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Interactive comment on "Volcanic ash infrared signature: realistic ash particle shapes compared to spherical ash particles" by A. Kylling et al.

Anonymous Referee #1

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This article by Kylling et al. is about the comparison of infrared optical properties of non-spherical volcanic ash particles with mass and volume equivalent spheres and their impact on volcanic ash mass retrievals. The scientific question is well posed and has not been investigated in literature so far. Therefore I recommend this paper for publication after addressing the comments below.

General Comments:

In Section 2.2 to 6 various terms ("ash particles and spheroids, spherical particles, spherical models") are used for the non-spherical ash particles and the spherical ash particles. This is somewhat confusing. Since in any discussion no difference is made between the highly irregular ash particles and the spheroidal ash particles, I

suggest to call them "non-spherical" ash particles. I'd also recommend to rename "spherical models, sphere models" to "equivalent spherical particles" or "equivalent spheres" throughout the paper.

Particle size is a very crucial aspect in this study, because scattering on aerosol in the mid-infrared is strongly size dependent. Unfortunately it is not clear what is meant by size (maximum dimension, or mass or volume equivalent diameter, or equivalent radius?). I highly recommend to make this clear once, e.g in Section 2.2, and then consequently use it throughout the paper (also in the figures).

The descriptions/discussions of the figures are imprecise (e.g. "The performance of the two effective medium theories varies. For some ash particle sizes Bruggeman is similar to the ash particle results, for some sizes it is rather different. The same is true for the Maxwell-Garnett spheres.") or even opposite to what the figures show. This should be improved. (see specific comments)

Specific Comments:

p.8937 I.23: I would recommend to use chronological order for references p.8937 I.25: Does Mishenko,2009, address the infrared? Please indicate which of the 150 references given there are relevant for the infrared.

p.8939 I.3-20: Since this study is about volcanic ash optical properties in the infrared, I'd recommend to motivate this study (also) in the infrared instead of only in the VIS. For example, for mineral dust, which has optical properties similar to volcanic ash in the infrared,

Hudson, P. K., Gibson, E. R., Young, M. A., Kleiber, P. D., and Grassian, V. H.: Coupled infrared extinction and size distribution measurements for several clay components of mineral dust aerosol, J. Geophys. Res., 113, doi:10.1029/2007JD008791, 2008

Hudson, P. K., Young, M. A., Kleiber, P. D., and Grassian, V. H.: Coupled infrared C3550

extinction spectra and size distribution measurements for several non-clay components of mineral dust aerosol (quartz, calcite, and dolomite), Atmos. Environment, 42, 5991–5999, 2008

showed that there are significant changes in extinction coefficient spectra for non-spherical particles. For polar stratospheric cloud particles composed of nitric acid dihydrate,

Robert Wagner, Ottmar Möhler, Harald Saathoff, Olaf Stetzer, and Ulrich Schurat: Infrared Spectrum of Nitric Acid Dihydrate: Influence of Particle Shape. J. Phys. Chem., 2005

showed also a sensitivity on particle shape in the infrared. Klüser (2011, AE) used Hudson (2008) extinction coefficient spectra for IASI mineral dust type retrieval and hence accounted for particle non-sphericity effects of mineral dust particles in the infrared.

p.8939 l.11-13: "For a refractive index with a larger imaginary part (larger absorption), the electromagnetic field will not penetrate that far into the particle." Could you please indicate which implications this has?

p.8940 I. 7: Please see general comment on terminology; insert "non-spherical" before ash particles

p.8942 I.17-19: Could you please shortly outline the basic principle of the DDA method? Without a short explanation the dipoles in I.26/27 are a bit surprising.

p.8942 l.20: What is meant by size? Is it the maximum dimension, or an mass or volume equivalent diameter, or equivalent radius? See general comment.

p.8942 I.21-23: Which refractive indices did you use?

p.8942 I.24: I suggest to give the refractive index information in the paragraph above. Why do you use Andesite from Pollack (1973) and not pumice from Volz (1973)? For porous volcanic ash I would think that pumice is the more representative material.

p.8942 I.26-27: If you explain the DDA in the paragraph above a bit more detailed it

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would be easier to understand that the dipoles actually represent the solid parts of the porous ash particles. The ash particle is composed of solid and vacuum dipoles, but only the solid dipoles contribute to the weight, right?

p.8943 l.8: To me it seems that you just skipped the simpler approximation of volume equivalent spheres with refractive index of the solid material. Could you please also include this scenario? I think this is an important point to also quantify the possible mass retrieval error when using volume equivalent spheres without a modified effective refractive index. Please explain why doTherefore I recommend this paper for publication after addressing the comments below. you compute an effective refractive index?

p.8943 I.22: I suggest to split this sentence and to use the terms "non-spherical particles" and "equivalent spherical particles".

p.8944 l.1: please use non-spherical ash particles instead of ash particles and spheroids

p.8944 l.8: it is not clear what is meant by particle size

p.8944 I.10-12: The discussion of figures 2 and 3 is very imprecise. Isn't it that Bruggeman and MG are relatively similar to each other and both are closer to the non-spherical particles Q_{ext} and Q_{sca} than the mass equivalent spheres? Figure 2 and 3 also show that B and MG are larger for particle sizes > 4μ m for large vesicles and smaller for particle sizes > 5μ m for small vesicles. Could you discuss the figures more specifically?

p.8944 I.12-14: you say "Thus, optical properties calculated for mass- or volume-equivalent homogeneous spheres do not generally agree with optical properties of morphologically complex inhomogeneous ash particles." but your figures actually imply that for particle sizes (whatever is meant by particle size) up to $4\mu m$ the volume equivalent spheres provide a quite acceptable approximation.

p.8944 I.15: Why is this surprising?

p.8944 I.15-19: Is the phase function important or not? This part is confusing.

p.8944 I.20-26: This paragraph is difficult to understand.

p.8944 I.20: Can you explain why this ratio is important? Can one expect negative BTDs as long as the ratio is positive?

p.8944 I.23: Why don't you discuss the good agreement between Bruggeman equivalent spheres and the non-spherical particles?

p.8944 I.24: I suggest to rephrase "It is thus anticipated that, compared to the mass-equivalent spheres, the other particles will give a negative brightness temperature difference signal dBT for a larger particle size range." to "Thus we expect that the non-spherical particles and the volume equivalent spheres will result in negative BTDS for larger particle sizes than the mass equivalent spheres." to be more precise. With "other particles" you mean non-spherical particles and volume equivalent spheres, right?

p.8945 l.1 & 15: I highly recommend to reorganise the structure of Section 3 and 4. Simulation setup and results should be in a single section. Optionally the authors could use subsections for setup and results.

p.8945 l.16-17: Leave out this sentence. It is redundant with l.12-13.

p.8945 l.17-18: Mono-dispersed: see comment above about reorganising the structure. This is part of the simulation setup description.

p.8945 l.20: "dashed lines"? There are only dotted lines. See comment above about particle size.

p.8946 l.6: Please think about restructuring. i suggest to discuss figure 5 first and then to go on with figure 6.

p.8946 l.7: See comment about particle size and replace sphere models with equivalent spheres.

p.8946 I.7-9: I don't understand this sentence. Please improve description and discussion of figure 6.

p.8945 I.15-17: Figure 6 and your description of figure 6 do not match. Your figure shows that the volume equivalent spheres for small vesicles fit the non-spherical ash particles very well. The mass equivalent spheres and the volume equivalent spheres

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for large vesicles do not fit well.

p.8945 l.18: Could you describe how figure 6 looks for polydisperse particles?

p.8947 l.5: Actually I would expect that the volume equivalent spheres are a better approximation than the mass equivalent spheres. Why don't you discuss the volume equivalent spheres?

p.8947 l.9: What is $\tau(\lambda)$?

p.8947 I.18: And what are the differences for volume equivalent spheres?

p.8947 I.20: Why is the effect of particle shape quantified by comparing non-spherical particles and volume equivalent spheres?

p.8947 l.21: I guess you mean "BT11 and dBT above," instead of "BT11 and BT12 above". Can you also give the exact values?

p.8948 l.6: Why is the total error only increased by 5-15% if the mass error is about 12-40%?

p.8948 point I.18: Can you also state how the optical properties of non-spherical ash particles compare with volume equivalent spheres?

p.8954 Figure2: What is meant by particle size? Check for terminology of non-spherical and equivalent spherical particles. Could you also add the scenario of volume equivalent sphere with andesite refractive index?

p.8956 Figure4: There are many indistinguishable red lines in this figure. I suggest to show one red line with bars indicating minimum and maximum as in figures 2 and 3. In the text the different red scenarios are not discussed separately, so there is no need to show all of them.

p.8958 Figure6: Same as for figure 4. There are many indistinguishable red lines in this figure. I suggest to show one red line with bars indicating minimum and maximum as in figures 2 and 3. In the text the different red scenarios are not

discussed separately, so there is no need to show all of them. Also now the particle size is a radius. How did you estimate the radius for the non-spherical particles?

Technical Corrections:

Since I'm not a native speaker I'd suggest to use Copernicus language service for this manuscript.

Abstract and Section 2.2: Please use past tense when describing what you have done.

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p.8937 I.17: there is a comma missing before "which"
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p.8941 I.6: I think it should be "after" instead of "once"

p.8942 l.2: "as large as" instead if "than"

p.8942 l.4: "except" instead of "save"

p.8943 I.17 and p.8949 I.23 "Bruggeman" instead of "Bruggemann" (one "n" too much)

p.8954: "the red line represents" instead of "the red lines represents" (one "s" too much)

p.8944 l.4: "equivalent spheres" instead of "sphere models"

p.8944 l.8: "scattering" instead of "sacettering"

p.8944 I.23: "volume equivalent spheres" instead of "other sphere models"

p.8945 l.7: comma before "which"

p.8945 l.11: comma before "which"

p.8947 I.2: delete "brightness temperature difference", dBT is sufficient

p.8947 l.11: "usually" instead of "normally"

p.8948 I.20: "is" instead of "are"

Interactive comment on Atmos. Meas. Tech. Discuss., 6, 8937, 2013.

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