

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

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Response to interactive comments from Referee #1

We thank the referee for the careful reading of and constructive comments to our manuscript, quoted below in italic font. Our responses to the comments are shown in roman font.

C4189

General Comments

1. *In Section 2.2 to 6 various terms (“ash particles and spheroids, spherical particles, spherical models”) are used for the non-spherical ash particles spherical and the 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.*

The nomenclature for the various particle shapes have been clarified throughout the paper.

2. *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 term “particle size” has been removed from the paper and replaced by the “mass-equivalent particle radius”.

3. *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).*

The discussion of the Figures have been extended and improved. Please see responses to specific comments below.

C4190

Specific comments:

- p.8937 I.23: *I would recommend to use chronological order for references.*

Chronological order of references adopted as suggested.

- p.8937 I.25: *Does Mishchenko,2009, address the infrared? Please indicate which of the 150 references given there are relevant for the infrared.*

There may be a slight misunderstanding of what we intended to express with this sentence. The statement is very general and not limited to a specific wavelength range or particle chemical composition and morphology. Note that, according to the scale invariance rule of electromagnetic scattering, the scattering characteristics of particles are primarily dependent on a dimensionless size parameter (e.g. Mishchenko, 2006), which is often defined by $x = 2\pi r/\lambda$, where r is the characteristic particle radius and λ is the wavelength of light. In particular, this means that differences in optical properties between spherical and nonspherical particles observed at, say, a visible wavelength λ_1 for a particle size r_1 will also be observed at, say, an infrared wavelength λ_2 for a particle size $r_2 = r_1\lambda_2/\lambda_1$, provided that the refractive index of the particles is the same. The article by Mishchenko (2009) was chosen as a reference here precisely because it is not limited to a certain wavelength range, but provides a rather general and pedagogical introduction into the field of scattering by nonspherical particles.

To avoid any misunderstandings, we suggest to re-formulate the sentence as follows:

"It is well established that, over a large range of sizes, wavelengths, and dielectric properties, the optical properties of non-spherical particles can be significantly different from those of spherical particles (For a readable and general introduction into the vast field of scattering by non-spherical particles we refer the reader to the recent review by Mishchenko, 2009)."

C4191

- 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 nonspherical particles. For polar stratospheric cloud particles composed of nitric acid dihydrate, Robert Wagner, Ottmar Möhler, Harald Saathoff, Olaf Stetzer, and Ulrich Schurath: 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.*

The suggested references and corresponding description have been incorporated into the text.

- p.8939 I.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?*

It implies that the scattering properties will be less determined by internal resonances induced inside the particle, but more by surface waves. Thus, morphological features, such as surface roughness, may become more important than other features, such as overall nonsphericity and internal structure. Since we merely want to provide a short literature overview in the introduction without di-

C4192

verting into the details of the electromagnetic scattering process, we suggest to reformulate this and the following sentence (lines 11-16) as follows:

"If the material of the particles is optically hard (large real part) or strongly absorbing (large imaginary part), the roughness of the particle, not considered by Krotkov et al. (1999), may play an important role for the optical properties (e.g. Kahnert et al., 2011; Kahnert et al., 2012)."

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

"non-spherical" inserted before "ash particles" as suggested.

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

We suggest to add the following text after the first sentence of Sect. 2.2 (line 19):

"The discrete dipole method is a volume-integral equation method, in which the volume integral is numerically evaluated by discretising the particle volume into (usually cubical) volume cells of size d . The main assumption is that d is sufficiently small, so that the electric field can be assumed to be constant over each cell volume. Thus, the oscillators within each volume cell respond to the external field by oscillating in phase, just like a dipole, and phase differences may arise only between cells. This means that in the DDA the target is, essentially, replaced by an array of dipoles with a dipole spacing d ."

- *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.*

By size is meant the mass equivalent radius. In the text we have replaced "sizes" by "mass equivalent radii".

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

C4193

The andesite refractive index from Pollack et al. (1973) was used.

- *p.8942 l.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.*

We have inserted the following sentence "The refractive index for andesite from Pollack et al. (1973) was used." We choose to use the andesite from Pollack et al. (1973) as it is commonly used for ash retrievals. Several authors have compared volcanic ash retrievals using andesite refractive indices from Pollack et al. (1973) and pumice from Volz (1973), see for example Corradini et al. (2008) and Francis et al. (2012). Generally the andesite refractive indices yield slightly larger ash mass loadings. Due to the computational burden of the single scattering simulations a comparison of the andesite and pumice refractive indices is outside the scope of the present paper.

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

The DDA has been described in greater detail as outlined above. Only the solid dipoles contribute to the weight. This has been clarified in the text by changing the last sentence in page 8942 to "The dipoles within the gas pockets, vesicles, were excluded."

- *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*

C4194

a modified effective refractive index. Please explain why do you compute an effective refractive index?

We assume that with “volume-equivalent spheres with the refractive index of the solid material” the referee means spheres which are not porous and with a volume-equivalent radius calculated from the non-spherical shapes. Comparing such spheres with the non-spherical particles implies comparing particles with different mass, an approach which we do not find appropriate. Similarly, the calculation of an effective refractive index is needed for volume-equivalent spheres.

- *p.8943 l.22: I suggest to split this sentence and to use the terms "non-spherical particles" and "equivalent spherical particles". The sentence has been split as suggested and the terms "non-spherical particles" and "equivalent spherical particles" have been adopted.*
- *p.8944 l.1: please use non-spherical ash particles instead of ash particles and spheroids*
The term non-spherical ash particles have been adopted as a common name for ash particles and spheroids.
- *p.8944 l.8: it is not clear what is meant by particle size*
“particle sizes” has been replaced by “mass-equivalent particle radii”.
- *p.8944 l.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 $> 4\mu m$ for large vesicles and smaller for particle sizes $> 5\mu m$ for small vesicles. Could you discuss the figures more specifically?*

C4195

To improve the discussion of Figs. 2 and 3 we suggest to move the discussion of the insensitivity of the asymmetry factor to a separate paragraph and discuss the Figures as follows:

“ For mass-equivalent particle radii below $3\ \mu m$ ($4\ \mu m$) the optical properties of mass and volume-equivalent spheres behave similarly to those of non-spherical particles for a wavelength of $10.8\ \mu m$ ($12.0\ \mu m$). For larger mass-equivalent particle radii, the optical properties calculated for mass- or volume-equivalent spheres do not generally agree with the optical properties of morphologically complex inhomogeneous ash particles. The mass-equivalent compact spheres (black lines Figs. 2-3) consistently underestimate the extinction and scattering efficiencies for mass-equivalent particle radii larger than 3 and 4-5 μm for wavelengths of 10.8 and 12 μm respectively, compared with both large and small vesicle non-spherical particles. The single scattering albedo is underestimated for 12.0 μm but shows agreement for 10.8 μm . The mass-equivalent spheres generally underestimate the asymmetry factor. The performance of the two effective medium theories varies for mass-equivalent particle radii above 3-6 μm compared to the non-spherical particles. Both the volume-equivalent Maxwell Garnett and Bruggeman spheres overestimate the optical properties compared to large vesicle non-spherical particles (Fig. 2). Compared to small vesicle particles (Fig. 3), the volume-equivalent Maxwell Garnett and Bruggeman spheres somewhat underestimate the extinction and scattering efficiencies and show good agreement for the single scattering albedo and the asymmetry factor. “

- *p.8944 l.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.*

Please see our answer to comment above.

C4196

- *p.8944 l.15: Why is this surprising?*

The wording here was unfortunate and we suggest to replace:

“The asymmetry parameter is quite insensitive to the shape assumptions, which is surprising.”

with the sentences:

“The asymmetry parameters are similar for each non-spherical ash model considered.”

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

We have clarified this by changing l.15-19 to

“ For thermal radiation the source term in the radiative transfer equation does not include the phase function. However, the phase function is of importance when scattering takes place, and, as shown in the third row of Figs. 2 and 3, the single scattering albedo is sufficiently large to make scattering effects have an impact for particles larger than about 2 μm . “

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

To clarify this paragraph we suggest to replace the first sentence with:

“For a semi-transparent plane-parallel ash cloud for which scattering is assumed to be negligible, ΔT may be written Prata and Grant (2001):

$$\Delta T = \Delta T_c (X - X^\beta). \quad (1)$$

where $\Delta T_c = T_s - T_c$, $X = 1 - \Delta_{10.8}/\Delta T_c$, $\Delta_{10.8} = T_s - T_{10.8}$, and T_s and T_c are the Earth’s surface and ash cloud temperatures, respectively. The ratio of the extinction coefficients is denoted by $\beta = k_{12}/k_{10.8}$. Normally we have $T_s > T_c$, and a value of $\beta > 1$ will thus give negative ΔT .”

C4197

- *p.8944 l.20: Can you explain why this ratio is important? Can one expect negative BTDS as long as the ratio is positive?*

Please see our response to above comment.

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

We suggest to expand the discussion with the following text:

“ It is noted that the β -ratio for Bruggeman volume-equivalent spheres exhibit the same behaviour as for the non-spherical particles. There is also agreement between the non-spherical particles and the Maxwell Garnett volume-equivalent small vesicle spheres for mass-equivalent radii larger than 3 μm . The β -ratio for Maxwell Garnett volume-equivalent large-vesicle spheres is overestimated for mass-equivalent radii between 3-7 μm and underestimated elsewhere when compared to the non-spherical particles. “

- *p.8944 l.24: I suggest to rephrase "It is thus anticipated that, compared to the massequivalent spheres, the other particles will give a negative brightness temperature difference signal dBTD 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?*

Change made as suggested.

- *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.*

We have reorganised sections 3 and 4 into one section as suggested.

C4198

- *p.8945 l.16-17: Leave out this sentence. It is redundant with l.12-13.*
The sentence has been left out as suggested.
- *p.8945 l.17-18: Mono-dispersed: see comment above about reorganising the structure. This is part of the simulation setup description.*
The section has been reorganised as suggested.
- *p.8945 l.20: "dashed lines"? There are only dotted lines. See comment above about particle size.*
"Dashed lines" have been corrected to "dotted lines". "Particle size" have been changed to "mass-equivalent particle radius" both in the text and Fig. 5 caption.
- *p.8946 l.6: Please think about restructuring. i suggest to discuss figure 5 first and then to go on with figure 6.*
We have reorganised the text and Fig. 5 is now fully discussed before Fig. 6.
- *p.8946 l.7: See comment about particle size and replace sphere models with equivalent spheres.*
"Particle size" has been changed to "mass-equivalent particle radius". "sphere models" has been replaced by "equivalent spheres".
- *p.8946 l.7-9: I don't understand this sentence. Please improve description and discussion of figure 6.*
To clarify the sentence we suggest to write is as:
"The equivalent spheres based on the Maxwell Garnett mixing rule give a slightly more negative ΔT compared to the Bruggeman spheres, as indicated in Fig. 6."
- *p.8946 l.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*

C4199

ash particles very well. The mass equivalent spheres and the volume equivalent spheres for large vesicles do not fit well.

To improve the discussion of Fig. 6, we suggest to replace l.15-17 with:

"The volume-equivalent spheres with small vesicles (dotted lines, Fig. 6) behave similarly to the non-spherical particles (red line, Fig. 6). This is consistent with the optical properties shown in Figs. 2 and 3. Volume-equivalent spheres with large vesicles (solid blue and green lines, Fig. 6) give larger negative ΔT for mass-equivalent radii larger than about $5 \mu\text{m}$ compared to the non-spherical particles. The mass-equivalent spheres give less negative ΔT for mass-equivalent radii larger than about $3 \mu\text{m}$ compared to the non-spherical particles. The volume-equivalent spheres with large vesicles do not give positive ΔT for any mass-equivalent radii, whereas volume-equivalent spheres with small vesicles give positive ΔT for radii larger than about $7 \mu\text{m}$ ($9 \mu\text{m}$) for the Bruggeman (Maxwell Garnett) mixing rule."

- *p.8946 l.18: Could you describe how figure 6 looks for polydisperse particles?*
To avoid any confusion and to keep the focus of the paper on particle shape, we suggest to omit the discussion on particle size distributions in the revised manuscript.
- *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?*
Please see our answer to comment p.8943 l.8 above.
- *p.8947 l.9: What is $\tau(\lambda)$?*
The following explanation have been added to sentence in l.10: " τ is the optical depth at wavelength λ ".

C4200

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

Please see our answer to comment p.8943 I.8 above.

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

This section has been completely rewritten with a new example. Please see answer to comment 4 by Referee #3.

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

This section has been completely rewritten with a new example. Please see our answer to comment 4 by Referee #3.

- *p.8948 I.6: Why is the total error only increased by 5-1512-40%?*

We assume independent errors and use standard error propagation theory which gives the total error as the square root of the square error sum. This has been better explained in the revised manuscript.

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

Please see our answer to comment p.8943 I.8 above.

- *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?*

Particle size has been replaced by mass-equivalent sphere radius. Concerning volume equivalent spheres, please see our answer to comment p.8943 I.8 above.

C4201

- *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.*

Figure 4 has been simplified and the results for non-spherical particles are now presented as in Figures 2 and 3. The caption have been changed to reflect this.

- *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?*

Figure 6 has been simplified similarly to Figure 4. The caption has been changed accordingly. Radius have been corrected to mass-equivalent radius.

Technical Corrections:

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

Past tense have been adopted in the Abstract and Section 2.2.

- *p.8938 I.17: there is a comma missing before "which"*

Comma inserted before which.

- *p.8941 I.6: I think it should be "after" instead of "once"*

"Once" changed to "After".

C4202

- *p.8942 l.2: "as large as" instead of "than"*
"than" changed to "as".
- *p.8942 l.4: "except" instead of "save"* "save" changed to "except".
- *p.8943 l.17 and p.8949 l.23: "Bruggeman" instead of "Bruggemann" (one "n" too much)*
The name of Bruggeman has been corrected.
- *p.8954: "the red line represents" instead of "the red lines represents" (one "s" too much)*
Changed "red lines represents" to "red lines represent".
- *p.8944 l.4: "equivalent spheres" instead of "sphere models"*
"sphere models" replaced by "equivalent spheres" as suggested.
- *p.8944 l.8: "scattering" instead of "sacettering"*
Spelling corrected.
- *p.8944 l.23: "volume equivalent spheres" instead of "other sphere models"*
"other sphere models" changed to "volume-equivalent spheres".
- *p.8945 l.7: comma before "which"*
Comma inserted before which.
- *p.8945 l.11: comma before "which"*
Comma inserted before which.
- *p.8947 l.2: delete "brightness temperature difference", dBT is sufficient*
"brightness temperature difference" has been deleted as suggested.

C4203

- *p.8947 l.11: "usually" instead of "normally"*
"Normally" replaced by "Usually" as suggested.
- *p.8948 l.20: "is" instead of "are"*
"are" corrected to "is".

C4204