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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Volcanic ash from Iceland over Munich: mass concentration retrieved from ground-based remote sensing measurements" *by* J. Gasteiger et al.

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We thank the referee for his/her comments which help us to improve the paper. In the following, comments by the referee are in italic font, answers by the authors in normal font.

The reviewer raises a lot of useful questions addressing different aspects of aerosol remote sensing and inversion. It should be clear that not all aspects could be elaborated in the present paper in full depth, otherwise the focus of our paper would be lost. We want to stress that several topics are discussed in already submitted papers. Nonetheless, in the following, we address all questions raised by the reviewer.

1) It remains unclear in how much the author deal in their computations with the fact that the observed plume likely did not consist of non-spherical ash particles only. There must have been sulfuric acid particles present.

All model particles are non-spherical. Unfortunately, no in-situ data for the observed ash plume are available. The history of dispersion showed no mixing with other aerosol types. Given the measured, very high, linear depolarization ratios and the very low relative humidity (<40%), it is unlikely that a significant amount of spherical/wet sulfuric acid particles were present in the plume. Significant amounts of non-spherical/dry sulfuric acid particles could be in agreement with the depolarization measurement. However, such kind of particles typically are smaller than our lidar wavelengths. If a significant amount of such particles would have been present in the ash plume, the extinction coefficient at 355nm would have been higher than at 532nm. Consequently, it is unlikely that sulfuric acid particles notably contributed to the optical properties of the observed ash plume. It could be that sulfuric acid accumulated on ash particles; this would hardly affect the spectral changes of the extinction of the ash, and if the non-sphericity of the ash particles is preserved after accumulation, the linear depolarization ratio could be as high as measured over Maisach. We are confident that our approach assuming only non-spherical particles is appropriate for the observed plume.

2) In view of 1) what is the effect on the results if two different refractive indices (two different particles types) are used?

As a first test of this, we allowed the ensembles to consist of two independent modes. The ratios of the number densities of both modes is sampled in the range 0.001<N_2/N_1<1000 (logarithmic sampling). The ranges of the other parameters of each mode are the same as given in Table 2, i.e., both modes consist of spheroids. For η (mass/ext-ratio) at $\lambda = 532$nm, we get 1.5 (0.9··3.4) g/m^2, i.e., the lower limit of the uncertainty range and the median values are only slightly higher, but the upper limit is approximately 50% higher than for mono-modal model ensembles. This is probably related to the insensitivity of the lidar signals to large particles (Section 3.5): several ensembles with a particle mode consisting of very large particles are in agreement with the lidar measurement. We add a sentence in Section 3.5 about the outcome of this test.

As a second test, the first test is modified by assuming spherical particles for the second mode. The ranges for the first mode, and the ranges for the size distribution and the refractive index of the second mode are not changed. We get 1.5 (0.9··4.1) g/m^2 for η at $\lambda = 532$nm (including the volume of second mode for calculation of η). There are several solutions, where the second mode consists of large ($r \approx$5-10μm) spherical particles with low m_r ($< \approx$1.4), which do hardly contribute to the extinction and backscatter, but contain most of the mass.

3) The authors should address in more detail the issue of choosing an appropriate axis ratio distribution. To my understanding the authors use results from Kandler et al., who present axis ratios for mineral dust particles observed over the Sahara (SAMUM experiment in Morocco).

From Kandler et al. 2007, we adopted only the parameterization of aspect ratio distributions by modified log-normal distributions (specified by two parameters in a similar fashion as it is done for log-normal size distribution, see Eq. 13). However, we do not use the parameters that were found specifically for mineral dust particles collected during SAMUM. Instead, we allow the parameters of the aspect ratio distribution to vary within the wide ranges given in Table 2.

4) How much proof do the authors have regarding the applicability of a log-normal size distribution in their computations? Volcanic ash may not show such a shape of the size distribution.

Log-normal size distributions, wavelength-independent size-independent bulk refractive indices, and the approximation of the particle shape by spheroids are approximations. Log-normal distributions are usual and established parameterizations for aerosol size distributions. The true size distribution will probably deviate from this assumption

to a certain degree.

Regarding the mono-modality of the size distribution of the ash over Maisach, we have the following indications: A potential optically-relevant second mode with particles smaller than the shortest lidar wavelength (i.e. fine mode) would result in a decrease of the extinction coefficient from 355nm to 532nm, as discussed in point 1). Inversion results given in point 2) show that a second mode consisting of very small particles is unlikely. As lidar signals are hardly sensitive to volume in very large particles ($r < \sim 3\,\mu$m, see Figure 5), ensembles with a second mode consisting of very large particles can be compatible with the lidar measurements, as shown under point 2). Such a mode of very large particles could have large impact on the ash mass concentration. To exclude this, we performed the consistency check using aureole radiances (sensitive to $r < \sim 10\,\mu$m, see Figure 11). A second mode consisting of very large particles is not found. These findings suggest that the size distribution in the observed ash layer can be well described by a mono-modal log-normal distribution, though "mild" bi-modality (modal radii close to each other) can not be excluded on the basis of our observations.

5) Do the authors have proof for the completeness of the solution space (in the mathematical sense)? There may be "gaps" in the grid of parameters they use in their computations. The parameters of the particle size distribution and particularly the complex refractive index are quite restricted. For instance, a denser grid could result in different mean values. Furthermore, the uncertainty level might increase if a broader range of complex refractive indices is used in the computations.

Maybe, there is a misinterpretation due to mixing up of Tables 1 and 2. We randomly sample the parameters of the model ensembles within the ranges given in Table 2. The density of the solution space is given by the resolution of the floating point variables. Of course, because the available computation time is limited, not all possible parameter-combinations are modeled, but a large number of them, until the distributions of the ensemble properties converged sufficiently.

The optical properties of the ensembles are calculated from the optical properties of single particles (available according to Table 1). To calculate the optical properties, e.g., for ensembles with an real refractive index m_r=1.45, the refractive indices from the single particle database are weighted accordingly, i.e., three particles with m_r=1.44 occur together with one particle with m_r=1.48. This approach introduces a deviation of the optical properties compared to the exact calculation for particles with m_r=1.45; tests, however, showed that this deviation typically is in the order of 1% or smaller for the lidar-relevant optical properties. To avoid misinterpretation, we change the caption of Figure 1 to "Grid points of parameters in single particle scattering database". Furthermore, we describe in more detail how the optical properties of ensembles are calculated from the single particle database.

6) Considering 5) in the computations could change the mass estimates.

We tested the influence of the m-interpolation by removing every second real part m_r and every second imaginary part m_i from the grid covered by the single particle database (Table 1), i.e. the step width for m_r is then +0.08 and for m_i is *2. The effect on (median and 2.5%- and 97.5%-quantile of) the retrieved parameters is very small: η and r_{eff} do not change within statistical uncertainty (≈0.5%); m_r: 1.428 (1.344··1.488) vs. 1.431 (1.351··1.495); m_i: 0.0071 (0.0024··0.0152) vs. 0.0069 (0.0024··0.0152).

7) The authors present ratios of radiances in their consistency check (section 4, page 14). They use two angles and the wavelength 1020 nm. I agree that 1020 nm is the best wavelength to consider. However, are the results consistent with computations at another wavelength? Particle shape may have some influence on the computations at different wavelengths, and in fact this test might tell a bit more about particle shape itself.

We applied the same procedure as for the 1020nm also for the 675nm (see point 11 of this review).

The shape of the main diffraction peak of large aerosols is primarily governed by the

cross sections of the particles. For cross-section-equivalent definition of particle size, like we use in the paper, the main diffraction peak is much more sensitive to particle size than to the particle shape (at least for the sizeparameter/shape-range relevant for ash). Consequently, we expect that not much can be learned about the particle shape from scattering at these angles.

8) The authors compare their results from the radiative transfer model to lidar observations? This comparison raises two questions: i) how do the authors account for the overlap effect? ii) How do the authors treat the fact that there is urban pollution in the boundary layer, whereas volcanic ash is on top.

i) To account for the overlap effect of the lidar we extended the extinction coefficient at the lower end of the lidar profile to the ground. The overlap of MULIS is comparatively low (≈600m), and near field telescope measurements of POLIS (overlap ≈100m, see paper in preparation "Characterization of the planetary boundary layer during SAMUM-2 by means of lidar measurements" by Groß et al.) were used to check the applicability of our approach. The estimated uncertainty is reflected in the range for the optical depth of the boundary layer aerosol given in Table 4 (0.056 - 0.084).

ii) As described in Section 4.2, the boundary layer aerosol is treated as a separate aerosol layer in the radiative transfer simulations, and the uncertainties about the properties were considered as given in Table 4.

9) The authors compare lidar in Maisach with sunphotometer at the university in Munich. How is the spatial difference accounted for? The data sets describe different portions of the plume, and the plume certainly was rather inhomogeneous. I assume that number concentration varied quite a bit, and particle size may have change, too.

For our purposes, the difference in the number concentrations at both sites is only relevant for the optical depth of the ash layer in the radiative transfer calculations for Munich. To account for such differences, the ash optical depth is varied from 0.216 to 0.324. More important than the similarity of the ash optical depth is the similarity of the intensive properties in the ash layer. Differences of the intensive properties would become relevant for the comparison of the results of Sect. 3 to results of Sect. 4. The lidar measurements in Maisach indicate no change of intensive properties of the ash between midnight and noon of 17 April. E.g., a paper by Groß et al. is in preparation showing the temporal stability of δ_l in the ash layer in the morning of 17 April. As stated in Sect. 4.1, the ceilometer in Munich showed similar vertical and temporal distributions of the volcanic ash as the lidars in Maisach; ceilometers in Augsburg, Weihenstephan, Hohenpeißenberg also showed similar distributions (Emeis et al., this special issue). We believe it is safe to assume that the intensive properties of the ash were similar at the horizontal distance of 25km.

10) Page 15, first full paragraph: the authors say that they use a scattering angle of 3 deg and 4 deg for their computations. Is there any reason that no angle closer to the sun disk was used?

The scattering angles 2° and 2.5° are affected by straylight. Measurements in an almost-aerosol-free situation on 7 April 2010 show that measurements at these scattering angles are not useable.

11) On page 15 the authors say that "the largest wavelength of the CIMEL is best suited .. less affected by the boundary layer aerosol." So let me ask again: are the results consistent with computations done at another wavelength? Or let me rephrase: what does "less affected" mean in quantitative terms?

We did the exercise also for 675nm (measured Λ is 0.823): same optical thickness of the ash layer as for 1020nm, but doubled optical thickness of the boundary layer aerosol (Angstrom approx. 1.7), the other parameters are same as given in Tab. 4.

The results are shown in the figure at the end of this reply (same style as for Fig. 9 in the paper): the different colors denote different shapes of size distributions, black dotted lines show the uncertainty range from the simulations, and black dashed lines the uncertainty of the measurement. r_{eff} up to 1.4μm are compatible with the measured

Λ at λ=675nm, which overlaps with the r_{eff} found for λ=1020nm. For example, r_{eff}=1μm agrees with the measured Λ for SD#2 (green) and SD#3 (blue) at both wavelengths (675nm and 1020nm).

12) Page 15, 4.2, MYSTIC: does this code treat particles of non-spherical geometry? From the text it does not become clear if MYSTIC applies Mie theory. In view of the unusual measurement situation it may be crucial that all tools used in this study (lidar, cimel, radiative transfer, forward simulations) can treat the particles on the basis of non-spherical scattering theory.

The MYSTIC simulations use single scattering properties of aerosol ensembles consisting of non-spherical particles as outlined in Section 4.2. The single scattering properties are calculated indepedently of MYSTIC and then used as input for MYSTIC. Section 4.2 is improved to clarify that single scattering properties are calculated independently of MYSTIC.

13) Page 16: how did the authors consider the fact that the AERONET algorithm cannot derive particle size distributions for which particle radius is above 15 mu? Papers by Mueller et al. (JGR 2010, paper # D07202; SAMUM results) and Mc Connell et al. (JGR 2008, paper # D14S05; DODO campaign) point out to this problem.

We did not use the AERONET inversion algorithm. The connection of our study to AERONET is only given by the fact that the Munich CIMEL is part of the AERONET network.

14) Page 20, second paragraph: the authors say that their results are consistent with AERONET retrieval results. Again let me point out that AERONET cannot retrieve effective radii larger than around 2 micrometer because the retrieved particle size distributions are restricted to 15 micrometer. The authors should discuss this specific effect in more detail.

We only use radiances measured by a CIMEL which is part of AERONET; we did not compare the outcome of our exercise in Section 4 with AERONET retrieval results. The comparison of our results to AERONET retrieval results is beyond the scope of our paper.

Abstract: The abstract should contain more information on the implications.

The revised abstract contains more information on the implications.

Page 2: The authors refer to Jaeger et al. As they speak of lidar techeniques they should are refer to papers by Ansmann et al, Wandinger et al., Carswell et al. (Raman lidar).

We assume that the reader is familiar with the basic principles of lidar. As we want to refer to lidar measurements of volcanic aerosols in general, we only refer to papers where this topic is covered. Later in the text, when advanced lidar techniques are explained, more references are included.

Page 3, first line: "in the order" should be "on the order".

Changed.

Line 2: "few years" should be "several years".

Changed.

Line 3: in the order should be on the order.

Changed.

First full paragraph on page 3: please rephrase the sentence "During the eruption of the ... effects for Europe". The sentence is difficult to understand.

Improved.

Page 3, line 13: "was known" should be "is known".

This sentence was removed due to another comment.

Page 3, line 6 from bottom: "allows" should be used with "allows for..." or "allows one to ..." I find this wording "allows" in many spots of the paper. Please make changes accordingly.

Changed throughout the paper.

Page 5, line 3: please provide the geographical location of the Iceland volcano.

Added.

Page 5, line 13: delete comma before "7 km".

Changed.

Page 6, last line: no comma after "because"?

Corrected.

Page 7, line 2 from the bottom: "an upper limit". Please specify.

A reference to Table 1 and Wiegner et al. is added, where the upper limit of the applicable size range of the T-matrix method is shown.

Page 8, first line: what means "improved geometrics optics code"? Please specify.

We restructured these sentences and mention that Yang's code considers egde-effects for the calculation of the efficiencies.

Page 8, line 3: You write "better reliability in the lidar-relevant backscattering direction". What means "better reliability"? Please specify.

Explained in more detail.

Page 8: "are weighted according to the actual ...": please explain this procedure in more detail. It is unclear how this is done.

We now provide more detail about the procedure.

Page 9, effective radius definition: you write 3V/4A. It should be 3V/A.

This depends on how A is defined. In Schumann et al. 2010, A is defined as the projected area density. We add more explanation in the paper.

Page 13, you refer to Patterson et al 1983. You should point out what Patterson et al actually measured, and whether it is comparable to your kind of data. Keep in mind that mineral dust may be highly variable. There likely is no "unique" value for the refractive index.

We agree that there is a high variability of the refractive index of ash. It is difficult to find statistics about the refractive index of ash in the literature. Studies dealing with the real parts for specific ash cases typically find m_r=1.50 or higher. Patterson et al 1983 is one example for ash collected at some distance from the El Chichon volcano. We now mention that Patterson is just an example for one specific case.

Page 16, line 7/8: how do you treat particles in the overlap region (Angstrom exponent assumption)?

We assume that the particles in the overlap region have the same properties as the boundary layer particles above the overlap region.

Page 16, line 12: you consider an aspect ratio of 1.8 in your computations. Is this a representative value, given the fact that the ash particles possess an aspect ratio distribution.

The real ash, of course, consists of particles with a wide aspect ratio distribution. According to microscopy measurements the median aspect ratio for volcanic ash is close to 2.0 (Schumann et al. 2010). We considered different aspect ratios for our simulations: E.g. shape C (Figure 6) has an aspect ratio of about 2.3. Thus, we did not systematically underestimate the aspect ratio.

Table 4: I am a bit confused about the numbers. Why do you assume the possibility of rather high absorption (imaginary part of up to 0.05) in the boundary layer, whereas

you neglect this possibility for the volcanic ash plume? Your effective radius is up to 3 micrometer in the volcanic ash plume. Is there a reason why you do not test any larger value for reff? Simply give it a try regardless of what Angstrom exponents tell you.

Regarding the refractive index of volcanic ash we looked in the literature and did not find highly absorbing ash aerosols. Based on our review of literature, we think that m_i=0.01 can be regarded as an upper limit for the m_i of ash at 1020nm.

Note, that the ratio Λ_{11} is hardly sensitive to the absorption properties. For example, if we compare the ratio Λ_{11} for an ensemble of spherical particles with $r_{\text{eff}} = 1.2\,\mu$m, $\sigma = 2.4$, $m = 1.55+0.01i$ to the Λ_{11} of an ensemble with the same properties but $m_i = 0.05$, we see only small differences for Λ_{11} though the absorption is quite different: Λ_{11}=0.833 for $m_i = 0.01$ vs. 0.827 for $m_i = 0.05$.

The refractive index of the boundary layer aerosol is much less known due to its high variability; thus wider ranges were covered for the boundary layer aerosol compared to the volcanic ash particles.

To investigate the effect of increasing the effective radius to $5\,\mu$m, we calculated the ratio Λ_{11} for the four forms of size distributions (Table 4): Λ_{11} for $r_{\text{eff}} = 5\,\mu$m is by 0.03 to 0.15 smaller than Λ_{11} for $r_{\text{eff}} = 3\,\mu$m; consequently, it can be concluded that Λ (Figure 8) would continue to decrease for $r_{\text{eff}} > 3\,\mu$m, further increasing the deviation from measured Λ.

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 26705, 2010.

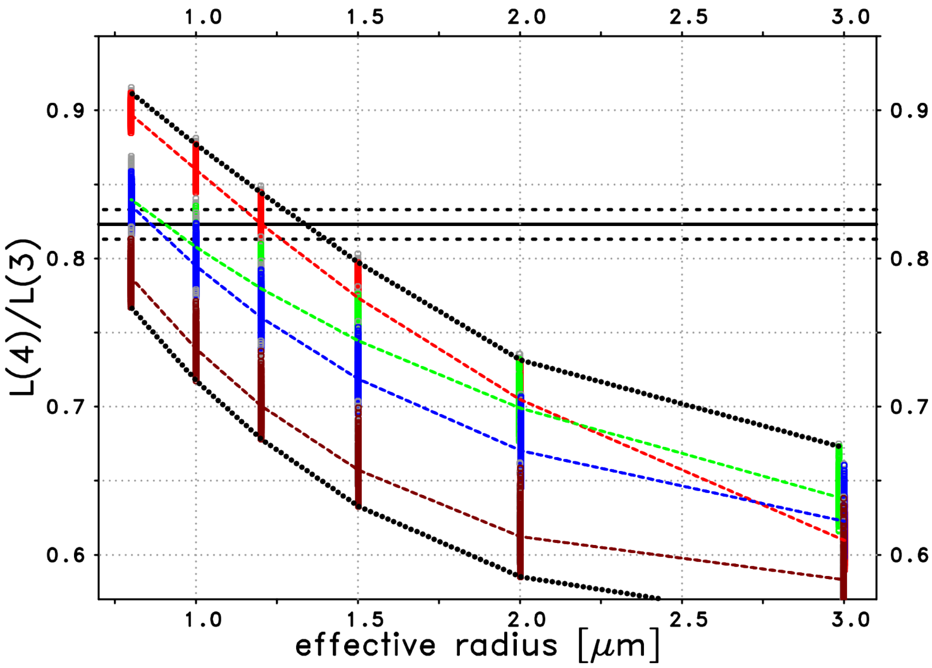

Fig. 1. Ratios of aureole radiances at 675nm (compare to Figure 9 of paper)