

We say thanks to both reviewers for their detailed and constructive comments, which most of them have been used to improve the paper.

One basic idea of Referee #1 was to make a clear distinction between “desert dust” and “mineral particles”. So we decided even to modify the title of our manuscript. The new title is: “Technical Note: Optical properties of desert aerosol with non-spherical mineral particles: data incorporated to OPAC”

In the following we discuss each point of the remarks in detail, with the reviewer comment partly repeated (in italics).

Referee#1

1.17 I ... would use rather Mineral dust or Desert dust, not both...

We mentioned “mineral desert dust particles” since also particles besides the mineral components may be part of desert aerosol. However, in the new OPAC only the shape assumptions for the mineral particles have been changed. But we improved the manuscript with respect to this aspect, and even the title as mentioned at the beginning.

1.20 It is quite unusual to use references in the abstract

We agree. But in our case the paper is directly an improvement of the old OPAC paper, which therefor must be mentioned already in the abstract with detailed information.

1.23 It is an (of course quite reasonable) assumption that the T-matrix approach improves the phase function. Nevertheless...I would rather suggest... to be a bit more conservative here...

We changed the wording.

1.25 ff It would be good to provide the corresponding changes in asymmetry parameter here also....

We added the information on the asymmetry parameter, since its small change is of special interest in comparison the large phase function deviations.

1.40 For full reliable optical properties you also need an absorption theory

Mie theory takes absorption into account, as well as T-matrix theory does. But we skipped the words scattering theory.

11.42ff ... another reason is that particles have no preferential direction...

Even under the assumption that the particles have no preferential direction the particle shape will have influence on the scattering function. Thus we agree with the referee that it is a good idea to advance beyond Mie theory for dust optical properties.

1.51 Is the largest fraction meant with biggest part?

Yes, we agree and changed the diction.

11 54 I do not agree with this statement.....

We eliminated the statement.

11 67ff That is too much simplification.... Passive remote sensing also includes infrared methods, which not so much rely on the scattering phase function

We changed the text

11 70ff You should clarify somewhere above that your major concern is on solar wavelengths and define your spectral region of interest.....

OPAC covers the broad spectral region from 0.25 to 40 μm , and thus the improved assumptions on the shape of mineral particles are taken into account also in the thermal infrared. That the effect of non-spherical particles is largest in the solar spectral range is one point of the results, mentioned in the text and shown in the figures, but nevertheless it is also considered up to 40 μm .

1 88show that T-Matrix really improves....

The improvement of the scattering properties of non-spherical particles using T-matrix instead of Mie-theory in the solar spectral range has been shown by references mentioned in the paper.

1 92 ...physical reasoning for that?

The variation of the aspect ratio distributions with particle size is fact, found be electron microscope measurements. Physical reason may be the particle formation.

I 106 ...campaigns showed abundance of much larger particles....effective radius around 5 μm or larger is not covered by OPAC...

OPAC takes the variability of mineral dust size distributions into account by providing the possibility to mix 3 different mineral components as required (e.g. using Eq. 2 – 4). When comparing with the SAMUM measurements we refer to the pre-defined mixture “OPAC desert”. The flexibility of OPAC with respect to large particles is limited by the coarse mode mineral component (MICM). This component has an effective radius of 8.1 μm and a maximum particle size of 60 μm (see Tab. 1), which means that OPAC covers mineral dust radii up to these values.

I 187 ...“mineral component” sounds like you are ...taking into account variable dust composition..

The three mineral components used in OPAC have the same refractive indices, as mentioned in the manuscript. The possibility of their individual mixture makes it possible to take into account variable dust compositions with respect to the size distribution.

I 197 ...physical reasoning for assuming prolate particles, i.e. from microscopic imagery?...

With electron microscopy generally the projection of the particle is analyzed. Thus it is not possible to detect whether the individual particle is oblate or prolate. As a consequence, the selection of the form with the better fit is a reasonable decision.

I 274 be more specific on spectral regions here...

and

I 275 ... dust also has significant longwave radiative forcing...

The referee is right that both the SW and the LW spectral range are essential for radiation budget and for remote sensing of desert dust. But, as mentioned, our improvement for the scattering properties of mineral particles holds for both spectral regions. Nevertheless we have corrected this paragraph.

I 306 It is not true that solar wavelength generally is use for aerosol remote sensing...

We have corrected the sentence.

I 313 ...very large particles also in transported dust... results of SALTRACE campaign ..

We agree with the referee that the size dependent loss of particles during transport is an assumption that no longer may be valid for desert dust. Since our institute took part and thus we are informed about the results of the SALTRACE campaign, we will consider this aspect carefully in future aerosol modelling. However, in the actual manuscript the assumption is used only to test the effect of non-sphericity for mixtures of particles with different size distribution. (As mentioned, OPAC has the advantage that any user can decide for an individual composition of the given components and thus for the size distribution of the mixture.) However, we added an explanation.

I 376 ... comment a bit that only one specific set of refractive index is used...

The referee is right that the refractive index is important for the optical properties of particles. But this manuscript, as a technical note, strictly focusses on the particle shape of mineral aerosol and thus leaves the other properties of OPAC unchanged. However we add a sentence belonging to the question of the uncertainty of the refractive indices in the conclusion.

I 429 ... spectral resolution in the different wavelength ranges...

The data in OPAC are available for the wavelengths that can be chosen from the data base. The spectral resolution is not given specifically since it is assumed that the spectral properties of aerosol particles vary rather weak, in contrast to gaseous absorption. Nevertheless, for some specific applications the spectral resolution may be too low. Here improvements will be discussed in future versions of OPAC.

Referee #2

3997 10 ... more representative publications...

Thank you for the suggested references.

4000 1 ...additional modern accounts of the T-matrix method...

We wanted to give credit to Waterman and his original paper. The used updated accounts of the TMM are mentioned in the text. Nevertheless we added the Mishchenko reference, but not Doicu, because we did not use his advanced method.

4000 4 und 13 ..distinction between the aspect ratio and the axial ratio ..

We agree that the definition of the aspect ratio in our text was incorrect and emended it in the revised manuscript.

General commentthe effects of non-sphericity are well known to depend on the imaginary part of the refractive index..... include a table showing the refractive indices used.... different types of dust with different origin...

The dependence of the effects of the particle shape on the refractive index, and especially on its imaginary part, is known and partly considered in the answer to referee #1 and mentioned in the improved conclusion of the manuscript. We prefer not to add a table or figure with the refractive index of the mineral components because it is referenced and the data can be looked up in the ASCII files of the OPAC package. The refractive index used in OPAC for the mineral particles is an average value, for an “average” mineralogical composition. We agree that mineral aerosol originating from different deserts may have different chemical composition and thus different refractive index. But considering a large variation of individual aerosol components is far beyond the idea of OPAC, which will be easy, to be used without too detailed information on the actual aerosol properties. Nevertheless, for a future version of OPA C, we plan to add an additional component with stronger absorbing mineral particles.

1 Technical note:
2 Optical properties of desert ~~dust~~aerosol with non-spherical mineral particles: Data incorporated to
3 OPAC

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16 ▲
17 Abstract

18
19 Mineral ~~desert dust~~ particles in general are no spheres and assuming spherical particles, instead of
20 more realistic shapes, has significant effects on modeled optical ~~dust~~ properties and so on the be-
21 longing remote sensing procedures for desert ~~dust~~aerosol and the derived radiative forcing. Thus in a
22 new version of the data base OPAC (Optical Properties of Aerosols and Clouds; Hess et al., 1998), the
23 optical properties of the mineral particles are modeled describing the particles as spheroids with size
24 dependent aspect ratio distributions, but with the size distributions and the spectral refractive indices
25 not changed against the previous version of OPAC. The spheroid assumption strongly improves
26 known to substantially improve the scattering functions, but pays regard to the limited knowledge on
27 particle shapes in an actual case. The relative deviations of the phase functionsoptical properties of
28 non-spherical mineral particles from those of spherical particles are for the phase function in the
29 solar spectral range up to +60% at scattering angles of about 130° and up to -60% in the backscatter
30 region, but the less than 2% for the asymmetry parameter. The deviations are generally small in the
31 thermal infrared and for optical properties that are independent of the scattering angle. The im-
32 proved version of OPAC (4.0) is freely available under www.rasein.netwww.rascin.net.

33
34
35 1. Introduction

36
37 The optical properties of aerosol particles are the basis for modeling their direct radiative forcing
38 (Lacis a. Mishchenko, 1995; Haywood a. Boucher, 2000; Yi et al., 2011) and thus correspondingly for
39 their effect on climate (McCormick a. Ludwig, 1967; Myhre et al., 2013). Moreover, the optical prop-
40 erties are necessary for all inversion techniques used for aerosol remote sensing (Koepke a. Quenzel,
41 1979; Kaufmann, 1993; Kalashnikova a. Sokolik, 2002; Nousiainen, 2009). Thus, for an easy availability
42 of spectral optical properties of aerosol particles, the software package OPAC, Optical Properties of
43 Aerosols and Clouds, had been created (Hess et al., 1998).
44 The optical properties of aerosol particles in general are modeled with a scattering theory using the
45 size distribution and the spectral refractive indices of the particles. In the past, commonly the as-
46 sumption has been made that the particles are spheres, using Mie-theory (Mie, 1908). This has
47 three different reasons: on the one hand, the assumption of spherical particles is reasonable in many
48 cases, especially for water soluble aerosol types under typical meteorological conditions with relative
49 humidity higher than 50%. On the other hand, the shape of individual particles is known only for a
50 limited number of examples, because it needs electron microscopy measurements. Thus, for actual
51 conditions, for practical use, the shape of particles, particularly as function of size, is not available.
52 But even if the particle shape would be available, the problem remains that modeling of non-

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53 spherical particles is complex and time consuming (Mishchenko a. Travis, 1998). et al., 2000; Kahnert,
54 2003). Thus the use of Mie-theory often is a good or the only possible assumption and it has also
55 been used in OPAC.

56 Desert ~~dust~~aerosol, besides sea salt, forms the ~~biggest part~~largest fraction of the atmospheric parti-
57 cles (D 'Almeida et al., 1991; Kinne et al., 2006). Thus desert ~~dust~~aerosol is very important for the
58 radiation budget and consequently for the climate, especially because it is distributed, often with
59 high optical depth, over large areas. Since its amount shows very strong spatial and temporal varia-
60 tions (Sokolik et al., 2001) ~~it generally is investigated with~~, remote sensing methods, ~~which~~ are im-
61 portant for desert aerosol research. However, remote sensing is always based on the assumed parti-
62 cle characteristics.

63 Especially for ~~desert dust~~ mineral particles the optical properties modeled under the assumption of
64 spherical ~~particles~~shape are ~~insufficient~~questionable, since ~~the mineral~~these particles are generated
65 by mechanical processes which give rise to highly irregular particle shapes, as to be seen by electron
66 micrographs (Falkovich et al, 2001; Kandler et al., 2011).

67 In comparison to spherical particles the phase function of irregular particles generally shows in-
68 creased sideward, but reduced backward scattering, ~~if the particles are relatively large in comparison~~
69 ~~to the wavelength~~ (Zerull et al., 1980; Koepke a. Hess, 1988; Nousiainen, 2009; and see Fig.1). Thus, if
70 ~~measured~~radiation data ~~measured at short wavelengths~~ are used to derive aerosol properties, the
71 assumption of spheres may lead to wrong results. ~~Errors in This holds also for~~ particle properties de-
72 rived from backscatter-lidar measurements (Gobbi et al., 2002; Wiegner et al., 2009; Sakai et al.,
73 2014) ~~may result from~~, since, amongst others, they are influenced by the lidar ratio ~~that has to be~~
74 ~~taken into account~~, which combines backward scattering with the extinction coefficient. For passive
75 remote sensing from satellite, ~~the retrieval error is dominated by the an~~ assumed wrong phase func-
76 tion of the particles (Mishchenko et al., 1997) ~~can introduce significant retrieval errors~~ and ~~in for con-~~
77 siderations of the radiation budget ~~consideration of mineral particles in the solar spectral range~~ the
78 assumption of spheres is a major source of error (Nousiainen, 2009). The amount of solar radiation
79 scattered back to a radiometer at a satellite depends on the scattering angle, i.e. the angles of Sun
80 and satellite, on the aerosol optical thickness, and on the reflectance at the ground. Thus the error in
81 the case of assuming spherical particles is highly variable, and it is essential to use the appropriate
82 scattering function (Horvath et al., 2006). The particle shape effect can cause up to 30% difference in
83 dust forcing at the top of the atmosphere (Yi et al., 2011).

84
85 These aspects are the reason to account for the non-sphericity of mineral particles in OPAC (Hess et
86 al., 1998) and so to improve this algorithm. The user-friendly data base and software package "Opti-
87 cal Properties of Aerosol and Clouds" presents the single-scattering properties of 10 aerosol compo-
88 nents that are given with size distribution and spectral refractive indices for a spectral range from
89 ultraviolet to far-infrared. These components easily can be combined by the user to individual mix-
90 tures, i.e. to variable aerosol types, for which phase functions and other optical and microphysical
91 parameters are modeled after user request.

92 If a particle no longer is assumed to be spherical, the possible variability of the particle shape is in-
93 creased dramatically, from spheres, over spheroids and cubes to really irregular particles (Cheng,
94 1980). Thus, if the shape of particles will be taken into account for general modeling of the optical
95 parameters, it is necessary to decide for simplifications. Moreover, a model is necessary that allows
96 one to consider reasonable forms of non-spherical particles. In this paper the non-spherical mineral
97 particles are approximated as spheroids, since this substantially improves the ~~results~~ ~~agreement~~
98 ~~between modelled and measured optical properties~~ (Mishchenko et al., 1997; Kahnert et al., 2005)
99 and an appropriate ~~scattering~~ theory exists, the T-matrix method (Waterman, 1971).

100 In the new version of OPAC the optical properties of the mineral components are modeled as spher-
101 oids with the T-matrix method (Mishchenko a. Travis, 1998), with the aspect ratio distributions of
102 the used spheroids varied with the particle size, ~~as found by electron microscope investigations~~. The
103 other microphysical properties of the components, the size distribution and the spectral refractive
104 indices, have not been changed against the old OPAC. During the Saharan Mineral Dust Experiment

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105 field campaign (SAMUM-1), which was located close to the Sahara and its mineral sources and used a
106 lot of different aerosol measurement systems (Heintzenberg, 2009), desert ~~dust~~aerosol size distribu-
107 tions have been measured both in situ at an air plane (Weinzierl et al., 2009) and inferred by the
108 AERONET network inversion algorithm from ground-based photometer measurements. The results
109 differ considerably (Müller et al., 2010), but the OPAC size distributions are in-between. Moreover,
110 photometer measurements in the solar aureole (where the non-sphericity has no influence) and val-
111 ues modeled with OPAC type “desert” agree very well (Gasteiger, 2011). Also optical properties of
112 Saharan dust measured by aircraft data in 1999 compare very favorably with OPAC results (Haywood
113 et al., 2001) for radiative properties that are independent of the scattering angle, like asymmetry
114 parameter, single scattering albedo and specific extinction coefficient, for which the non-sphericity
115 has negligible influence. Thus the OPAC size distributions for ~~mineral~~ desert aerosol are assumed to
116 be adequate for a combination with the information on particle shape from SAMUM.
117 Also not changed against the old OPAC is the possibility of the flexible mixing of the components and
118 of the outcome of OPAC, like optical properties depending on relative humidity and available for a
119 large wavelength range. In the new version of OPAC (4.0), which is freely available for non-
120 commercial use, now the optical properties modeled for non-spherical mineral particles are taken
121 into account, directly for practical useapplication.

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124 2. Methods

126 2.1. Non-spherical particle scattering

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128 The most suitable method to model the optical properties of mineral aerosol particles on a systemat-
129 ic basis (Wiegner et al., 2009) is the T-matrix method, TMM. It provides a solution of Maxwell’s equa-
130 tions for the interaction of radiation with arbitrarily-shaped particles (Waterman, 1971) and is most
131 efficient for rotationally symmetric particles. In our model the ~~desert dust~~mineral particles are given
132 as spheroids, originating from rotation of ellipses about one of their axis. Thus, an additional micro-
133 physical parameter that has to be taken into account is the aspect ratio ϵ , which is the ratio between
134 the ~~rotational axis~~longest and the ~~shortest~~ axis ~~perpendicular to it~~ (Dubovik et al., 2006). Moreover,
135 the particles can be prolate (cigar like) and oblate (disk like) spheroids.
136 For the results in this paper and the new version of OPAC, the state-of-the-art TMM code from
137 Mishchenko ~~and~~a Travis (1998) for randomly oriented particles has been used for the mineral com-
138 ponents. The T-matrix calculations are supplemented by geometric optics calculations with the code
139 of Yang et al. (2007) for large particles not covered by the TMM code. Wiegner et al. (2009) show the
140 size coverage of the TMM code, which can model dust spheroids up to size parameters, $x=2\pi r/\lambda$,
141 around 110 – 120 for aspect ratio 1.6 and smaller. For aspect ratio 3.0 the maximum size parameter
142 of TMM is around 25. These codes have been used to create a data set of single particle scattering
143 properties of spheroids covering a wide range of particle sizes, aspect ratios, and refractive indices.
144 The grid of particle parameters in this data set is given in Gasteiger et al., 2011. The optical properties
145 of the OPAC mineral components were calculated from this data set according to their microphysical
146 properties described below. For the selection of the adequate aspect ratio distributions depending on
147 particle size, measurements of the Saharan Mineral Dust Experiments (SAMUM I and SAMUM II) have
148 been used (Kandler et al., 2009; Kandler et al., ~~20~~11).

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151 2.2. Particle properties

153 This paper presents an improvement of OPAC, by modifying the shape of mineral ~~dust~~ particles. The
154 other microphysical parameters used in OPAC, as the particle size distribution and the spectral refrac-
155 tive indices, have been left unchanged.

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156 In OPAC the aerosol particles are given as components (Shettle a. Fenn, 1979; Deepak a. Gerber,
 157 1983) resulting from an internal mixture of particles of a certain origin. The particles of a component i
 158 have a log-normal size distribution (Eq.1).

$$159 \frac{dN_i(r)}{dr} = \frac{N_i}{r \sqrt{2\pi} \log \sigma_i \ln 10} \exp\left(-\frac{1}{2} \left(\frac{\log r - \log r_{mod,i}}{\log \sigma_i}\right)^2\right) \quad (1)$$

160 N_i is the total number of particles of the component i per cubic centimeter, r the particle radius, $r_{mod,i}$
 161 the mode radius of component i with respect to the particle number, and σ_i measures the width of
 162 the distribution. The radius r of each spheroid is assumed to be the radius of a sphere with the orien-
 163 tation-averaged geometric cross section of the spheroid. The relative optical properties do not de-
 164 pend on N , thus they are given always for $N=1$. For absolute values of optical properties, for actual or
 165 individual conditions, N_i must be chosen adequately for each component that will be taken into ac-
 166 count.

167 The mineral dust is described in OPAC with three components as given in Tab. 1: Mineral Nucleation
 168 Mode (MINM), Mineral Accumulation Mode (MIAM), and Mineral Coarse Mode (MICM), with r_{mod}
 169 and σ the data of the size distributions, and r_{min} and r_{max} the borders that have been taken into ac-
 170 count for modeling the optical properties.

171 Tab. 1. Microphysical properties of mineral aerosol components

Component	Mineral...	r_{mod} [μm]	σ	r_{min} [μm]	r_{max} [μm]
Nucleation mode	MINM	0.07	1.95	0.005	20
Accumulation mode	MIAM	0.39	2.00	0.005	20
Coarse mode	MICM	1.90	2.15	0.005	60

Component	Mineral...	r_{mod} [μm]	σ	r_{min} [μm]	r_{max} [μm]
Nucleation mode	MINM	0.07	1.95	0.005	20
Accumulation mode	MIAM	0.39	2.00	0.005	20
Coarse mode	MICM	1.90	2.15	0.005	60

181 These mineral components can be mixed externally, also together with other components, to form
 182 individual aerosol types. In general, both over deserts and for other aerosol conditions with a domi-
 183 nant mass of mineral particles, also water-soluble particles (WASO) are present. These particles can
 184 be assumed to be spherical. Their amount usually is small with respect to their mass per volume, but
 185 since the particles are small their numbers per volume may be large.

186 In OPAC the aerosol type "desert" is a mixture of more than 200 $\mu\text{g}/\text{m}^3$ mineral particles and only 4
 187 $\mu\text{g}/\text{m}^3$ water soluble particles (WASO), however, resulting in 2000 particles per cm^3 of WASO, and 300
 188 cm^{-3} of mineral particles belonging to their three components. A small amount of WASO generally
 189 is taken into account in the following results, which show optical properties of mixtures of mineral
 190 particles.

191 The refractive indices of the components are wavelength dependent (d Almeida et al., 1991; Koepke
 192 et al., 1997). The particles of the mineral components all have the same refractive indices, since they
 193 are assumed to result from the same sources at the surface. The refractive index is given with an im-
 194 aginary part that is responsible for the absorption properties of the particles.

195 To describe the shape properties of mineral particles of different size, for each of the three mineral
 196 components, the data of the "reference" case of SAMUM-1 have been used (Wiegner et al., 2009).
 197 The reference case was a situation with a very homogeneous mineral dust desert aerosol layer up to 5
 198 km above sea level which was very stable in time. The aspect ratio distribution of the particles was
 199 measured using electron microscopy and is given depending on particle size intervals by Kandler et al.
 200 (2009). For modeling the optical properties of mineral aerosol particles these wide aspect ratio dis-

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203 tributions are applied, to account for the large variety of the natural dust particle shapes. The belong-
 204 ing modeling results, compared to measured phase functions, are remarkably better than results
 205 when using only a single aspect ratio (Mishchenko et al., 1997; Nousiainen a. Vermeulen, 2003).
 206 Moreover, all mineral particles are assumed to be prolate, because this gives better agreement with
 207 measured scattering matrix elements of dust particles than ~~those of~~ using oblate or mixtures of
 208 prolate and oblate spheroids (Nousiainen a. Vermeulen, 2003).
 209 It is worth mentioning that the aspect ratio distribution of mineral particles did not vary significantly
 210 during SAMUM-1 and also not during the SAMUM-2 campaign, which was conducted further away
 211 from the dust source Sahara (Kandler et al., 2009; Kandler et al., 2011). Thus the selected aspect ratio
 212 distribution might be regarded as representative for Saharan dust.
 213 The aspect ratio distributions depend on the size of the particles. For the reference case the relative
 214 frequency of particles with a given aspect ratio is available for 6 ranges of particle size (Kandler et al,
 215 2009; Wiegner et al., 2009). Some of them have similar aspect ratio distributions so that only three
 216 radius ranges must be differentiated: For particles with $r < 0.25 \mu\text{m}$ the frequency decreases strongly
 217 with increasing aspect ratio. For particles with $r > 0.5 \mu\text{m}$ the shape distributions for all analyzed size
 218 intervals are similar with a small maximum for the aspect ratio of about 1.5. Between these two re-
 219 gimes the particles between $r = 0.25$ and $r = 0.5 \mu\text{m}$ have an aspect ratio distribution that gives a
 220 transition between the other two regimes (see Tab. 2).

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221
 222 Tab. 2. Aspect ratio distributions as function of particle radius interval according to Kandler et al.
 223 (2009). The first line represents the range from $\epsilon = 1.0$ to 1.3, the last line is valid for $\epsilon > 2.9$ and the
 224 other values cover ϵ -intervals of 0.2.

225

ϵ	$r < 0.25 \mu\text{m}$	$0.25 \mu\text{m} < r < 0.5 \mu\text{m}$	$r > 0.5 \mu\text{m}$
1.2	0.535	0.220	0.103
1.4	0.289	0.205	0.234
1.6	0.108	0.150	0.218
1.8	0.040	0.105	0.157
2.0	0.015	0.070	0.101
2.2	0.007	0.050	0.065
2.4	0.003	0.040	0.041
2.6	0.001	0.030	0.027
2.8	0.001	0.020	0.018
3.0	0.001	0.080	0.036

236

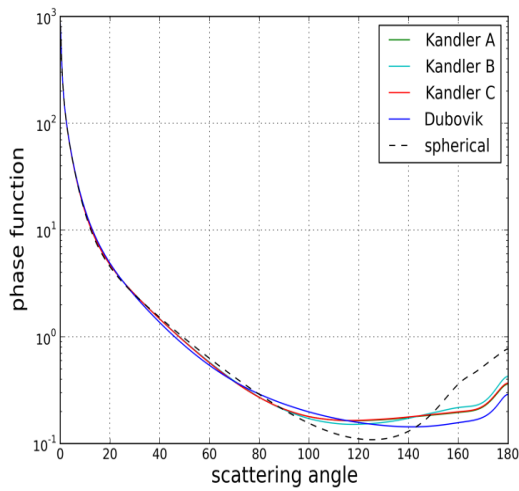
ϵ	$r < 0.25 \mu\text{m}$	$0.25 \mu\text{m} < r < 0.5 \mu\text{m}$	$r > 0.5 \mu\text{m}$
1.2	0.535	0.225	0.103
1.4	0.289	0.212	0.234
1.6	0.108	0.156	0.218
1.8	0.040	0.110	0.157
2.0	0.015	0.075	0.101
2.2	0.007	0.054	0.065
2.4	0.003	0.039	0.041
2.6	0.001	0.028	0.027
2.8	0.001	0.022	0.018
3.0	0.001	0.079	0.036

237
 238 Each OPAC mineral component contains particles in all radius ranges given in Tab. 2, with proportions
 239 that are varying according to the size distribution of the components (Tab. 1). To check the shape
 240 effects, as a first test (Kandler A) each mineral component is divided into the three radius ranges of
 241 Tab. 2 and the belonging aspect ratio distribution of each range is applied. This test is the most exact
 242 approach based on the available aspect ratio data. As a second test -- with respect of the idea of
 243 OPAC to keep things simple -- for all particles of each of the three OPAC mineral components a

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244 fixed aspect ratio distribution has been used: the distribution of $r < 0.25 \mu\text{m}$ for MINM, of $0.25 \mu\text{m} < r$
245 $< 0.5 \mu\text{m}$ for MIAM, and of $r > 0.5 \mu\text{m}$ for MICM (Kandler B). This test setup seems appropriate since
246 the mode radii of the three components (Tab.1) fall into these three radius intervals used to separate
247 the aspect ratio distributions (Tab.2). As a third test (Kandler C), the second test is modified by assum-
248 ing also for all particles of the accumulation mode (MIAM) the aspect ratio distribution that has been
249 measured for particles with $r > 0.5 \mu\text{m}$. This use of the aspect ratio distribution measured for the
250 larger particles also for MIAM was tested, since the maximum of the surface area distribution of
251 MIAM is close to a radius of $1 \mu\text{m}$. Finally a further association of radius and aspect ratio distribution
252 has been tested: Dubovik et al. (2006) has derived aspect ratio distributions by analyzing measured
253 phase functions, with the assumption that they are independent of the particle size. These are inves-
254 tigated as a fourth test (Dubovik) for the particle shape effects.
255 As example for the different considerations of the aspect ratio distributions, in Fig.1 are shown the
256 phase functions under the assumption of spherical particles and for non-spherical particles after the
257 4 tested radius dependent aspect ratio distributions. The phase functions are given for a wavelength
258 $0.55 \mu\text{m}$ (however the results at other wavelengths are similar, see Fig.2), and as size distribution the
259 combination of the three mineral components of the aerosol type "desert" after OPAC, including
260 WASO at 0% relative humidity, has been used.

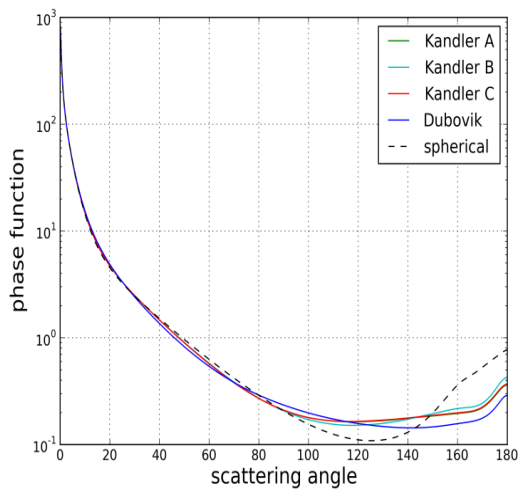
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265 Fig. 1. Phase functions at 0.55 μm for the mixture of the mineral components after the aerosol type
266 “desert”, under the assumption of spherical mineral particles and for mineral particles with various
267 aspect ratio distributions (see text for details).

268
269 In Fig. 1 the increased sideward and reduced backward scattering clearly is to be seen which holds for
270 all phase functions resulting from particles with non-spherical shape. The phase function after
271 Dubovik is clearly noticeably separated against those after Kandler A to C. But this result is not really
272 astonishing, since the direct electron microscopic investigations show that the aspect ratio distribu-
273 tions are size dependent, in contrast to the size-independent assumption by Dubovik. The phase
274 functions after Kandler A (exact approach) and Kandler C are nearly identical, which means that the
275 simpler assumptions in Kandler C give already correct results. Thus for all optical property modeling
276 of non-spherical mineral particles, both for the results shown in the following and for the new OPAC,
277 the size dependent aspect ratio distributions distribution after Kandler C is used.

278
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3. Results

281
282 The effects of the particle shape are different for different optical properties, which is shown in this
283 paragraph for a variation of the optical properties available from OPAC. Examples are presented for
284 the deviations between optical properties caused by mineral particles that are assumed as spheres,
285 on the one hand, and those assumed as spheroids with the aspect ratio distributions after Kandler C,
286 on the other hand.

287
288 As mentioned above, theThe phase function is very important for remote sensing of desert
289 dust aerosol and for its radiative forcing, and moreover, as mentioned above, for this optical quantity
290 the effect due to non-sphericity is large, especially large in the solar spectral range.
291 Thus Fig. 2a shows the phase function for the two particle shape assumptions, for the mixture “de-
292 sert” (Hess et al., 1998) and for different wavelengths. The assumed shape variation (spherical or
293 non-spherical) is modeled only for the mineral particles: MINM 269.5 cm^{-3} , MIAM 30.5 cm^{-3} , MICM
294 0.142 cm^{-3} . The 2000 cm^{-3} particles of the WASO component always are assumed as spherical.

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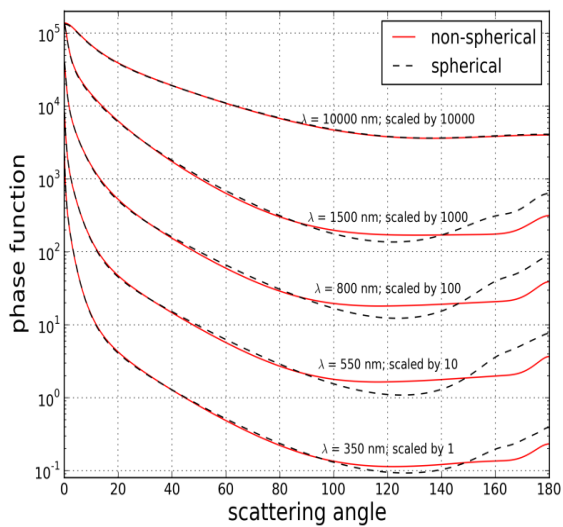
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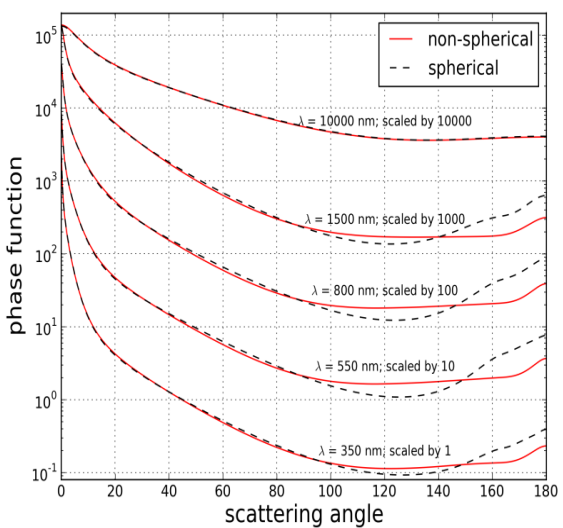
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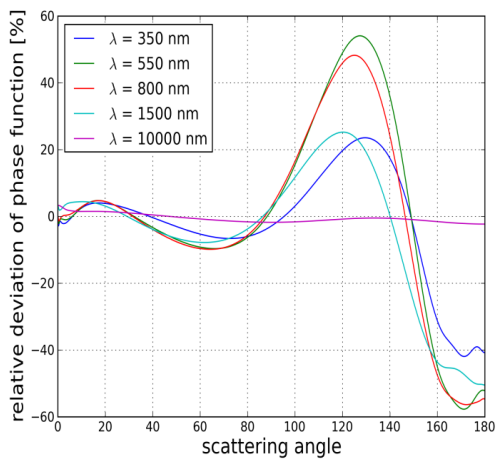
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Fig. 2a. Phase functions of “desert dust” aerosol for different wavelengths, assuming spherical and spheroidal mineral particles with a size dependent aspect ratio distribution after Kandler C. For details see text. The scale of the phase functions for the different wavelengths is shifted by a factor 10 in each case.

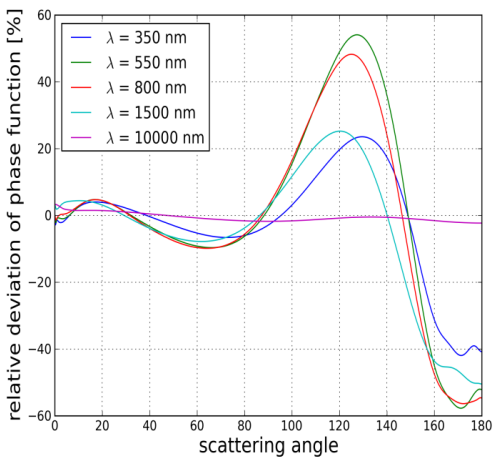
The phase functions show the known strong forward peak of aerosol particles, which is not influenced by the particle shape. It is increasing with increasing size parameter, and thus decreasing with wavelength. The particle shape effect is clearly to be seen in Fig. 2a in the backward scattering region, but more pronounced in Fig. 2b, where the belonging percentage deviations between the phase functions for particles with size dependent aspect ratio distributions and for spherical particles are shown.

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312 Fig. 2b. Relative deviations (%) of phase functions assuming spheroidal mineral particles from phase
 313 functions of spherical particles, for "desert dust" aerosol and the conditions shown in Fig. 2a.

314

315 The effect of the particle shape is up to almost + 60 % at scattering angles around 130° and – 60 %
 316 around 170°, in the backscatter region. The effect decreases with increasing wavelength, since and is
 317 nearly negligible at 10 μm, as also shown. The reason is that the shape properties of the particles
 318 become less relevant if the wavelength of the radiation becomes larger relative to the particle size. In
 319 contrary, the effect of the particle shape is relatively low at 350 nm, but this result#results from the
 320 strong absorption of the mineral particles at this wavelength, which reduces the scattering effects in
 321 general and thus overcompensates the shape effect. As to be seen, the effect of the particle shape is
 322 strongest in the solar wavelength range, which is generallyoften used for aerosol remote sensing and
 323 which is essential for radiative forcing and thus for climate effects. This documents again the need to
 324 take the non-spherical particle-shape of desert dust mineral particles into account for remote sensing
 325 or climate studies.

326 As mentioned, the aspect ratio distribution depends on the particle size. Thus size distributions with
 327 different amount of small and large particles may result in different variations of the phase function
 328 compared to that under the assumption of spheres. Since the life time of big particles in the atmos-

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329 there is less than that of smaller particles, in a dust storm not only the total amount of mineral parti-
 330 cles in the air is high, but also the relative amount of large particles. During the transport, i.e. with
 331 the time after the dust generation, the particle amount will be reduced due to sedimentation, but
 332 this effect is can be stronger for larger particles. Finally, for background conditions, the total amount
 333 of mineral particles is low, with strongest reduction for the lowest amount of large particles
 334 (d'Almeida, 1987; Longtin et al., 1988; Tanré et al., 1988). The relative increasing amount of large
 335 particles with increasing turbidity is given that we assume to test the effect of non-sphericity with
 336 respect to particle size distribution is shown in Eqs. 2 – 4 (d'Almeida, 1987; Koepke et al., 1997). Given
 337 are correlations between the total number of desert mineral dust particles and the belonging
 338 numbers for the three mineral components.

339

340 $\ln N_{MINM} = 0.104 + 0.963 \ln N_{mineral}$ (2)

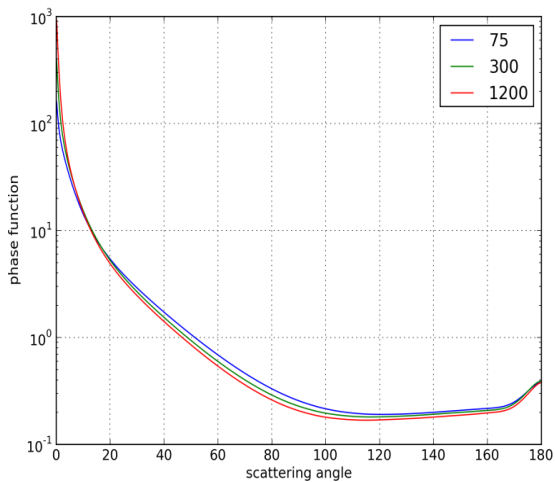
341 $\ln N_{MIAM} = -3.94 + 1.29 \ln N_{mineral}$ (3)

342 $\ln N_{MICM} = -13.7 + 2.06 \ln N_{mineral}$ (4)

343

344 For desert dust aerosol with different turbidity, given implemented with different total particle num-
 345 ber and belonging different number of particles of the three mineral components, in Fig. 3 the phase
 346 functions for non-spherical desert particles are shown. N gives the total number of mineral particles,
 347 the value of $N_{mineral}$ in Eqs. 2 - 4. N = 75 stands for "background desert" conditions, N=300 for average
 348 "desert" and N=1200 for "dust storm". It can be seen that the general effect of the non-spherical par-
 349 ticle shape is always given, but does not differ considerably for the different size distributions, as result
 350 of different total particle number. The effect of varying size distribution is more pronounced in
 351 the forward peak and the sideward scattering.

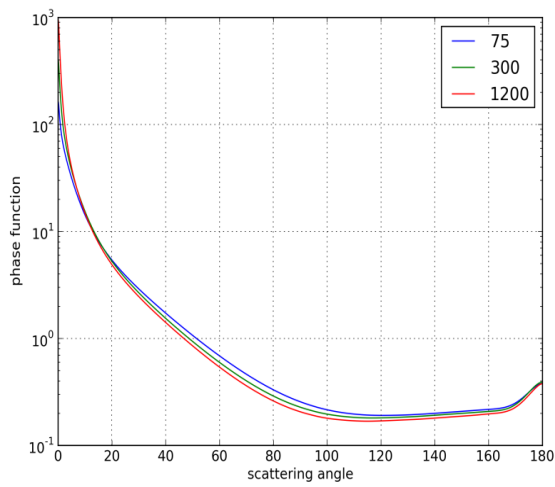
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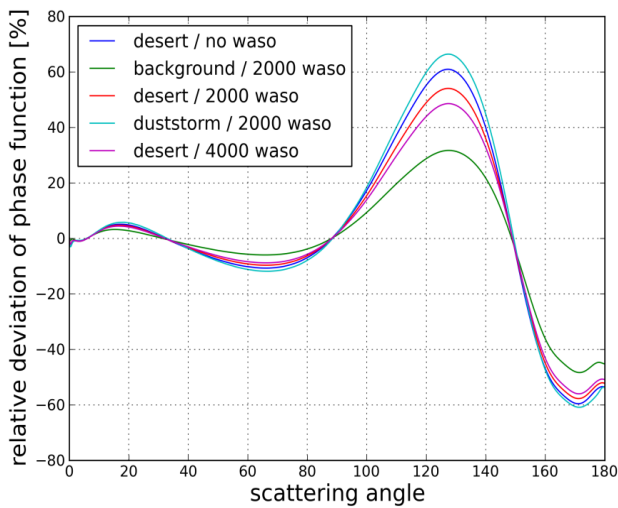
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356 Fig. 3. Phase functions of ~~non-spherical~~ desert aerosol ~~particles~~ at 0.8 μm, with a mixture of the ~~non-~~
357 ~~spherical mineral~~ components MINM, MIAM and MICM after the Eqs. 2-4, using the total number of
358 mineral particles given in the figure. In each case 2000 WASO particles assuming 0% r. h. are included.
359

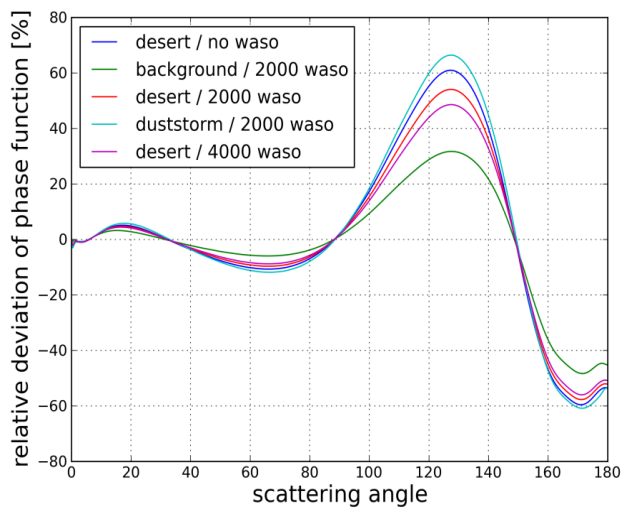
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361 As mentioned, the WASO particles are spheres, with the consequence that the variation of their
362 amount changes the phase function of the mixture. This is shown in Fig. 4 for “desert” with different
363 amount of WASO, on the one hand, and for average amount of 2000 WASO particles, but in combina-
364 tion with mineral particles for “background” and for “dust storm” conditions, on the other hand.
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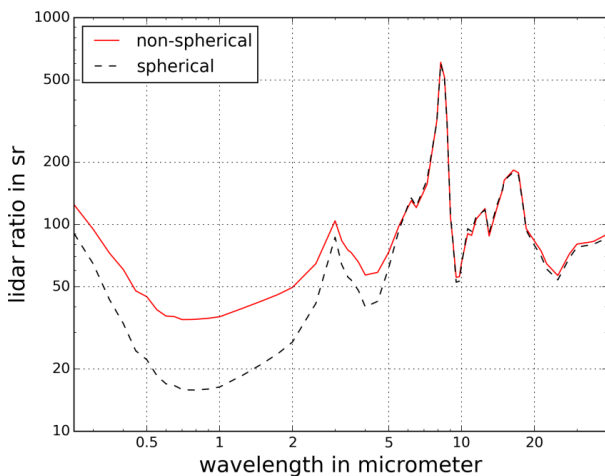
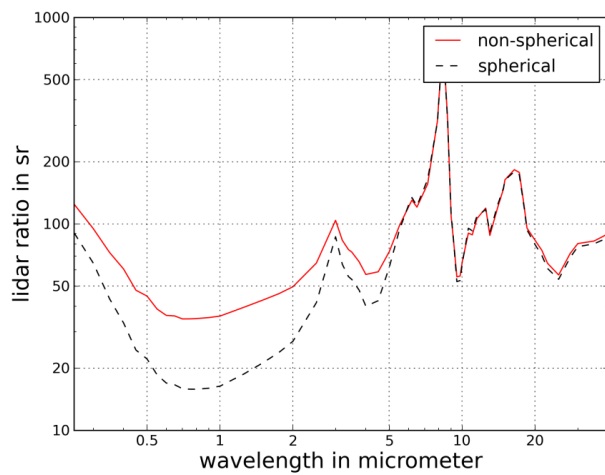
Fig. 4. Relative deviations (%) of phase functions at $0.55 \mu\text{m}$, assuming spheroidal mineral particles, from phase functions of spherical particles, for different combinations of the components WASO, MINM, MIAM and MICM (for details see text).

374 Figure 4 shows that the effects due to the particle shape increase from background over desert
375 dust storm if the number of WASO is fixed, simply due to the increasing amount of non-spherical
376 mineral particles. In contrary, the effect due to non-spherical shape is reduced, to be seen for the
377 type “desert”, if the amount of spherical WASO particles is increased. But it should be mentioned that
378 the effect due to doubling or omitting WASO for the relative deviations of the phase function is less
379 than the effect due to the variation of the amount of the mineral particles.

380 For the determination of the height dependent aerosol extinction coefficients, often backscatter lidar
381 systems or ceilometers are used, because they are cheaper than higher sophisticated lidar instru-
382 ments (Mona et al., 2012; Wiegner et al., 2014). However, for these instruments the measured signal
383 is result of both the scatteringextinction coefficient and the phase function at 180° . Thus, to get the
384 interesting height dependent extinction coefficient, it is necessary to use a quantity “lidar ratio”,
385 which depends on the phase function and thus on the particle shape.

386 Fig. 5 shows the lidar ratio for the aerosol type “desert”, both under the assumption of non-spherical
387 and spherical mineral particles. The values are given for a wavelength range up to $40 \mu\text{m}$, although no
388 lidar instruments are available for wavelength larger $\approx 2 \mu\text{m}$. Moreover for the large wavelengths, the
389 particles behave more and more like spheres, as already to be seen in Fig. 2b. For the interesting
390 wavelength range around and below $1 \mu\text{m}$, however, the consideration of non-sphericity is essential.
391 With respect to independently measured lidar ratios, the agreement with modeled values is much
392 better under the assumption of spheroids than of spheres (Gobbi et al., 2002). The lidar ratios to be
393 seen in Fig. 4 are in good agreement with measured values from SAMUM (Groß et al., 2011). This also
394 generally is valid for all lidar-wavelengths that have been used during SAMUM, but here the agree-
395 ment between measured and modeled lidar ratios was reduced for 355 nm , probably due to wrong
396 assumptions with respect to the refractive index (Wiegner et al., 2009).

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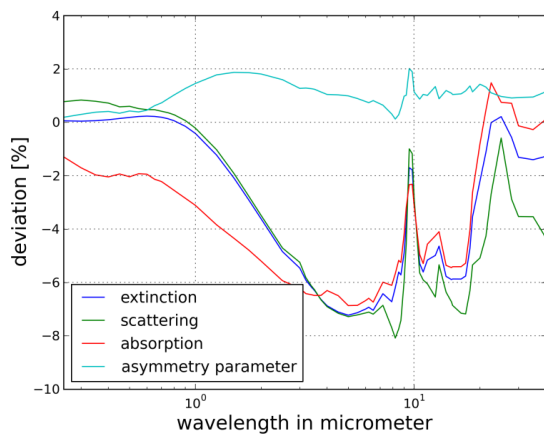
402 Fig. 5. Modeled values of the lidar ratio for "desert" aerosol under the assumption of spherical and
 403 non-spherical particles.

404

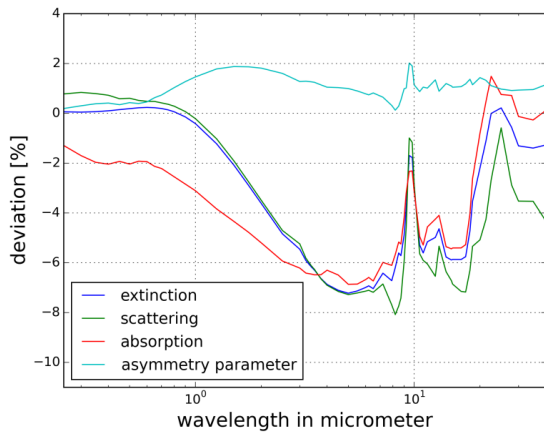
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406 Optical quantities that are independent of the scattering angle or given as ratio between wavelengths
 407 are expected to be less sensitive with respect to the particle shape. To investigate this aspect, in Fig. 6
 408 relative differences between spherical and non-spherical desert particles are presented for the spec-
 409 tral scattering -, absorption - and extinction- coefficients and for the asymmetry parameter. For all
 410 these quantities the deviations are less than 6 % and even less than 4 % in the part of the solar spec-
 411 trum that is most relevant for climate effects. The same low dependency on the particle shape also
 412 holds for the single scattering albedo and the Ångstrom coefficient, not shown in a figure.

413



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417 Fig.6. Deviation (%), between spherical and non-spherical “desert” aerosol for different optical quantities
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4. New version: OPAC (4.0)

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The main improvement of the new version of OPAC is the consideration of the non-sphericity of ~~desert dust~~ mineral particles. In OPAC always the large wavelength range between 0.25 and 40 μm is considered, with the consequence that the improved particle shape of mineral particles works both in the solar and in the infrared spectral region. Additionally new in OPAC (4.0) is the possibility to model ~~PM10, PM2.5 and PM1~~ the particle mass for ~~the individual mixtures of components~~ different cut off radii, as used e.g. for PM10. On the other hand, the component “mineral transported”, MITR, no longer is considered. This component had been used to describe desert aerosol under very remote conditions, as part of aerosol in polar regions. However, the amount of ~~desert mineral~~ dust particles should be reduced continuously on its way from the source, depending on their life time, ~~which~~. This is possible with the remaining mineral components (e.g. using Eqs. 2-4), instead of switching to MITR. Thus the aerosol type “~~antarctic~~“Antarctic” in OPAC has been modified.

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435 As discussed in the paper, the shape of the mineral particles has been improved. To avoid mistakes,
436 the new mineral components are named in the new OPAC version with an N at the end, standing for
437 non-spherical.

438 The change from spheres to spheroids was made on the basis of cross section equivalence, resulting
439 in a small reduction of the particle volume and thus the particle mass, with resulting in the reduction
440 factors shown in Tab. 3.

441

442 Tab. 3. Reduction factors for particle volume and mass for the non-spherical mineral components,
443 compared to the old components.

444 ~~MINM >> MINN 0.9754~~

445 ~~MIAM >> MIAN 0.9273~~

446 ~~MICM >> MICN 0.9273~~

447

<u>MINM → MINN</u>	<u>0.9754</u>
<u>MIAM → MIAN</u>	<u>0.9273</u>
<u>MICM → MICN</u>	<u>0.9273</u>

448

449 All the other microphysical aerosol properties are unchanged against the previous version of OPAC.

450 Also the new version of OPAC gives the possibility to combine different aerosol components, in each
451 case with individually decided particle number density for each component.

452 Results of OPAC (4.0) are ~~microphysical properties, like particle mass per volume and PM10, and the a~~
453 large number of optical properties (like phase function, scattering- absorption- and extinction coeffi-
454 cient, asymmetry parameter, single scattering albedo, Ångstrom coefficient, ~~and lidar ratio~~), and visi-
455 bility) and particle mass per volume. All properties can be modeled for different relative humidity and
456 the optical properties are available as spectral values for the wide wavelength range of 0.25 to 40 µm
457 and spectrally weighted for the solar and terrestrial range. For non-commercial use OPAC (4.0) is
458 freely available: www.rascin.net.

459

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461

462 5. Conclusion

463

464 Aerosol particles are one of the main gaps in the present knowledge of radiative forcing (Myhre et al.,
465 2013), and mineral ~~partiele~~particles are especially essential due to their large amount and temporal
466 and spatial variability. Since mineral particles in general are no spheres, Mie-theory ~~as scattering the-~~
467 ~~ory~~ may lead to wrong values ~~of, both, if~~ their optical properties, ~~if they~~ are modelled based on size
468 distribution and refractive index, and vice versa, if remote sensing data are used to get aerosol prop-
469 erties. Thus, ~~as a major improvement~~, the optical properties of mineral particles in the new version of
470 OPAC are derived using T-Matrix method for spheroids. ~~As described in~~ this paper the non-
471 sphericity is ~~described given~~ by typical size dependent aspect ratio distributions of spheroids, which
472 have been derived from measurements at observation campaigns. The predefined components in
473 OPAC, now also for non-spherical mineral particles, are ~~an advantage of OPAC a big convenience~~, be-
474 cause users do not need to decide for individual single particle properties, as available from various
475 studies and data bases (Nousiainen, 2009; Meng et al., 2010).

476 The differences between spherical and non-spherical mineral particles are shown for a wide range of
477 optical properties of desert ~~dust~~aerosols. They are small, nearly negligible, for angular-independent
478 optical quantities, like extinction-, scattering- and absorption- coefficient, asymmetry factor, single
479 scattering albedo and Ångstrom coefficient. However the differences between spherical and non-
480 spherical particles are large, up to 60 %, in the sideward and backward scattering regions of the phase
481 functions. in the solar spectral range. As a consequence the deviations also are large in the lidar ratio,
482 a parameter required to get height dependent extinction values from often used backscatter lidar
483 measurements. The effect of the particle shape decreases with wavelength, since for wavelengths

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484 that are rather large with respect to the particle size, the irregular particle shape is of less relevance.
485 ~~However, in the solar spectral range the shape effects can be large. Since this is the wavelength range~~
486 ~~that is generally~~
487 ~~It should be born in mind that the size distribution and the complex refractive index of the aerosol~~
488 ~~particles are very important for their optical properties. For the radiative properties in the thermal~~
489 ~~infrared the uncertainty in the refractive index will outperform the shape effect, which moreover~~
490 ~~depends on the absorption of the particles (Legrand et al., 2014). However, in this article only the~~
491 ~~aspect of the shape of mineral particle is discussed, and in the new version of OPAC the shape of the~~
492 ~~mineral particles has been improved, but the assumed size distributions and spectral refractive indi-~~
493 ~~ces have not been changed. This will be done in the future, where it is planned also to add a stronger~~
494 ~~absorbing mineral component that allows for a larger variability of mixtures to describe desert aero-~~
495 ~~sol.~~
496 ~~Since the solar spectral range is often~~ used for remote sensing of aerosol particles, on the one hand,
497 and relevant for aerosol radiative forcing, on the other hand, ~~the use~~ consideration of the phase func-
498 tions of non-spherical mineral particles ~~will be~~ is a real improvement. ~~To allow an easy use of the opti-~~
499 ~~cal properties of desert aerosol with non-spherical particles, the data are made~~ OPAC, now available
500 ~~in OPAC (as version 4.0).~~

502 Acknowledgements

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