Response letter to the Reviewers Helsinki, 24.10.2013

We wish to thank the reviewers for their careful reading of our manuscript published in ACPD and their comments to it. The manuscript has been modified accordingly and we hope that the reviewers will find the changes satisfactory. The comments of the reviewers are shown below in italics and answered separately.

Sincerely, in behalf of all co-authors,

Hannakaisa Lindqvist

Reviewer #1:

Synopsis: scanning-electron microscope images were used to construct the 3D geometries of dust particles whose compositions were inferred from energy-dispersive spectroscopy measurements. The obtained dust morphologies and compositions were applied to light scattering simulations performed with the discrete dipole approximation.

Overall, the manuscript is well written. No major technical errors were found. The study can be a useful contribution to the literature of the scattering of light by small particles. The manuscript can be accepted for publication after some minor revisions.

We thank the reviewer for considering the manuscript worth publishing, and next address his/her specific comments in detail:

Specific comments:

1) *The major weakness of this study is the upper limit of the size parameter range is only 16, a value that is small in the case of dust particles at a visible wavelength.*

The upper limit of the size parameter range is due to computational limitations. To emphasize this, we have added information to Sect. 4.2 on the computational times for largest particle sizes: ”With the chosen resolution, the running times of the parallelized computations varied from hours to a few days; the most demanding case was Sil I, which required 154 hours on 46 processors, resulting in the total of 7084 hours of CPU time.”

2) *It is shown that the single-scattering properties, averaged over a size distribution, are not sensitive to the detailed size distribution features for given effective variance and effective particle size. If the cutoffs of the size parameters are 0.5 and 16 at the lower and higher ends, respectively, the single-scattering properties may be sensitive to the specific size distribution used.*

We agree that our computations do not cover the whole size distribution (SD) and this truncation impacts the obtained SD-integrated values. In other words, the computations should not be interpreted to represent the single-scattering properties of dust particles with the given SD. Rather, the SD is simply used to provide reasonable weights for the SD integration in such a way that all computed size parameters contribute. At different wavelength our computations would cover different parts of

the SD. To clarify this, we added this sentence: ”Our chosen size parameter range naturally covers only part of this example distribution.”

3) *Typo in the first paragraph of Section 3: “elecron microscopy” should be “electron microscopy”.*

Fixed.

4) *It suggested that the variation of the extinction efficiency versus the size parameter be presented for the various cases considered in this study.*

ADDA calculates Q_{ext} by dividing the corresponding cross section with the area of the geometrical cross section of the sphere with a volume equal to that of dipole representation of the particle. This does not correspond to the definition of Q_{ext} which should involve the actual cross sectional areas in each orientation used. As the computation of the real Q_{ext} would be overly complicated, and we feel plotting the Q_{ext} values reported by ADDA would be misleading, we have decided to leave out such plots.

Reviewer #2:

This manuscript reports the use of stereogrammetric shape retrieval method to derive dust three dimensional particle shapes and to determine inhomogeneous composition of dust particles by mineralogical interpretation of localized elemental information based on energy-dispersive spectroscopy. The optical properties of these particles are then modeled. The manuscript is well written. This reviewer recommends this manuscript be published after revisions as follows

We thank the reviewer for considering our work suitable for publication and for suggesting improvements to it. These are addressed next in detail.

1. *Different particle shapes and compositions definitely cause different light scattering properties. This has already been well known for many years. A good research is not satisfied by finding only the differences, but by finding more general things, i.e. The general properties of these individual cases. This manuscript should derive more on the latter.*

We definitely agree with the reviewer that light scattering research for mineral dust particles should strive for this goal altogether: to find the general properties in scattering by different types of dust particles. However, in this study, we considered an ensemble of only four single dust particles which we consider to be much too few for generalizations. We also note that these four particles had substantially different single-scattering properties. In the future, we intend to use the stereogrammetric method to a larger set of particles hoping that some general features could be identified.

2. *The word “invaluable” in the statement “Our results could be invaluable as references in validation of such a method.” Is not appropriate.*

This was a bit of a mistake from our part. What we really wanted to say is that we believe our method to be a valuable tool for validating approaches based on simpler, faster methods. Data provided by our method could be used as reference for such validation. We have rephrased the

text accordingly: ”For validation of such a method, the approach presented here could be used for producing reference data when applied to a suitable set of target particles.”

3. *Texts for surface and shape retrievals by stereogrammetry are not well written. The methods must be clearly described step by step.*

We went through the text sentence by sentence and could not recognize any particularly unclear statements. As the stereogrammetric method is a complicated process with several phases, we decided to improve the clarity of the description by adding a schematic presentation of the method as a figure to Sect. 4.1. We believe that the details of the process are well and clearly explained and described with equations and appropriate citations in the text.

4. *“The scattering properties of different, individual dust particles are highly dependent on their physical properties, and this essential link : : :” What is “this essential link”?*

In this context, ”this essential link” refers to the link between physical and optical properties. We modified this sentence in the following way: ”The scattering properties of different, individual dust particles are highly dependent on their physical properties, and this relation can be established either by measuring the single-scattering properties for particles whose physical properties are known, or computing them with a single-scattering model.”

5. *“Both of these are essential since the single-scattering properties of dust have been estimated to be very sensitive to the scale and type of surface roughness (Nousiainen, 2009, and references therein) and inhomogeneity” A much earlier paper of Wenbo Sun, Norman G. Loeb, Gorden Videen, and Qiang Fu, "Examination of surface roughness on light scattering by long ice columns by use of a two-dimensional finite-difference time domain algorithm", Appl. Opt., 43, 1957-1964 (2004) must be cited here for sufficient bibliography.*

This publication was not originally cited because it considers ice crystals, not mineral dust particles. However, we agree with the reviewer that surface roughness is an important topic in scattering and therefore added the reference as follows: ”More generally, surface roughness has been considered as a significant feature of small atmospheric particles with respect to its impact on scattering (see, e.g., Macke et al., 1996; Nousiainen et al., 2003; Sun et al., 2004).”

6. *Section “2 Relevant single-scattering theory” is unnecessary. This section should be removed. Eqs (4) and (5) can be showed in the section for numerical results simply as some notes.*

We would like to have this section in the paper because it provides essential information for understanding the results section, as well as emphasizes that this is first and foremost a light-scattering study, not a shape analysis of mineral dust particles.

7. *When talking about “The Gaussian random sphere : : :” An accurate numerical calculation of light scattering properties of Gaussian-type particles was done in Wenbo Sun, Timo Nousiainen, Karri Muinonen, Qiang Fu, Norman G. Loeb, and Gorden Videen, "Light scattering by Gaussian particles: A solution with finite-difference time domain technique", J. Quant. Spectrosc. Radiat. Transfer, 79-80, 1083-1090 (2003). This paper*

needs to be cited properly.

The Gaussian random sphere model has been used in a large number of studies. As this is not a review, nor particularly focused on the Gaussian random sphere geometry, we did not consider it necessary to cite all these works, and in particular not those considering ice particles instead of dust particles. However, since the reviewer considers this a necessary addition, we have added the requested reference in the following sentence: "The Gaussian random sphere is a statistical shape model for irregular particles introduced by Muinonen et al. (1996) and used in, e.g., scattering modelling of ice crystals (Sun et al., 2003)."

8. *"From the orientation-averaged computations, we obtained the 10 scattering-matrix elements of : : :" Why 10 elements? not 8? not the 6 independent nonzero elements for randomly oriented particles? Do these dust aerosols have orientation preference in the air?*

The particles are considered as randomly oriented. Note, however, that these are single particles, not particle ensembles. Only in the case where a particle ensemble consists of particles and their mirror particles in equal numbers, does the scattering matrix result in 8 nonzero elements out of which 6 are different.

9. *It is also interesting to see the depolarization properties of these dust particles. The authors may consider showing the curves for depolarization ratio as in Wenbo Sun, Zhaoyan Liu, Gorden Videen, Qiang Fu, Karri Muinonen, David M. Winker, Constantine Lukashin, Zhonghai Jin, Bing Lin, and Jianping Huang, "For the depolarization of linearly polarized light by smoke particles," J. Quant. Spectrosc. Radiat. Transfer, 122, 233-237 (2013).*

The depolarization ratio for size-integrated scattering is already presented in Fig. 10. Moreover, Fig. 7 presents the S22/S11 ratio, which is connected to the depolarization ratio, as a function of scattering angle and size parameter. We consider these to be sufficient for this paper, although it would be an interesting idea to concentrate more on dust depolarization and study the behavior of the phenomena seen there (see, e.g., Lindqvist et al., 2009).

Reviewer #3 (Maxim Yurkin):

The manuscript provides an important step towards realistic simulations of light scattering by atmospheric aerosols. It not only describes and successfully applies a new methodology, but also compares it with existing simpler approaches. The manuscript is well-written and is definitely worth publishing, but a few issues must be addressed before publication.

We thank Dr. Yurkin for this consideration, and for his suggestions that definitely helped us improve the quality of the manuscript.

1) *The authors state that their results can be used as a reference/benchmarks, and they deduct certain conclusions from comparison of their realistic simulations to simpler models. Thus, they should discuss the parameters and accuracy of the DDA simulations in much more details.*

This is an excellent remark. By putting the parameters and accuracy considerations in the text, the computations become more transparent and could be repeated. The additions are indicated below, in the context of more detailed comments. We also reconsidered the statements about reference/benchmark computations, and decided to rephrase them because the number of particles in our study is fairly small to be considered as references. In the Abstract, we now write: ”For validation of such a method, the approach presented here could be used for producing reference data when applied to a suitable set of target particles.” and the sentence in the Conclusion as ”If applied to larger particle sets, the method presented here could be used to provide reference data for validating simpler shape models for mineral dust.”

> *1a) The authors should describe the parameters of the DDA simulations in Section 4.2, at least the DDA formulation (is it LDR, the default in ADDA?), the dpl (number of dipoles per wavelength), and version of the ADDA code. Was the number of dipoles changing with size parameter? The authors should also specify the typical computational requirements (at least, for the largest particles). This can also explain/justify the upper limit of the size parameter, which was mentioned by another reviewer.*

Regarding this, we added the following text to Section 4.2: ”Details of the particle shape and inhomogeneity treatment are closely connected to, and partially dictated by, the light-scattering method chosen for the scattering computations. Of the publicly available methods, the discrete-dipole approximation was considered to be most suitable for the task. The computations were carried out using the parallelized version of the discrete-dipole approximation code ADDA 1.1 (Yurkin and Hoekstra, 2011). In the DDA, the particle is presented with small, discrete volume elements which are treated as dipoles, and scattering is computed by integrating over the electric fields induced by these dipoles. Therefore, the resolution of the volume discretization, described with an interdipole separation d, dictates how small-scale morphological details can be considered. Also, the accuracy of the computed fields depends on d: it should be small compared to the wavelength of radiation inside the particle. This criterion is often evaluated by the $|m|kd$ value but, according to Yurkin and Hoekstra (2011), no specific value that would guarantee a reasonable accuracy in all cases can be given; rather, they advice to first test the accuracy of the DDA in the desired situation to find the most suitable resolution. This is as fine as needed for the accuracy, but no finer, to keep the computational demands feasible.

The volumes of the stereogrammetric shapes were discretized with approximately 100000 dipoles. This resolution was first used through size parameters 0.5--16, resulting in $|m|kd \sim 0.90$ for the largest size parameter. Zubko et al. (2010) concluded for their irregular particles that $|m|kd < 1$ is an adequate accuracy criterion but, in our case, already $|m|kd \sim 0.90$ turned out to result in an insufficient accuracy with relative errors as high as 30% for S44/S11 at specific scattering angles. S22/S11 was also moderately sensitive. Consequently, the number of dipoles was increased for $x \geq 10$ by dividing each dipole into eight smaller dipoles, thus preserving the resolution of the morphological details through all size parameters. After this, the number of dipoles per wavelength was 22.7 and $|m|kd \sim 0.45$ for the largest size parameter $x = 16$. With the chosen resolution, the running times of the parallelized computations varied from hours to a few days; the most demanding case was Sil I, which required 154 hours on 46 processors, resulting in the total

of 7084 hours of CPU time."

We also added one sentence to the next paragraph: "The connection between dipole polarizability and macroscopic refractive index was established in the computations using the lattice dispersion relation, which is the default setting in ADDA."

> *1b) It is important to control/quantify the simulation uncertainty. The author cite (Zubko et al., 2010) on p. 18463. However, it is unclear if it is a general reference, of if the authors adopt the convergence (accuracy) criterion from it. If the former is true, the authors should at least also refer to [Yurkin M.A. and Hoekstra A.G. The discrete dipole approximation: an overview and recent developments, J. Quant. Spectrosc. Radiat. Transfer 106, 558–589 (2007). http://dx.doi.org/10.1016/j.jqsrt.2007.01.034], which contains a comprehensive overview of DDA accuracy in different cases. If the latter (adopting convergence criterion) is true, this is very arguable without additional justification, since results of Zubko et al. are rather specific. Anyway the authors should explain why (based on what criterion) they used so many dipoles for the largest size and quantify (at least roughly) the accuracy of their simulations.*
> *The methods to do it are described, e.g., in Section 2.1 of (Yurkin and Hoekstra,2011), based on simulation of the same particle with different dipole sizes. The largest number of dipoles (10^5) is not that large (even considering orientation averaging), so substantially refining the discretization (decreasing dipole size at least 2 times) should be computationally feasible for several representative examples. Especial care is required for backscattering quantities, since DDA with default parameters may calculate them with large errors in certain cases.*

We had assumed that, since particle shapes considered by Zubko were similar to those considered here, their finding that $|m|kd < 1$ would be sufficient also for us. Prompted by your insightful comments, we carried out test simulations and found out that it is not the case.

Two types of test simulations were considered. On the one hand, we rediscretized the targets from the original high-resolution triangle mesh using double the original resolution. On the other hand, we replaced each of the dipole in the original discretized targets by eight dipoles (2x2x2 cube). Both approaches lead to high-resolution target files with eight times the original number of dipoles. The former approach increases both the accuracy of the target description and the accuracy of the DDA solution due to the increased the number of dipoles per wavelength, while the latter approach only improves the accuracy of the DDA solution. Only homogeneous cases were considered, for simplicity. The test simulations revealed that, while these two high-resolution cases did not show significant differences in their scattering from each other, they both differed quite significantly from the results obtained for the original, low-resolution targets at the largest size parameters considered. This suggests that the original target discretization accuracy was sufficient, but the original computational accuracy is not. S44/S11 was most affected, showing differences that exceeded even 30% at certain scattering angles; S22/S11 was also clearly affected, while for other elements the original low resolution seemed sufficient.

We therefore redid all the scattering computations for size parameters $x \geq 10$ using the double-resolution targets. For simplicity, we decided to use the double-resolution targets of the second type, where the double-resolution targets are obtained from the low-resolution targets by

replacing each original dipole with eight new dipoles (2x2x2). This provided us with sufficient accuracy, while it assured that the spatial distribution of mineral components is consistent for both resolutions.

Accordingly, we have revised all the affected figures and rewritten the paragraph concerning the simulations as explained in our response to the previous comment.

> *1c) The authors should specify the orientation-averaging scheme in Section 5.2 additionally to the plain number of orientations. Is it the built-in scheme of ADDA? If yes – what is the number of alpha, beta, and gamma angles? Why those numbers were selected, what is the estimated uncertainty (of different computed quantities) due to orientation averaging? If ADDA's scheme is used, the raw output does contain estimates of this uncertainty.*

We added the following text about this to Sect. 5.2: "Scattering results were averaged over 8704 particle orientations with the built-in orientation averaging scheme of ADDA, using the following number of different Euler angles: $\alpha = 32$, $\beta = 17$, and $\gamma = 16$. The relative error of C_{ext} resulting from orientation averaging was estimated to be below 0.015 for the highest size parameter considered."

2) The missing element in the whole approach (important for validation) is measurements of the scattering from single particles. The authors correctly mention that no such measurements has been made for these specific particles. However, there exist techniques potentially capable of performing such task, although coupling these systems with the shape reconstruction of the _same_ particle is not easy: Air flow systems measuring 2D scattering patterns – developed by Kaye et al. See the review in [Kaye P.H., Aptowicz K., Chang R.K., Foot V., and Videen G. Angularly resolved elastic scattering from airborne particles, in Optics of Biological Particles, eds. A.G. Hoekstra, V.P. Maltsev, and G. Videen, Springer, Dordrecht, pp. 31–61 (2007). http://dx.doi.org/10.1007/978-1-4020-5502-7_3] Air flow systems measuring holographic patterns – see e.g. [M. J. Berg and G. Videen, Digital holographic imaging of aerosol particles in flight, J. Quant. Spectrosc. Radiat. Transfer 112 p. 1776-83 (2011). http://dx.doi.org/10.1016/j.jqsrt.2011.01.013] (Liquid) flow cytometers, measuring 1D and 2D scattering patterns: [Strokotov D.I., Moskalensky A.E., Nekrasov V.M., and Maltsev V.P. Polarized light-scattering profile - advanced characterization of nonspherical particles with scanning flow cytometry, Cytometry A 79A, 570–579 (2011). http://dx.doi.org/10.1002/cyto.a.21074] [Jacobs K.M., Lu J.Q., and Hu X.-H. Development of a diffraction imaging flow cytometer, Opt. Lett. 34, 2985–2987 (2009). http://dx.doi.org/10.1364/OL.34.002985]. So the authors should discuss (some of) these techniques in the introduction and, probably, in the discussion or conclusion with respect to possible future work.

Finally, a minor comment – the authors may consider pointing out in the Introduction an analogy between their approach to mineral aerosols and the realistic modeling of light-scattering from biological particles, based on confocal images. See e.g. [Brock R.S., Hu X., Weidner D.A., Mourant J.R., and Lu J.Q. Effect of detailed cell structure on light scattering distribution: FDTD study of a B-cell with 3D structure constructed from confocal images, J. Quant. Spectrosc. Radiat. Transfer 102, 25–36 (2006). http://dx.doi.org/10.1016/j.jqsrt.2006.02.075] and [Orlova D.Y., Yurkin M.A., Hoekstra A.G., and Maltsev V.P. Light scattering by neutrophils: model, simulation, and experiment, J. Biomed. Opt. 13, 054057 (2008). http://dx.doi.org/10.1117/1.2992140]

We thank Dr. Yurkin for pointing out these important references. They have been appropriately notified in the introduction with this added

paragraph: ”The stereogrammetric approach to model scattering by single dust particles more realistically has some analogies to the confocal microscopy measurements method used by Brock et al. (2006) and Orlova et al. (2008) to model light scattering by biological particles: both methods result in a retrieved, three-dimensional model shape for a single particle. Orlova et al. (2008) even validate their computed scattering results by comparisons to measurements performed with a scanning flow cytometer. Indeed, several approaches in addition to flow cytometers (Jacobs et al., 2009; Strokotov et al., 2011) have been taken towards measuring scattering by individual particles: for example, air flow systems measuring two-dimensional scattering patterns (Kaye et al., 2007) and digital holographic imaging (Berg and Videen, 2011).”