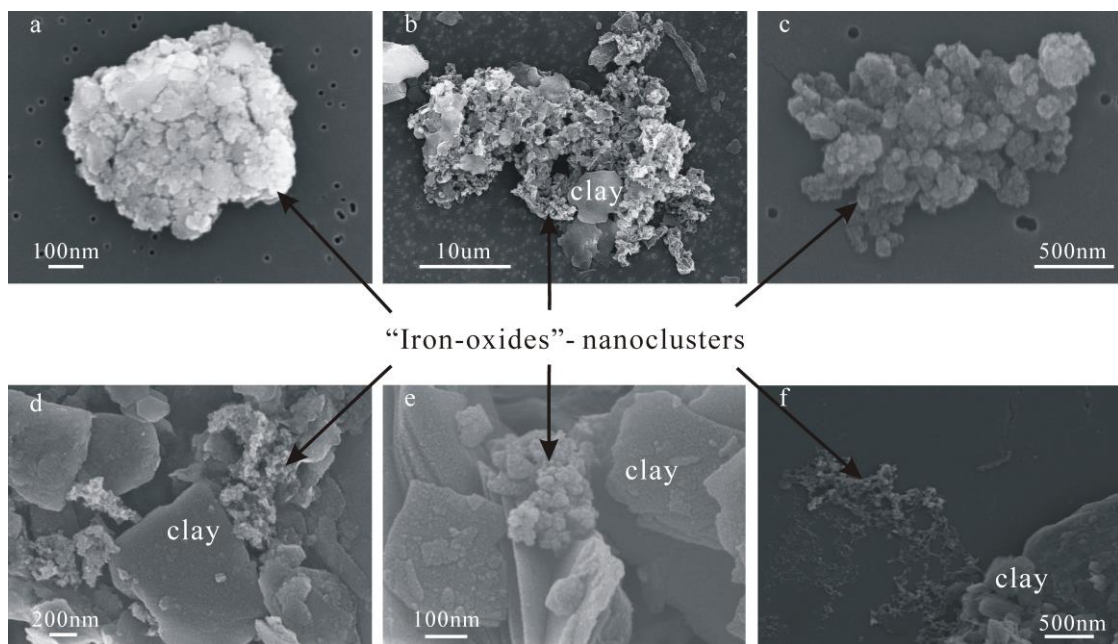
300 **3.2 Physical and mineralogical properties**

301 **3.2.1 Size and Morphology**

302 Many electron microscopy observations (Greeland et al., 1968; Tipping, 1981; Postma and
303 Brockenhuus-Schack, 1987; Poulton and Canfield, 2005; Raiswell and Anderson, 2005; Shi et al.,
304 2009; Deboudt et al., 2012; Wagner et al., 2012; Guo and Barnard, 2013) have shown that poorly

305 ordered iron-oxides commonly occur as spheroidal to ellipsoidal nanoparticles that may be single
306 or aggregated, and may be unattached or attached to quartz or clay minerals (Figure 3). The
307 reasons for the aggregation and the attachment are not well understood but are likely to be related
308 to interactions of surface charge characteristics between iron-oxides and quartz or clay minerals
309 (Poulton and Canfield, 2005). According to Hinds (1982), the binding mechanisms that hold
310 separate aerosols together in an agglomerate formed in the air include the van der Waals force, the
311 electrostatic force and the surface tension of adsorbed liquid films. As mentioned above, the
312 dispersed nanoparticles of iron-oxides which are attracted to larger dust particles have more
313 prominent optical absorption than aggregated iron-oxides, but the use of size distributions for
314 nanoparticles (such as $r_0 = 0.01 \mu\text{m}$ and $\sigma = 2.0$) will overestimate the optical absorption of
315 iron-oxides in natural dust aerosol samples.

316



317

318 **Fig. 3.** Representative morphology of iron-oxide aggregates in dust samples observed by SEM.

319

320 3.2.2 Abundance of total iron and iron-oxides

321 By employing the optical parameters of hematite and clay minerals calculated above, we can
322 model the optical effects of hematite in dust aerosols. Problems associated with this are the actual
323 variation of iron-oxides content and the state of the mixture with other minerals and these should
324 be accounted for when modeling the optical properties of dust aerosols.

325 The total iron content in dust aerosol bulk samples is always measured in terms of the Fe_2O_3

326 mass percentage of the total oxide mass by elemental analysis (i.e. XRF, PIXE or ICP). Ganor and
327 Foner (1996) gave a median Fe content of 2.9% for dust storms in Israel. The observed Fe
328 percentage for the Dunhuang site in China during ACE-Asia is $4.0 \pm 0.9\%$ (Zhang et al., 2003). A
329 value of $4.45 \pm 0.49\%$ (Guieu et al., 2002) has been proposed as characterizing Saharan dust. The
330 elemental analysis by XRF yielded total iron oxide contents between 2.0 and 5.0 weight % for
331 four Saharan mineral dust samples of different color and origin (Linke et al., 2006). Moreover,
332 Lafon et al. (2004) and Lafon et al. (2006) reported that the total iron content (the Fe_2O_3
333 percentage) varies from 6.2% to 8.7% in six atmospheric samples and three wind tunnel generated
334 samples. The total iron content ranges from 1.82% to 11.8% (with an especially high value of 30.0%
335 in the sample collected from Bamako, Mali) in entrained $\text{PM}_{2.5}$ from ten soil samples representing
336 the Arabian Peninsula, the Sahara and Sahel regions and samples from northeast Africa and
337 south-central Asia (Moosmüller et al., 2012). Furthermore, percentage values of iron content from
338 several datasets reported by previous studies vary mainly between 4 and 11% (e.g., Gomes and
339 Gillette, 1993; Chiapello et al., 1997; Gao et al., 2001; Journet et al., 2014).

340 One additional aspect should be discussed here for the proper interpretation of the obtained
341 data: how representative is the total iron concentration of the free iron oxide content of dust
342 aerosols? Free-iron is present as a major aerosol component affecting the short-wavelength
343 absorption of mineral dust. However, iron oxide represents only part of the total iron, which may
344 also exist in the crystal lattice of numerous other dust minerals. The iron oxide-to-total iron ratio
345 in natural and soil-derived aerosols has been characterized by applying an adapted reductive
346 extraction method as commonly used in soil science (Lafon et al., 2004; Lafon et al., 2006). This
347 method provides no structural information about the extractable iron and therefore cannot
348 distinguish between the presence of goethite or hematite in the samples. Fortunately, this method
349 can provide an upper limited to the free-iron content for the optical modeling.

350 Lafon et al. (2004) and Lafon et al. (2006) reported considerable variability in the iron
351 oxide-to-total iron ratio for various regions and sampling conditions and that there is no clear
352 relationship between the oxide-to-total iron ratio and dust origin or aging. Based on all the values
353 for the oxide-to-total iron ratio reported in the published literature (Lafon et al., 2004; Lafon et al.,
354 2006; Alfaro et al., 2004; Formenti et al., 2008; Klaver et al., 2011a), we have calculated an
355 average of 0.52. Formenti et al. (2014a) reported that iron oxides account, by mass, for 0.38 to

356 0.72 of the total elemental iron based on X-ray absorption analysis of samples of mineral dust
357 emitted from or transported to western Africa. Reynolds et al. (2013) reported that the percent iron
358 in goethite and hematite relative to iron in all iron-bearing phases ranges from 0.2 to 0.52 for dust
359 samples in Australia as determined from Mössbauer spectra. Based on an average compiled from
360 the literature, Kandler et al. (2009) and Kandler et al. (2011) have assumed that only 20% of the
361 total iron content is hematite when determining the complex refractive index of dust aerosols.
362 Alfaro et al. (2004) found in their dust samples comparable total iron contents in the range of 3.0
363 to 6.5 weight %, and they assigned a significant amount of 2.8 to 5.8 % of this iron as present in
364 iron oxide mineral phases. This result is consistent with the reported 2.8 - 5.0% of free-iron in
365 aerosol samples collected from three different locations over the world (Lafon et al., 2004; Lafon
366 et al., 2006). Takahashi et al. (2011) indicated that the content of iron-oxides is less than 5 weight %
367 in Asian dust. Moreover, Klaver et al. (2011a) reported that the iron oxides-to-total iron ratio for
368 the analysed samples varied between 0.4 and 0.61, accounting for between 1% and 3% of the total
369 gravimetric mass, and Formenti et al. (2008) also illustrated that iron oxides (speciation hematite
370 and goethite) represented 2.4% and 4.5% of the total mineral dust mass. As mentioned above, only
371 about half of the total iron content is represented by free-iron.

372 XRD analytical technology has also been applied to identify the content of hematite and
373 goethite in some cases and less than 2% iron oxides was detected (Shi et al., 2005; Linke et al.,
374 2006b; Kandler et al., 2009; Lawrence et al., 2010; Klaver et al., 2011a; Wagner et al., 2012;
375 Formenti et al., 2014b). Depending on the crystal phase of interest, this method has a detection
376 limit of 0.1 to 0.5 weight % for iron oxides (Balsam et al., 2014). Discrepancies between the
377 quantified free-iron content detected by the method of Lafon et al. (2006) and the hematite or
378 goethite contents determined by XRD could be due to difficulties of the Rietveld method
379 associated with poor crystallographic ordering of iron oxides in mineral dusts.

380 Single particle analysis has also been conducted for detecting the free iron oxides. Fe-rich
381 particles (iron oxides) represented no more than 5% of the particle number in aerosol samples and
382 hematite or goethite were found more often in the fine fraction (Chou et al., 2008; Kandler et al.,
383 2009; Schladitz et al., 2009; Kang et al., 2009; Scheuvens et al., 2011; Malek et al., 2011; Wagner
384 et al., 2012; Menéndez et al., 2014).

385 **Table 3.** Summary of global reported ratios of hematite to goethite (Hm/Gt) in dust aerosols.

| Location (Number of samples) | Type | Method | Average value of Hm/Gt | Reference |
|---------------------------------|----------|--------|---------------------------|-------------------------------|
| Niger (1) | Aerosol* | DRS | 0.5625 | Lafon et al. (2006) |
| Tunisia (1) | Aerosol* | DRS | 0.4085 | Lafon et al. (2006) |
| China-Zhenbeitai (1) | Aerosol | DRS | 0.3514 | Lafon et al. (2006) |
| Niger (99) | Aerosol | DRS | 0.4286 | Formenti et al. (2008) |
| Niger (12) | Aerosol | XAS | 0.5771 | Formenti et al. (2014a) |
| Gran Canaria (19) | Aerosol | DRS | 0.9048 | Lázaro et al. (2008) |
| North Atlantic (9) | Aerosol | DRS | 0.9276 | Arimoto et al. (2002) |
| Muztagata (7) | Aerosol | DRS | 0.6918 | Xu et al. (2014) ⁺ |
| Golmod (29) | Aerosol | DRS | 0.7262 | Yang et al., (2014) |
| Tazhong (6) | Aerosol | DRS | 0.9157 | Lu et al. (2011) |
| Dunhuang (29) | Aerosol | DRS | 0.8762 | Shen et al. (2006) |
| Yulin (32) | Aerosol | DRS | 0.7158 | Shen et al. (2006) |
| Horqin (22) | Aerosol | DRS | 0.7448 | Shen et al. (2006) |
| Australia (6) | Aerosol | MS | 0.4571 | Reynolds et al. (2014) |

386 * Dust aerosol produced by wind tunnel; ⁺ Unpublished paper of the fourth author, private communication.

387 The technology of diffuse reflectance spectroscopy (DRS) has normally been to quantify the
388 ratio of hematite to goethite in a particular dust sample (Lafon et al., 2006; Shen et al., 2006;
389 Lázaro et al., 2008; Formenti et al., 2008; Formenti et al., 2014a). The accurate quantification of
390 goethite and ferrihydrite in dust is extremely difficult owing to similarities in structure and
391 associated absorption spectra of these two minerals (Scheinost et al., 1998; Torrent and Barrón,
392 2002; Schroth et al., 2009). This has the critical implication that the content of goethite measured
393 by absorption spectroscopy is actually the sum of goethite and ferrihydrite. This does not, however,
394 affect the optical calculations due to their optical similarity. Table 3 summarizes the measured
395 ratios of hematite to goethite in global dust aerosol samples and shows higher ratios of Hm/Gt in
396 Asian dust samples compared to African samples. Over the whole world, it is concluded that
397 goethite predominates over hematite with a relative abundance of 50% - 75% of iron oxides in
398 dust aerosols.

399 Based on the above reported results, we conclude that the iron-oxides account for
400 approximately half of the mass of elemental Fe and for between 2 and 5 % of the dust mass. Most

401 of them are composed of goethite, representing between 50 and 75 % of the iron oxide mass.

402

403 **3.2.3 Mixing states**

404 As free-iron particles are always mixed with other kinds of particle, the condition of the
405 mixture could be important for their ability to scatter and absorb radiation. The 3D structure of
406 iron-oxide particles obtained by tomography reveals that these Fe-rich inclusions are often found
407 at the surface of aluminosilicate particles but that some are also included inside particles (Deboudt
408 et al., 2012). Inversions calculated assuming external mixing are better able to explain the
409 wavelength dependence of dust absorption by varying only hematite concentration than inversions
410 using internal mixing (Koven and Fung, 2006; Formenti et al., 2014a). Thus, a semi-external
411 mixing assumption is clearly an optimal approximation for iron-oxides mixed with aluminosilicate
412 particles. Moreover, this assumption has the advantages of simplicity of calculation, interpretation,
413 and the possibility of comparing with model results.

414

415 **3.3 Further simulation and verification**

416 Sokolik and Toon (1999) also suggested that the radiative properties of a mixture would
417 strongly depend on the relative abundance of individual minerals due to the large variations in the
418 optical properties of individual minerals. Therefore, we model the optical properties for a range of
419 possible weight fractions of hematite in the clay-size mode while the remaining mass is illite
420 which represents the clay minerals. As mentioned above, we adopt 0% hematite as the lower limit
421 for the aerosol samples with no free-iron particles, 2.5% hematite for the transported dust aerosol
422 samples, 5% hematite for the locally emitted dust samples and 7.5% hematite for the upper limit.
423 Due to the limited and discontinuous refractive indices of goethite, this setting may underestimate
424 the actual optical effects of goethite in dust aerosol. Using the density of hematite (5.3 g/cm^3) and
425 illite (2.75 g/cm^3), volumetric hematite fraction was converted from the mass hematite fraction to
426 calculate the effective complex refractive indices for dust.

427 Dust mineralogical composition is often expressed as a weighted fraction of individual
428 components in the total dust sample. Because the relationship of number concentration and mass
429 concentration can be expressed as:

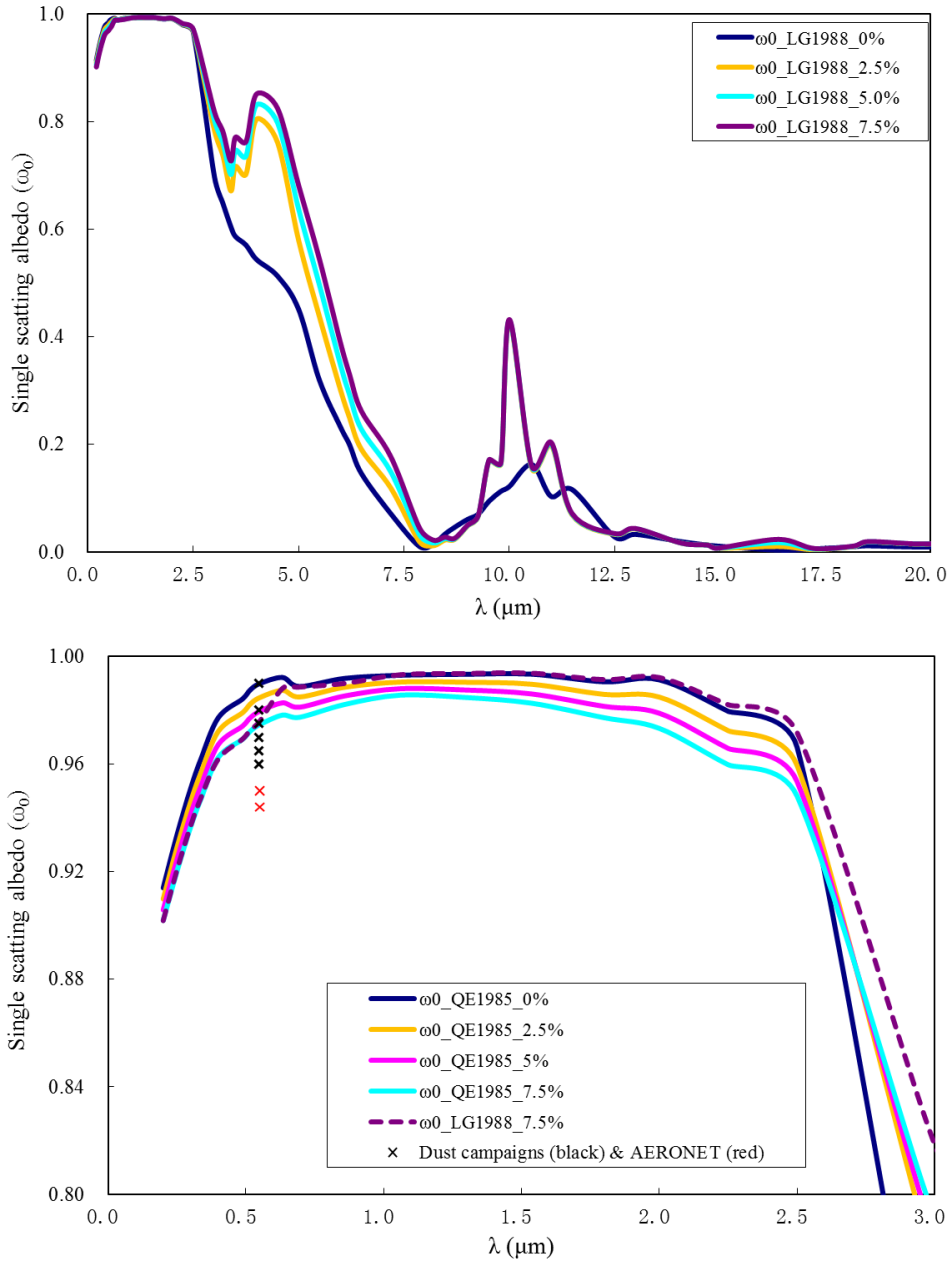
430
$$M_i = M_0 \cdot W_i = N_i \cdot \rho_i \cdot \frac{4}{3} \pi (r_0)^3 \cdot \exp\left[\frac{9}{2} (\ln \sigma)^2\right],$$

431 where M_i , N_i , W_i and ρ_i are the particle mass concentration, number concentration, weight
 432 fraction and density of the i -th mineral in the mixture respectively and M_0 is the total particle
 433 mass concentration of the mixture sample, the optical properties of external mixtures of minerals
 434 can be modeled by:

435
$$K_{ext}^{mix} = \sum (K_{ext(i)}^* \cdot N_i) = \sum (K_{ext(i)}^* \cdot \frac{M_0 \cdot W_i}{\rho_i \cdot \frac{4}{3} \pi (r_0)^3 \cdot \exp\left[\frac{9}{2} (\ln \sigma)^2\right]})$$

436 Figure 4a shows the differences between the single scattering albedo calculated for the
 437 mixture of illite and hematite with complex refractive indices from LG1988 at visible and infrared
 438 wavelengths. Compared to ω_0 for illite with no hematite, it is not hard to see that ω_0 for mixtures
 439 with different amount of hematite show significant differences in four wavelength ranges, namely,
 440 0.2 - 0.7 μm , 2.5 - 8.7 μm , 8.7 - 12.5 μm and 15.0 - 17.5 μm . For $0.2 < \lambda < 0.7 \mu\text{m}$ and $15.0 < \lambda <$
 441 $17.5 \mu\text{m}$, ω_0 for the mixtures is smaller than ω_0 for pure illite, which means the presence of
 442 hematite enhances the optical absorption of the mixtures. For $2.5 < \lambda < 8.7 \mu\text{m}$, ω_0 for the
 443 mixtures is larger than for pure illite, which means the presence of hematite enhances the optical
 444 scattering of the mixtures. But for $8.7 < \lambda < 12.5 \mu\text{m}$, ω_0 shows more complicated fluctuations.

445
 446



447

448 **Fig. 4.** Spectral single scattering albedo (SSA) for mixtures of illite and hematite with varying percentages and
 449 refractive indices at wavelengths of 0-20 μm (a) and 0-3 μm (b).

450

451 Figure 4b shows the differences between ω_0 for illite mixed with different amounts of

452 hematite with refractive indices from QE1985 at solar wavelengths. The magnitude of ω_0 for a

453 mixture increases when the median radius r_0 increases. In order to compare the effects for

454 hematite with refractive indices from two different sources for the same mixture state, we also plot

455 ω_0 for illite mixed with 7.5% hematite with refractive indices from LG1988 in Figure 4b.

456 Evidently, for $0.55 < \lambda < 2.5 \mu\text{m}$, the dataset of QE1985 will lead to higher optical absorption,

457 although the two datasets have the same optical scattering and absorption for $\lambda < 0.55 \mu\text{m}$.

458 Another question is what is the competition between theoretical calculated values and field
459 observed results? In order to compare our calculated SSAs with measured values, we review all
460 reported SSAs during different dust campaigns or inferred from AERONET measurements, and
461 listed them in Table 4. The measured results for dust mixed with BC were excluded during our
462 review progress, such as the results from the campaign of AMMA. The measurements from
463 DABEX are comparable but on the lower edge of previous measurements performed at 550 nm
464 during the TARFOX, SHADE, GERBILS, SAMUM, NAMMA and Fennec 2011 field campaigns.
465 Absorption from the mineral dust as measured using the corrected nephelometer and Particle Soot
466 Absorption Photometer (PSAP) combination suggests that single scattering albedos at 550 nm
467 (SSA550) range from 0.91 to 0.97 (with a mean of 0.97) for iron oxide mass fractions between 1.3
468 and 3.5% (Klaver et al., 2011a). The SSAs at 532 nm were reported as 0.99 ± 0.001 and $0.98 \pm$
469 0.002 for the Cairo 2 and Morocco dust samples with Fe_2O_3 mass fractions of 4.5 and 3.63%,
470 respectively (Linke et al., 2006). Johnson and Osborne (2011) revealed that the use of the mineral
471 dust refractive indices from Balkanski et al. (2007) assuming 1.5% hematite gave reasonable
472 agreement with the measured single scattering albedo, consistent with the findings of Klaver et al.
473 (2011a). Haywood et al. (2011) shown that mineral dust is relatively non-absorbing at 550 nm due
474 to the relatively small fraction of iron oxides present (1–3%). Balkanski et al. (2007) addressed
475 this difference and argued that dust absorption at visible wavelengths might be lower than
476 previously thought because mixing rule calculations with hematite content of 1.5% by volume,
477 supposedly representative of median dust absorption, showed a very good agreement with the
478 AERONET measurements. The same question is presented in Figure 7a of Formenti et al. (2014a):
479 that the calculated SSA using a higher imaginary index of iron-oxides also overestimated the
480 absorbing ability of Saharan dust in comparison with field observation.

481 We compare our calculated SSA curves with the measured values in Figure 4b. It is shown
482 that the observed SSAs were mostly ranged in 0.94-0.99 during different dust campaigns, but
483 much lower (0.944-0.95) for the AERONET which cannot exclude the presence of black carbon
484 with higher absorbing. Our calculated result could consistent with the higher part (0.97-0.99) of
485 measured SSAs, but higher than the lower part (0.95-0.97) due to the effect of coarse particles
486 during different dust campaigns. Thus, the iron oxide content alone cannot explain the variability

487 of the single scattering albedo. This suggests that more complete knowledge of the dust
 488 mineralogical composition and size distribution with varied multi-modes is needed as input to
 489 more rigorous modeling.

490 **Table 4.** Review of measured dust single scattering albedos during dust campaigns and AERONET observations.

| References | SSA_550nm (Mean) | S.D. | Observations |
|----------------------------|------------------|-------|----------------|
| Haywood et al. (2001) | 0.97 | 0.02 | TARFOX |
| Haywood et al. (2003) | 0.97 | 0.02 | SHADE |
| Johnson and Osborne (2011) | 0.97 | 0.02 | GERBILS |
| Müller et al. (2011) | 0.96 | 0.03 | SAMUM_2 |
| Petzold et al. (2011) | 0.975 | 0.15 | SAMUM |
| Jeong et al. (2008) | 0.96 | 0.01 | NAMMA |
| Osborne et al. (2008) | 0.99 | 0.02 | DABEX |
| Ryder et al. (2013) | 0.965 | 0.015 | Fennec 2011 |
| Lack et al. (2009) | 0.95 | 0.01 | TexAQS/GoMACCS |
| Linke et al. (2006) | 0.985 | 0.006 | Laboratory |
| Zhu et al. (2007) | 0.95 | 0.01 | AERONET |
| Kim et al. (2011) | 0.944 | 0.005 | AERONET |

491

492 In order to check the further effects of the mixing state of hematite on the single scattering
 493 albedo, we use two sets of assumptions that bracket the actual state of hematite mixing: internal
 494 mixing (Int), in which individual dust particles are a combination of all components present; and
 495 external mixing (Ext), in which different components exist as separate particles.

496 For the case of an external mixture of particles, the average optical properties are calculated
 497 by summing over the optical properties of the individual species. Approximations have to be made
 498 to calculate the optical properties of internal mixed particles. Three common approximations for
 499 the calculation of these latter optical properties are the volume mixing method, the Bruggeman
 500 approximation and the Maxwell-Garnett approximation (Chýřlek et al., 1988; Bohren and
 501 Huffman, 1998). Detail information about the three methods is given by Sokolik and Toon (1999).
 502 We have calculated the single scattering albedo (SSA) of illite-hematite mixtures with different
 503 hematite contents using internal mixing according to the above three internal approximations and
 504 also using external mixing.

505 The calculated SSA values for illite-hematite mixtures using internal and external mixture
 506 assumptions as a function of wavelength and hematite mass fraction are illustrated in Figure 5. For
 507 the case of external mixing, the SSAs at 405 nm show good agreement for refractive indices from

508 QE1985 and LG1988, but the calculated SSAs at 870 nm for hematite with refractive indices from
509 QE1985 are much smaller than those using LG1988. This is explained by Figure 4b where the two
510 datasets have the same optical scattering and absorbing properties for $\lambda < 0.55 \mu\text{m}$ but the dataset
511 of QE1985 leads to higher optical absorption for $\lambda > 0.55 \mu\text{m}$. The calculated SSAs with the three
512 different internal mixing methods are all much smaller than those for external mixing both at 405
513 nm and 870 nm since the assumption of an external mixture results in less absorption and less
514 wavelength dependence of absorption than does the assumption of an internal mixture for small
515 amounts of hematite. The basic reason for this is due to the extremely high imaginary refractive
516 index for hematite at short wavelengths. For the case of internal mixing, the SSAs from the
517 volume mixing method are smaller than for the other methods. This is due to the averaged
518 imaginary refractive index being larger than for the other two approximations. On the basis of the
519 study of Peterson (1968), only the effective refractive index of the non-metallic part of the dust
520 can be calculated using the volume mixing method. Thus, adopting the volume mixing method to
521 calculate the optical properties of aerosol samples will lead to a smaller SSA (Levoni et al., 1997;
522 Sokolik and Toon, 1999; Shi et al., 2005; Höller et al., 2003; Ebert et al., 2004; Kandler et al.,
523 2007; Kandler et al., 2009; Petzold et al., 2009; Otto et al., 2009; Wagner et al., 2012).

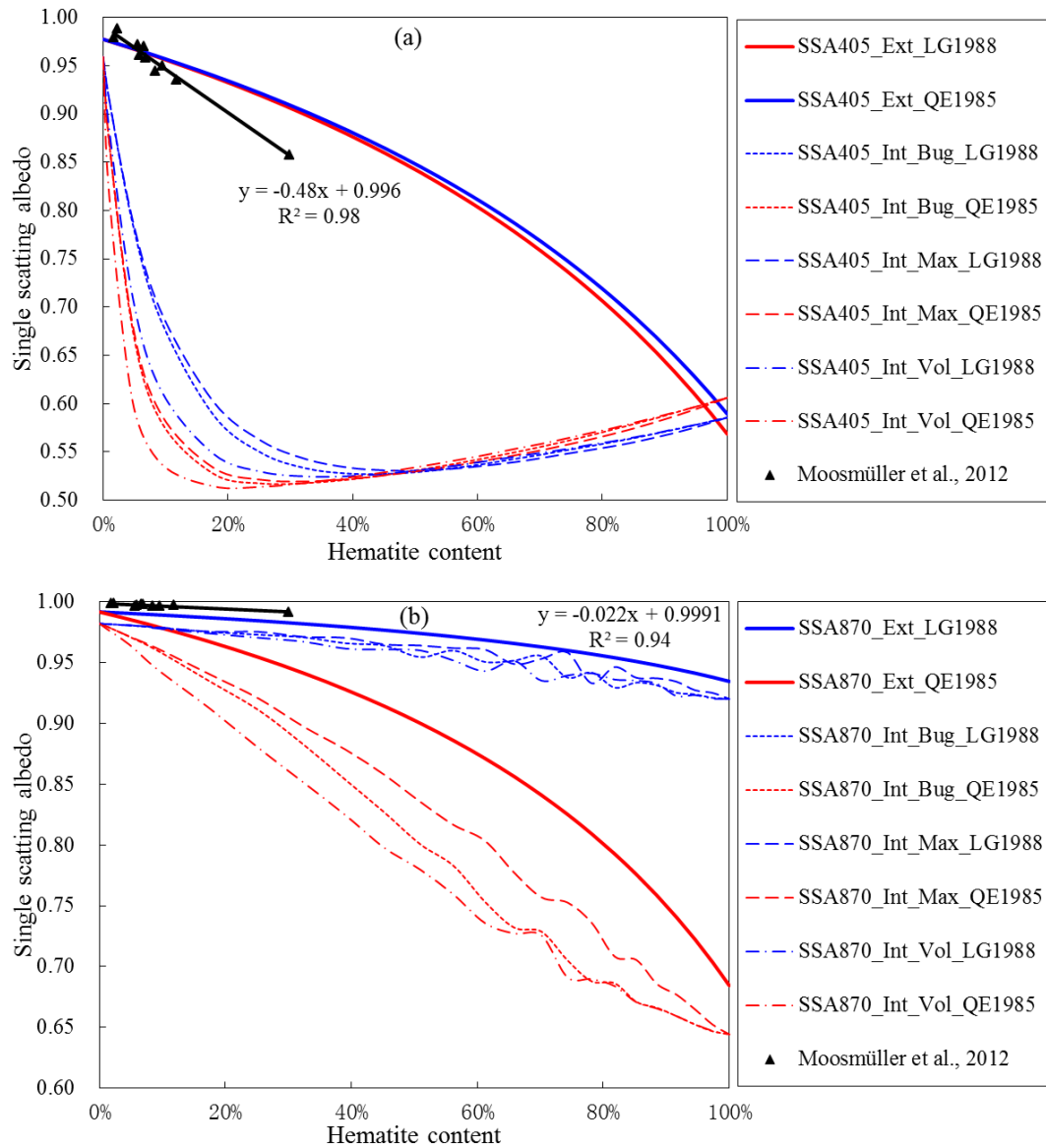
524 The calculated SSAs using the Bruggeman approximation are consistent with those from the
525 Maxwell-Garnet approximation for low hematite contents at both 405 nm and 870 nm but differ
526 from them for hematite content larger than 10%. Both the Maxwell-Garnet and Bruggeman
527 approximations are derived from the same integral equation for the propagation of electromagnetic
528 waves in an inhomogeneous medium but under a different set of approximations (Chyřek et al.,
529 1988; Bohren and Huffman, 1998). In previous studies, the Bruggeman approximation (Sokolik
530 and Toon, 1999; Lafon et al., 2006; Koven and Fung, 2006; Mishra and Tripathi, 2008; Thomas
531 and Gautier, 2009; McConnell et al., 2010; Klaver et al., 2011a; Wagner et al., 2012; Mishra et al.,
532 2012) has been more often used for calculating the complex refractive index of silicate-hematite
533 mixtures than the Maxwell-Garnet approximations (Balkanski et al., 2007; Hansell Jr et al., 2011).

534 The Bruggeman approximation allows for the calculation of an effective dielectric constant
535 of multicomponent mixtures without distinguishing between matrix and inclusions. If we do not
536 know which is the main body for the silicate and the hematite in an aerosol, it is better to choose
537 the Bruggeman approximation. For the Maxwell-Garnet approximation a decision must be made

538 as to which component is the matrix and which is an inclusion. The Maxwell-Garnett
539 approximation is designed for small inclusions inside a host matrix and thus it is not suitable for
540 hematite >50% in the mixture, so the calculated SSAs show abnormal fluctuation for hematite >50%
541 in Figure 5. This phenomenon disappears if we consider the inverse Maxwell-Garnett
542 approximation which makes hematite the host matrix. Actually, the variation of Fe₂O₃ has been
543 constrained within the range 0–10%, so we advise the use of the Maxwell-Garnett approximation
544 in which the inclusions should be identical in composition but may be different in volume, shape
545 and orientation.

546 The comparison between the laboratories measured SSAs with known abundance of
547 iron-oxides with our theoretically calculated SSAs by different mixing rules, would give us further
548 insights into the actual mixing states of iron oxides and accuracy of different refractive indices.
549 Laboratory measured SSAs by extinction and photoacoustic absorption measurements at different
550 wavelengths have been specific reported (Linke et al., 2006; Moosmüller et al., 2012).
551 Moosmüller et al (2012) has demonstrated that SSAs are much smaller at 405 nm than at 870 nm
552 and all SSAs are linearly correlated with the iron content. These measured results are shown in
553 Figure 5 for comparison with our theoretically calculated SSAs. The measured SSAs at 405 nm
554 show good agreement with our external mixing SSA values calculated from both LG1988 and
555 QE1985 for the abundance of hematite less than 10%, while they are much larger than our internal
556 mixing SSA values at 405nm (Figure 5a), potentially indicating that the dust samples are mainly
557 external mixing and are accompanied by a very small degree of internal mixing.

558 In contrast, the measured SSA values at 870 nm are much larger than our external and
559 internal mixing SSA values calculated from refractive indices from QE1985 but show good
560 agreement with our external mixing SSA values calculated using LG1988 values (Figure 5b). This
561 illustrates the fact that the complex refractive indices of hematite from QE1985 have greatly
562 overestimated absorption at 870 nm.



563

564 **Fig. 5.** The single scattering albedo (SSA) as a function of varying hematite content at wavelengths of (a) 405 and
 565 (b) 870 nm, with different mixing states (Internal and External mixing), different complex refractive index sources
 566 (QE1985 and LG1988) and mixing approximations (Volume, Maxwell-Garnet and Bruggeman). For comparison,
 567 the measured values using a photoacoustic instrument with integrating reciprocal nephelometer and linear fits from
 568 Moosmüller et al.(2012) are also shown.

569

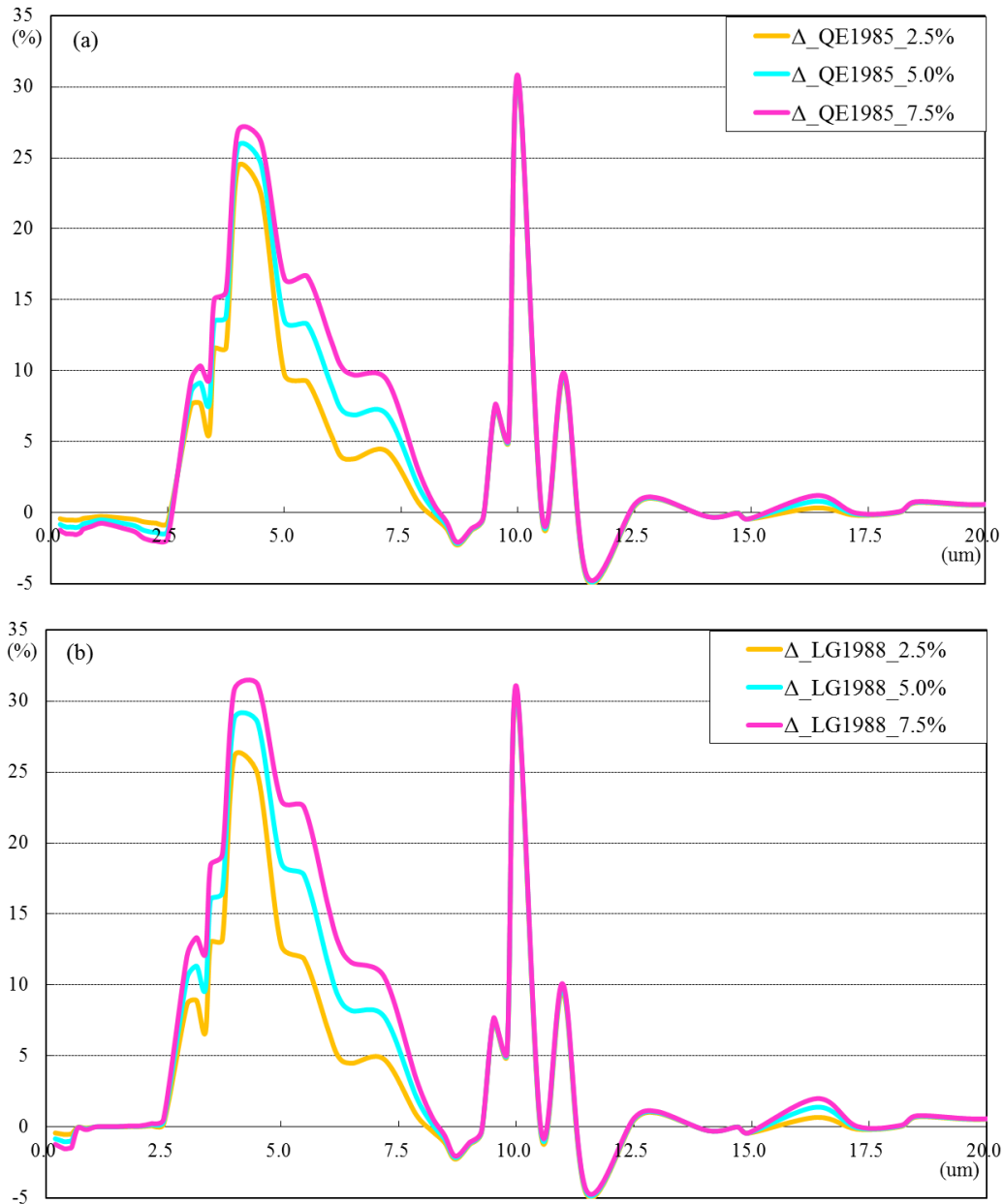
570 For purposes of further quantitatively illustrating the optical effects of hematite to the
 571 mixtures, we calculate $\Delta\omega_0$ as the difference between the single scattering albedo of mixtures
 572 with given abundances of hematite and pure illite. Thus, we have

573
$$\Delta\omega_0 = \frac{\omega_0^{mixture} - \omega_0^{illite}}{\omega_0^{illite}} \times 100\% .$$

574 Here, we assume $M_0 = 100 \mu\text{g}/\text{cm}^3$ and a lognormal size distribution with $r_0 = 0.5 \mu\text{m}$, $\sigma =$

575 2.0. The single scattering albedo of pure illite represents the case when the dust contains no
576 hematite. If the calculated values of $\Delta\omega_0$ are negative, this means that hematite has high
577 absorption at the corresponding wavelengths. Figure 6 shows the calculated $\Delta\omega_0$ for hematite
578 with refractive indices from QE1985 and LG1988 at $\lambda < 20 \mu\text{m}$. It demonstrates that hematite
579 with refractive indices from LG1988 only enhances the optical absorption of dust mixtures for $\lambda <$
580 $0.55 \mu\text{m}$, but hematite with refractive indices from QE1985 enhances the absorption for $\lambda < 2.5$
581 μm . The magnitude of $\Delta\omega_0$ for 5% hematite over these wavelengths is approximately about 1%.
582 Therefore, the use of refractive indices for hematite from QE1985 in climate models would lead to
583 overestimation of the optical absorption at both visible and near-IR wavelengths. Another apparent
584 difference is the positive magnitude of $\Delta\omega_0$ for hematite with refractive indices from the two
585 datasets at wavelengths of $2.5 < \lambda < 8.7 \mu\text{m}$.

586 As about 87% of solar spectral irradiance is distributed at wavelengths from $0.4 \mu\text{m}$ to 2.5
587 μm (ASTM G-173, <http://rredc.nrel.gov/solar/spectra/am1.5/>), hematite with refractive indices
588 from LG1988 shows approximately equal absorption to the clay mineral illite over the whole
589 region from $0.2 \mu\text{m}$ to $2.5 \mu\text{m}$, and only enhances the optical absorption of aerosol mixtures at $\lambda <$
590 $0.55 \mu\text{m}$. Thus, a more precise description of the nature of clays and iron oxides is necessary at
591 lower wavelengths because of large differences in their spectral optical properties.



592

593 **Fig. 6.** The difference $\Delta\omega_0$ between the single scattering albedo (SSA) of pure illite and the single scattering albedo
 594 of mixtures with different contents and refractive indices of hematite. (a) Complex refractive index of hematite
 595 from Qurrey (1985), (b) Complex refractive index of hematite from Longtin (1988).

596

597 Given all that, the complex refractive index of iron-oxides is therefore a key parameter in
 598 effects of dust aerosols on the radiation balance, and the optical constants of hematite from
 599 different sources become a major source of uncertainty in radiative forcing calculation. Further
 600 work is needed to provide experimental measurements of the refractive index of iron-oxides,
 601 especially for hematite and goethite in the visible region of the spectrum. More complete
 602 knowledge of the dust mineralogy, morphology and size distribution with varied multi-modes is

603 needed as input to more rigorous modeling.

604

605 **4. Summary**

606 In this paper we have investigated the spectral optical properties of iron-oxides with
607 considering different refractive indices, size distributions, and more logical weight fractions and
608 mixing states of iron-oxides. The iron-oxides account for approximately half of the mass of
609 elemental Fe and for between 2 and 5 % of the dust mass. Most of them are composed of goethite,
610 representing between 50 and 75 % of the iron oxide mass. The iron-oxides commonly occur as
611 spheroidal to ellipsoidal nanoparticles that may be single or aggregated, and may be unattached or
612 attached to quartz or clay minerals, which could be expressed as semi-external mixing state.
613 Moreover, the spectral SSA values determined in the present study show a strong
614 wavelength-dependence with a steep decrease from the visible to the near-UV. There are still
615 problems that need to be solved in order to accurately study the real role of iron-oxides in
616 determining the overall impact of dust aerosols on climate perturbation, as follows:

617 1. Although there have been many published investigations of the complex refractive index of
618 different iron-oxides, uniformly continuous optical constants for a single type of iron-oxides from
619 0.2 μm to 50 μm are very scarce. Some of them are inconsistent and careful checking of their
620 accuracy is therefore essential.

621 2. The abundance of specific iron-oxide types (such as goethite and magnetite) remains
622 unknown. Although many studies have measured the mass ratio of goethite to hematite as about
623 7:3, the absence of goethite optical constants at 0.75-8.5 μm restricts the usefulness of this ratio.
624 Thus, using hematite to represent all types of iron-oxides is a popular hypothesis.

625 3. Microscopic observations and optical simulations have shown that semi-external mixtures
626 employing both external mixtures of Fe-aggregates and other minerals and partly internal mixing
627 between iron-oxides and aluminosilicate particles is the optimal mixing approximation.

628 4. For hematite, there are two datasets of complex refractive indices that differ significantly.
629 Compared with LG1988, the complex refractive indices of QE1985 greatly overestimate the
630 optical absorption at both visible and near-IR wavelengths. Comprehensive laboratory
631 measurements of the refractive indices of iron-oxides, especially of hematite and goethite in the
632 visible spectrum, should therefore be made in order to accurately assess the effect of mineral dust

633 on climate perturbation.

634 Theoretically calculated SSA values are comparable to values observed in recent laboratory
635 and field studies in the range of 0.97-0.99. The iron oxide content alone cannot explain the
636 variability of the single scattering albedo, and the lower SSAs could be explained as the presence
637 of coarse dust particles and high-absorbing black carbon in natural transported dust aerosol. More
638 complete knowledge of the dust mineralogical composition and size distribution with dynamic
639 varied multi-modes is needed as input to more rigorous modeling.

640

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646

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