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Interactive Comment

Interactive comment on "Modelling light scattering by mineral dust using spheroids: assessment of applicability" *by* S. Merikallio et al.

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Anonymous Referee #3:

"This is a nice and comprehensive study on scattering properties of non-spherical mineral dust compared to measurements. I urge publication in ACP because the presented facts are worth knowing and of highly scientific interest. I have some comments and questions listed below in detail which can be summarised as follows: a) What about the possible influence of other types of size equivalence? b) What about the validity of the shape distribution applied? c) How do the polarisation and particle measurements a la Volten et al. really fit together? Some statements with respect to these questions should be included in the paper (Section 6?). It seems to me that the spheroidal model is rather discredited, although not all uncertainties related to the questions a) to c) are

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discussed or cannot be discussed in one (this) paper. I am aware that the spheroid model is only an approximation but it is the best we have (we are able to compute the scattering properties for most situations half-decently). On the other hand, this paper demonstrates clearly that we must not rest."

Answer: We thank the referee for his thorough and positive comments. A description of all changes we made in the text in response to the comments, and an itemized answer to all questions is given below. Referee comments are in *italic*. The revised manuscript can be found in the supplement.

Detailed comments and questions: 1) Page 3978, Line 24/25: The size of the dust particles are important. As noted later in the manuscript size distribution measurements are difficult, e.g., to measure coarse mode particles.

Answer: We agree with the reviewer that size is important. We merely quote from the paper of Myhre and Stordal, which concluded that, although size is important, the refractive index is even more important. To reduce the risk for misunderstanding, we changed "to a lesser extent" to "to a slightly lesser extent". This should make it clearer that we make a statement about the relative importance of different sources of error, not about the absolute importance.

2) Page 3979, Line 28: What is meant with 'larger'? 30, 50 or 150?

Answer: Most previous fitting studies based on spheroids only consider the feldspar sample which has a size distribution representative of background conditions but not those for dust events. Here we consider samples which have effective size parameters up to over 8 times larger. For clarity, we now added some details in the sentence in question and it now reads "Little is known about the performance of the spheroidal model particles for mimicking scattering by dust particle ensembles with effective radii larger than about 1 um."

3) Page 3981, Line 13: Obviously, volume equivalence is assumed. However, this is

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only one possible assumption. I think that the quantity 'size equivalence' is an important free parameter. Applying another type differing results are obtained. It would be nice to see results for various kinds of size equivalence. But I suspect that this would mean too much effort beyond the results of this paper.

Answer: We agree with the reviewer that considering other measures of size equivalence would involve substantial extra work. However, they were not completely overlooked, we did perform some tests also with area-equivalent particles initially, but as these did not produce significantly altered results, we chose to consider only the volume equivalence. Due to the very large amount of different analyses conducted, the use of multiple size equivalences would have been quite a complication. Also, we note that there are important applications in which it may not be desirable to allow the measure of size equivalence to be a free parameter. For example, in chemical transport models coupled with a so-called 'double-moment' aerosol dynamics model, both the mass and number size distributions are prognostic parameters of the model. Therefore such models predict the mass per particle in each size bin. (Such models are employed both in air pollution forecasting, and in modern Earth-system climate modelling, in which atmospheric chemistry, aerosol dynamics, and atmosphere-ocean general circulation are dynamically coupled. Aerosol optics models are included in such models either for computing aerosol climate forcing, or for assimilating remote sensing observations of aerosols.) By using any measure for size equivalence other than volume equivalence, one manipulates the mass per particle. This basically means that one would discard one of the main prognostic parameters of the model (either size-resolved mass or number concentration), just for making the aerosol optics computations somewhat more convenient. Thus the only measure of size equivalence that is consistent with a double-moment aerosol dynamics model is the measure of volume equivalence.

We have now added a mention of this in the manuscript in the beginning of page 3983, right after the passage " The volume-equivalent size is assumed." as: "The use of area equivalence was also briefly tested, but its performance appeared to be comparable to that of the volume equivalence in reproducing the measured scattering, so further considerations using different size equivalences were deemed unnecessary."

We also added in first paragraph of Conclusions "The volume-equivalent size has been assumed." and couple of paragraphs after (p. 3998 last paragraph): "The impact of using a different size equivalence would most likely not have extended beyond minor details in the results. In particular, it is noted that different size equivalences weight different aspect ratios differently, which can be partially compensated by the shape distribution weights, thus the retrieved values of n might be somewhat affected. "

4) Page 3982, Line 10, Eq. 6: As I understood the expression, case n=0 means an equiprobable distribution. It would be interesting to get an impression of the functional relationship, e.g., by plots of shape distributions (typical for the paper) as a function of the spheroid axis ratio. How do these distribution fit to measurements, e.g., carried out recently during the SAMUM campaign?

Answer: Comparison of measured and fitted shape distributions would be interesting, but there is also much potential for misunderstandings. For one thing, the shape distributions of our samples may not be similar to those obtained during the SAMUM campaign. Second, a shape distribution of spheroids that reproduces scattering by a distribution of real dust particles best may not be similar to the actual dust-particle aspect ratio distribution. Indeed, we have investigated this in another manuscript currently under revision, confirming that shape distributions that best match the shapes and best match the scattering can be very different. Thus, if we added such a comparison here, we would have to add considerable amount of explanations to avoid any misunderstandings. Therefore, we have decided against it. Figure 4 shows the fitted shape distributions considered in the paper. We have now added a n=3 curve in the plot 4 and mention of it in the text: "Also n = 3 line (red) is plotted in the figure for reference."

5) Page 3982, Line 14/15: The database of Mr. Dubovik was applied which uses different scattering codes, since the Mishchenko code converges only for size parameter lower than approximately 50. The larger the axis ratio the less convergent the code, especially in the transition range (to other codes, e.g., of Mr. Yang/Liou) of the axis ratio of about 2:1. How well does the database map the transition from one to another scattering code? This is important since axis ratios up to 2.8 are applied. Beyond, as I know scattering kernels are saved in the database. How are, e.g., the cross sections derived numerically from these kernels?

Answer: The referee is correct that two different methods have been used to compute the kernels of the Dubovik's database. The convergence of these methods is discussed by Yang et al. 2007 ('Modeling of the scattering and radiative properties of nonspherical dust-like aerosols', J. Aerosol Sci., vol. 38, pp. 995 – 1014). As can be seen, the convergence is good. As to the cross sections, they are included in the database output. In our case, wet only need them for weighting when computing shape-distribution integrated single-scattering properties. To clarify this in the text, we changed the sentence in page 3982: "We make use of a database of pre-computed phase matrices for mineral dust particles" into "We make use of a database of pre-computed single-scattering properties for mineral dust particles". And added a sentence "Scattering cross sections are also extracted, as they are needed for weighting when computing shape-distribution integrated quantities."

6) Page 3982, Line 20-23: Some literature to the values applied would be helpful to stress these choices of the refractive indices.

Answer: We have adopted the refractive indices from Volten et al. (2001) and Munoz et al. (2001), where they were estimated based on mineralogical composition and some literature values. We added a clarifying sentence on the text: "These values are based on the estimated range of m provided by Volten et al. (2001) and Munoz et al. (2001)."

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7) Page 3982, Line 29: As noted in 3), volume equivalence is only one possibility.

Answer: We made some clarifying changes in the manuscript as noted in Answer 3, above.

8) Page 3983, Line 11: 'corresponding size distribution'. I do not understand this. The size distribution is constant for the sample, right?

Answer: We mean the size distribution corresponding to the particle type measured, i.e. loess. We wanted to stress that each of the colored lines in Figure 1 is calculated over a size distribution of the loess sample. We have tried to clarify by changing "An example of a measured Mueller matrix (with error bars) is shown in Fig. 1 along with example computations for varying spheroids with the corresponding size distribution." into: "An example of a measured Mueller matrix (with error bars) is shown in Fig. 1 along with example computations of spheroids integrated over the size distribution of the loess sample."

9) Page 3983, Line 14/15: Could the authors summarize the accuracy of the particle measurements? Were the size distributions measured parallely to the polarisation measurements to ensure that always the same sample was considered? The authors know about the difficulties of size measurements. Couldn't it be possible that there are inconsistencies between both measurements? Can it be screened out that the polarisation measurements only 'saw' smaller particles due particle losses in the experimental setup?

Answer: The accuracy of the scattering measurements are indicated by the error bars. The accuracy of the size distributions is not known. The size distribution of each sample have been measured separately from the light scattering measurements. The aerosol generator that produces the aerosol beam can work with particles smaller that 100 micron in radii. When particles are larger (or of the order of) 100 micron, the larger particles may remain in the aerosol generator i.e. they are not measured by the scattering setup. Therefore it is true that there might be some size selection, but just for

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particles larger that 100 micron in radii which are very scarce in the studied samples. We are just using published data and are not intimately accustomed to every step taken to guarantee the data quality. We have, however, spoken to the authors responsible for the measurements on multiple occasions in conferences and are confident that these are the best data currently available for this kind a study. Every attempt has been made to make the measurements as reliable as possible.

Further, in Nousiainen (2009) there is a plot where spheroids are used to simulate the scattering matrices using the refractive index and the same shape distribution for all the samples, using the same database as done here. The different samples show surprisingly little differences in their scattering matrix elements, implying that small errors in the size distributions would not impact our analyses much. The differences between the size distributions of different samples are, after all, larger than the uncertainties we can expect from the size distribution measurements.

10) Page 3983, Line 18-19 and Page 3984, Line 8-11: How representative are then the samples?

Answer: The representativeness is difficult to quantify. The samples are composed of the same minerals observed in atmospheric dust. The samples cover well the observed effective radii obtained during the SAMUM experiment, for example. Some of the samples, such as the Loess, are actually obtained from deposits of wind-transported dust, while some samples have been artificially generated by crushing larger rocks (e.g., the feldspar sample). If full scattering matrices existed for samples collected from the air, we would use those. Alas, no such measurements currently exist. Our thinking is that if spheroids work for these samples, then they should work also for atmospheric dust. The size distributions and refractive indices may wary, but these can be changed to match the airborne dust if the proper values are known. The shape distribution is probably the biggest issue, which is one reason why we are looking for a generic shape distribution that would perform adequately for different dust samples and thus, hopefully, for a large range of dust particles expected in the atmosphere. Further remarks were added in the text in Chapter 5: “Also, possible shape distribution differences between available measured samples and real atmospheric dust lead us to seek for a generic shape distribution that would work for a large range of dust particles thus also including those in the atmosphere. ”

11) Eqs. 7 and 8; Page 3985, Line 2: The cross section of a spheroid is not Pi times $r2$. How is the shape really be considered in size integrations? How is the integration performed with regard to the volume equivalence case? What size ranges were considered, e.g., what minimum/maximum particle diameter assumed?

Answer: Equations (7) and (8) are quite generic equations that could be used to describe either real dust particles or the model spheroids. In either case, some kind of size equivalence needs to be chosen, and r refers to equivalent radius based on this choice. The size integration is done in the same way regardless of the size equivalence, in other words, by weighting by the number concentration and scattering cross section of the corresponding size. The referee is correct that the mean cross-sectional area of a spheroid is not pi*r^2 if r refers to the volume-equivalent size. However, we use these equations only for describing the measured samples. In the size measurement, the size is probably neither volume- nor area-equivalent size, but rather some kind of optically equivalent size. As far as we can tell, only the instrument manufacturer knows what it is exactly, the details of the retrieval algorithm being commercial secrets. When we use spheroids with our size equivalence, we are essentially assuming that the measured r are volume-equivalent radii. We have not corrected the values, because we do not know what they really are and thus do not know how to correct them. We are fairly certain, however, that possible errors caused by this are not large compared to other uncertainties involved in size distribution measurements.

When Eqs. (7) and (8) [Eqs. (8) and (9) in the revised manuscript] are used, the whole measured size range is used. The lower limit is about 80 nm for all samples, but the upper limit varies between samples. When the corresponding single-scattering properties are retrieved from the database, we are subject to the size limitation of the

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database: particle sizes are fully covered at the small end, but at the large end, we can only go up to size parameter 625. At wavelengths 442 and 633 nm it corresponds to particle radii of about 44 and and 63 micrometers, respectively. The samples measured contain a few particles larger than these, but not sufficiently many to influence the size-averaged scattering matrix elements noticeably. This can be tested, for example, by using a Mie theory which is not subject to the size parameter limitations of the database.

See also previous answer #9 and the answer to remark #2 of Referee #1.

12) Page 3986, Line 3: Here, one could also refer to recent results, e.g., from SAMUM.

Answer: We now added a sentence: " More recently, physical and optical properties have been measured for different size classes of airborne Saharan dust in the SAMUM campaign (Heintzenberg, 2009). Measured refractive indices were found to be varying in-between different size classes (Otto et al., 2009), which is not surprising considering that also the chemical composition was found to vary (Kandler et al., 2009)"

13) First two paragraphs of Section 4.1: To define 'coverage', are here single spheroids considered, that is, no shape distribution? A more clear description would help the reader.

Answer: Yes, no shape distribution, but each shape aspect ratio as its own. To clarify the text, we changed the word 'shapes' into 'aspect ratios' and inserted a word 'single': "The term 'coverage' refers to the percentage of measurement points for which the measured values overlap with the range of values calculated for single spheroids of different aspect ratios."

Also, we have rephrased the coverage introduction as: "If a measurement point lies outside the range of those matrices covered by differen aspect ratios, then it is impossible to fit that measurement point with any shape distribution. This leads us to consider how well this necessary condition for successful fitting is met for different samples. The

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non-linear fits are only performed for selected cases and are considered in more detail in Section 4.2.

Investigations on how well the measured scattering-matrix elements can be covered by spheroids of different shapes and refractive indices are thus performed. The term 'coverage' refers to the percentage of measurement points that are within the range obtained by considering the spheroids size-integrated values for all aspect ratios separately. This gives an indication of how well the measurements can be modeled by using spheroids."

14) Page 3987, Line 3: 'spheroids' –> 'spheroid'?

Answer: Thank you, corrected.

15) Page 3987, Line 7-11, 16-20: Would it be possible that the measurement merely detected smaller particles and larger ones were lost?

Answer: This is very unlikely. When the aerosol particles come out of the tube, just above the measurement laser, it is the smallest particles that would most likely move away from the aerosol jet before hitting the laser. We are under the impression that no detectable loss of small particles takes place, however. On the other hand, particles larger (or of the order of) 100 micron might remain in the aerosol generator i.e. they are not measured by the scattering setup. Therefore it is true that there might be some size selection but just for particles larger that 100 micron in radii. Then again, particles this large are not included in the simulations, either.

16) Fourth paragraph of Section 4.2: Could the authors present (or refer to literature) a formula of the shape distribution (not only a proportionality as in Eq. 6) as a function of the weights?

Answer: We are not sure if we understand the question correctly. The shape distribution p(ξ) does provide the integration weights for the shape-averaging, e.g.

$$< C_{sca} >= \int C_{sca}(\xi)p(\xi)d\xi.$$

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To replace the proportionality in Eq. (6) by an equality, you simply include a normalisation factor, i.e.

$p(\xi) = N|\xi|^n, n \geq 0,$

$N = \int_{\xi_{min}}^{\xi_{max}} |\xi|^n d\xi.$

We have replaced Eq. (6) in the revised manuscript by the normalised shape distribution given above.

17)Page 3988, Line 26: Shouldn't be the exponent outside the absolute signs?

Answer: Yes, corrected, thank you.

18) Page 3990, Line 15-16: Couldn't it be possible that the assumed shape distribution is not entirely accurate? See also point 4). What about a shape distribution of lognormal shape centered at a typical axis ratio found by in-situ measurements reported in literature?

Answer: For sure the assumed shape distribution is not entirely accurate, especially when parameterized with only one parameter, in this case the exponent n. But as the n-distribution produces pretty good results overall, it is telling that the values of n differ so much when different criteria are used. We did consider many differing shape distributions with little additional benefit, including distributions smoothly peaking at some value of xi in mid-range of our n-value spectrum (n= 0-18). These were not very successful, improving on power-law distribution only sporadically, and were thus abandoned from further analyses. As mentioned in the manuscript, we also tried fitting arbitrary shape distributions with the measurements. These resulted in distributions that favoured high-aspect ratio spheroids, resembling our parameterized distributions with $n > 0$ (Figure 4.). AERONET shape retrievals of atmospheric dust particles reported by Dubovik et al. (2006) also resulted in shape distribution that favored high-aspect ratio spheroids. This is also inconsistent with the in-situ measured shape distributions and further shows that shape distributions resulting in optimal scattering do not neces-

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sarily correlate with actual shapes. In our study, this is evident from obtaining different shape distributions at different wavelengths.

We have added a sentence on the first paragraph of chapter 4.3.: " We also tested other shape distributions, which is discussed in the end of this chapter." See also answer to question 4. Additionally, we added in text, after introducing the power law (p. 3982): "AERONET shape retrievals of atmospheric dust particles reported by Dubovik et al. (2006) also resulted in shape distribution that favored high-aspect ratio spheroids."

19) Page 3991, Line 10-12: Isn't it valid only for the shape distributions applied in this paper?

Answer: Yes, it is. But we also tried out different distributions including those that consist of only oblate or only prolate model particles, which are briefly discussed in the end of section 4.3. As these other distributions produced no improvement over power-law distribution, we abandoned them from further analyses.

20) Page 3994, Line 6-8: This is true, but isn't it in contrast to the statement in the introduction (Page 3978, Line 25)?

Answer: In page 3994 we are considering matching of the matrix elements whereas in the study cited in the Introduction, error sources for radiative impacts were considered. The one big difference is that a size distribution error can contribute quite a lot to the optical depth of aerosols even if their impact on the scattering matrix elements would be small. Or, it is possible that their impact on optical depth and scattering matrix elements partially compensates for each other, in which case we would be more affected by the size distribution inaccuracy than the radiative impact considerations. So, it is possible that size distribution errors are significant for one application and not the other.

In our case the size distribution error would probably have most impact through diffraction extrapolation and would impact the P11 element the most. The element ratios are not affected by this except at the extrapolated forward angles, and since these angles do not have substantially larger values compared to other angles, errors there would not have large impact. For P11 they could, especially if they caused the measurement points to shift out of the coverage range.

21) Page 3994, Line 20-22: The versatility was 'only' tested for one type of shape distributions and size equivalence.

Answer: Other types of shape distributions tested were mentioned in chapter 4.3, but mentioning this was missing in conclusions. We thus replaced the sentence "It is found that parameterisations with more free parameters do not lead to consistent improvements." with "Other types of shape distributions were also considered, some with more free parameters, but they did not result in any significant or consistent improvements."

22) Page 3995, Line 3: Size equivalence is also an important free parameter which was not investigated.

Answer: See answer to question #3.

23) Page 3997, Line 19: 'validity', see 21).

Answer: The range of validity was investigated using many different types of shape distributions, including single shapes (coverage) and completely arbitrary best-fit shape distributions.

24) Page 3998, Line 6-10: See 15). If the measurements were performed in the presence of merely smaller particles, the respective effective refractive index might be different to that one of the total ensemble because chemical composition and hence refractive index may vary as a function of particle size.

Answer: This is true although there is no evidence that only large particles would be missing, except maybe those above 100 microns. The choice of best-fit refractive index was done based solely on model performance and could have been different, were the measurements done by different means. There are many things we might have done differently and some of these paths will be in the future studies. Size-

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dependent refractive index would make fitting more difficult than it already is, but it could be considered. Likewise, fitting could be done simultaneously at multiple wavelengths.

25) Page 3999, Line 5: What is meant with 'simplified model shapes'? Single spheroids of one axis ratio?

By simplified model shapes we refer to shapes such as spheroids. An effort has been made to clarify the text changing "Indeed, these findings suggest that, when inverting dust physical properties from the single-scattering properties, the use of simplified model shapes may lead to erroneous results even when the agreement is good... " into "Indeed, these findings suggest that, when inverting dust physical properties from the single-scattering properties, the use of simplified model shapes, such as spheroids, may lead to erroneous results even when the agreement is good".

26) Page 4003, Line 1: 'Press et al. (1992)' –> '(Press et al., 1992)'?

Answer: Fixed, thank you for helping us to improve on the manuscript.

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C2819/2011/acpd-11-C2819-2011-supplement.pdf

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 3977, 2011.