**Atmospheric
Chemistry
and Physics
Discussions**

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 #1: *"Good knowledge of the light scattering behavior of irregular dust particles is mandatory for studying their radiative impact in the atmosphere. Its theoretical study is far from trivial due to the high variety of geometries of dust particles. Moreover, they are usually distributed over broad size distributions. Therefore, new efforts in that direction are always of a great interest. In this paper, the authors present a thorough study to asses the validity of the spheroidal model for simulating the scattering behavior of irregular dust particles. Moreover, they study whether a generic shape distribution of spheroids can be applied to a broad range of dust samples candidate to be in the Earth atmosphere. To do that, calculations are compared to experimental scattering matrices of a broad range a dust samples. Apart from the detailed description of*

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Interactive Discussion

Discussion Paper

the methodology employed in this study, the authors give a quite critical presentation of their own results which is highly laudable. Therefore, I strongly recommend publication of this paper on Atmospheric Chemistry and Physics. However, there are some minor issues that I would like the authors to clarify before publication."

Answer: We thank the referee for positive and constructive comments and address the raised questions here one by one. The revised manuscript is attached as a supplement.

Specific Comments 1. Section 3, "Measurements", paragraph 145: In my opinion the way Mueller/phase/scattering matrix are defined in Sections 2 and 3 is a bit confusing. By definition, a Mueller matrix is a 44 matrix that describes each linear change of a column Stokes vector into a similar column Stokes vector (see e.g. Transfer of Polarized light in Planetary atmospheres, Hovenier, Van der Mee, Domke; 2004). As such, any of the matrices presented in the paper can be called Mueller matrices. The phase matrix relates the Stokes parameters of the incident and scattered beams, defined relative to their respective meridional planes. In contrast, the scattering matrix relates the Stokes parameters of the incident and scattered beams defined with respect to the scattering plane (See e.g. "Light scattering by nonspherical particles", Mishchenko, Hovenier, Travis, 2000; "Transfer of Polarized light in Planetary atmospheres", Hovenier, Van der Mee, Domke; 2004). According to that, F and P are both scattering matrices but with different normalizations. If the authors are following different definitions they should specify the sources."

Answer: You are absolutely right and to avoid misinterpretations, we have replaced the term Mueller matrix throughout the text by either scattering matrix or phase matrix (for normalized matrices). We also added a clarifying sentence " Both F and P are so-called Mueller matrices." in page 3984 after discussing the relation between P and F. We also found one inconsistency in nomenclature and changed the F22 into P22 In section 4.1 in the sentence "The length of the error bar covered is accounted for each matrix element when calculating coverages, so that one single outlier point with a huge error bar might lower the coverage percentage significantly, which is exactly what

happens with the feldspar F22 element in the upper right corner of Fig. 2."

2. Section 3, paragraph 150 and Section 4 paragraph 160: Do the computations cover the complete size distribution of all five samples?

Answer: The database covers size parameters from 0.012 to 625. The smallest size parameters required are about 0.8 (80 nm radius at 633 nm wavelength), so for small particles the database provides full coverage. The upper size-parameter limit, 625, however, only reaches up to 44 micrometer in radius at the shorter wavelength, 442 nm. There are particles larger than these in the samples, but their contribution to the total scattering cross section is small. The impact on the angle-dependent phase matrix elements should be negligible. We added a clarifying remark on the sentence introducing the light-scattering database (Sec 2, paragraph 105): "From the database, we can directly retrieve the scattering-matrix elements for any given aspect ratio averaged over a given size distribution within $0.012 < x < 625$, Dubovik et al. 2006. In the samples there are particles whose size parameters exceed this range. These particles are thus neglected, but their contribution to the matrix elements is estimated to be negligible." See also answer $\#$ 9 for Referee $\#3$ for information on the accuracy of the scattering measurements .

3. Section 4.1, paragraph 225: 'It is interesting to note that all the other scattering-matrix elements show strong dependence on size except for P12/P11 and P34/P11, which are reproduced quite well with spheroids regardless of the size range." It is also interesting that those two element ratios do not seem to depend that much on the refractive index used in the calculations. On the contrary the P11 element seem to be quite dependent on the refractive index in all studied cases.

Answer: This is an acute observation and presumably results from the F12 and F34 elements in having the largest coverage (see Fig. 1.) and thus being most flexible in finding and producing the minimal errors. For Figure 3, discussed in the commented text, the best possible values of n are chosen for each refractive index. For certain

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matrix elements, such as P12/P11 and P34/P11, the coverage provided by the model spheroids is good for all refractive indices, allowing us to find near-optimal fits regardless of the refractive index. For the other elements however, the span of modeled values often lies on the one or the other side of the measurements and change in the refractive index can thus go directly into change of modeling capacity. The P11 element is particularly dependent on the refractive index because the refractive index impacts both the simulations AND the measurements, the latter through extrapolation at the forward angles. We added a sentence " Moreover, these elements do not seem to depend much on the refractive index assumed. This is probably mostly due to the extensive coverage provided to these elements by the model spheroids, allowing us to obtain good fits with different refractive indices" in the manuscript.

We also noticed an inaccuracy in the original text, giving an impression that Fig. 3. would have matrix-elements plotted ("It is interesting to note that all the other scattering-matrix elements show ...", when we are instead plotting error values. We have thus replaced 'matrix elements' with 'psi values' on the paragraph in question: "It is interesting to note that all the other psi values show...".

4. Section 4.3, paragraph 285: "Curiously, unlike P11, the best-fit n for the asymmetry parameter g is slightly larger. This may be because the calculation of the asymmetry parameter takes into account also the extrapolated diffraction peak". In principle the forward diffraction peak is not that sensitive to the shape of the particles but on the size distribution. Indeed, that is the assumption used for the extrapolation of the phase function at small scattering angles (Liu et al. 2003; Kahnert and Nousiainen 2006; Kahnert and Nousiainen 2007). If for getting a good-fit for g we need a higher value of n it seems like the diffraction forward peak could be shape-sensitive. Can you comment on that or could you think of any other reasons for the requested values of n for the best-fit of g?

Answer: The P11 and g are different (if related) quantities. It is quite reasonable that they lead to slightly different n, because when fitting P11, all angles are

treated with equal weights, whereas when fitting g, the angles have been weighted by sin(theta)*cos(theta) term. So, differences at any angle will weight differently when fitting g or P11. It does not have to be in the forward diffraction. To avoid misleading the reader, and to fix our original, possibly incorrect speculation, we decided to remove the speculation on the cause of the n-value differences for P11 and g.

5. Section 4.3, paragraph 290: Although the HSA (Homogeneous Sphere Approximation) is defined on page 2, it would facilitate the reading if the definition is included again on page 9. There are a lot of information and definitions of variables in the previous pages and it is difficult to retain all of them specially when it was given 7 pages earlier.

Answer: Fixed as suggested.

6. Section 4.4, paragraph 360, The authors mention the results for loess but they are not shown in Figure 5. That should be mentioned in the text.

Answer: Fixed as suggested.

7. Section 5, paragraph 435, "When the asymmetry parameter is the criterion, a reasonable first assumption for a spheroid shape distribution is to use the power law function with n=3. For P11 element, the equiprobable distribution often works the best, whilst for the polarisation elements it might prove profitable to favour heavily the most extreme shapes (n=18)." According to Figure 6, the asymmetry parameter as well as the F11, and F12 elements (except in the Sahara case) are reasonably well reproduced with the power law distribution with n=3 and n=10 with very small differences between each other. I do not see that in any of the studied cases, the equiprobable distribution is the best option for the F11 element. Moreover, you write "Pij" in the text but Fij in the figure. Please, correct that. I assume that in all plots when you say P11 you mean that they are normalized according to equation (2). If that is not the case you should mention which normalization is used and they should not be called P11.

Answer: If the minimum value of error is sought among all model runs with various refractive indices, the n=0 rises often as the best option for F11 (this can be seen in first column of Table 3.). In Figure 6, only one refractive index is used for each particle type, that deemed to be most suitable overall. So the mention of the equiprobable distribution working for F11 element is misplaced and thus misleading, and has now been removed. And yes, in the figure there should be P instead of F, which is now fixed.

8. Section 5, paragraph 440: What do you mean with a "generic size distribution"? Do you mean a generic shape distribution?

Answer: Yes, thank you for spotting this, it is now corrected!

9. Section 6, paragraph 495 "It turns out that it is impossible to suggest a single shape distribution that would be the best choice in all cases. Not only does the best-fit distribution vary between the samples, but it also varies between the wavelengths, the metrics used for specifying the goodness of fit, the quantities fitted, and the refractive index assumed." In my opinion this is a too strong/negative statement. It is clear that the P22 element is not well reproduced by any of the studied shape distributions. Thus, the spheroidal model does not seem to be a good model for studding depolarization properties of dust particles. However, it seems like the shape distributions with n=3 and n=10 are in general a good approximation for all studied samples specially if we focus on the asymmetry parameter, and the F11 and F12/F11 elements (see point 7 of this review). The fits can be further improved by using n=18. Moreover, it is clear that even when the fits are not perfect, a shape distribution of spheroids (for samples with relatively small reff) is a better approximation by far (including the F22/F11 ratio!) than the Homogeneous Spherical Approximation.Therefore, I recommend to reformulate the paragraph. Apart from that, in this paper the chosen set of samples correspond to a quite broad scenario attending to their origin. However, it seems like the scattering behavior of different dust samples is confined to rather limited domains when they are grouped according to their origin (see e.g. Munoz et al. JGR, 109,

D16201, doi:10.1029/2004JD004684). That seems to be in agreement with the results presented in this paper for red and green clay. I would recommend as a future work to perform a similar study on several different groups of samples like e.g. clays, volcanic ashes, or desert dust. That would probably facilitate the definition of a single shape distribution representative for each of the groups. That can provide a powerful diagnostic tool for irregular aerosols in the atmosphere.

Answer: Thank you for your insight, it would indeed be interesting and probably beneficial to study groups of different dust types.

We rephrased the start of the commented paragraph to remind the reader of the overall encouraging results of this study: "Although relatively good results can be acquired by various shape distributions, it turns out..."

Please also note the supplement to this comment:
http://www.atmos-chem-phys-discuss.net/11/C2812/2011/acpd-11-C2812-2011-supplement.pdf

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

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