

Response to Referee # 2

We thank the referee for his detailed review and valuable comments. The manuscript has been modified according to the suggestions proposed by the reviewer. The remainder is devoted to the specific response item-by-item of the reviewer's comments:

Reviewer comments

General comments.

Manuscript discusses characterization of volcanic aerosols from Mt. Etna and Mt. Stromboli degassing plumes using both in situ and remote sensing techniques. In situ observations consisted of aerosol size distribution (ASD) measurements using Forward Scattering Spectrometer probes (FSSP) and remote sensing approach was based on inversion of combined observations of angular scattering intensities and extinction obtained by airborne Polar Nephelometer. Information content of Nephelometer observations was analyzed using Principal Component technique which showed possibility to distinguish scattering pattern of volcanic aerosols from the one of clouds (cirrus and contrails). Inversion of Polar Nephelometer data resulted in relatively low values of the real part of refractive index of volcanic aerosol: 1.35 to 1.38. This was attributed to the presence of cavities inside particles which effectively decrease the real part of refractive index. Manuscript is very well written and the goals and the techniques used are clear. I believe that the subject of the manuscript is in scope of ACP. Paper certainly can be published.

Specific Comments.

1. My main concern is the effect of uncertainty in extinction coefficient (25%) and the limited range in scattering angles (15 to 162) on the accuracy of aerosol retrievals. The authors do not discuss these issues at all. However absence of aureole measurements can affect the ASD retrievals, especially Deff. In addition the uncertainty in extinction coefficient can affect the accuracy of retrieved complex refractive index. Therefore I suggest authors to conduct a simple sensitivity studies: calculate synthetic measurements for the complete range of scattering angles and then invert them using 15-162 range only. In addition, add/subtract 25% to/from extinction coefficient and estimate corresponding uncertainty in retrieved aerosol parameters. I believe these sensitivity tests will make the conclusions of the manuscript much more solid.

A large set of sensitivity tests related to the air-borne and laboratory nephelometers of the Laboratoire de Météorologie Physique (LaMP) were performed in the mid-noughties with the participation of the first author of the manuscript. The main attention was paid to cases of angular scattering intensities (ASIs) measured within a limited range of scattering angles. Some results of the tests were reported in the work by Verhaege et al., (2008). It was shown that despite the absence of aureole and backward measurements the real part of the refractive index and the microphysical parameters can be retrieved in the case of the low absorbing particles.

The following text has been added to Section 5.

That conclusion corroborates with the results of sensitivity tests performed by Verhaege et al., (2008) for ASIs measured within a limited range of scattering angles. It was shown that despite the absence of aureole and backward measurements the real part of the refractive index and the microphysical parameters can be retrieved in the case of the low absorbing particles.

2. Did authors really try different initial guesses for inversion code to make sure the global minimum is reached as they discussed at page 9?

The application of the Dubovik's package in our work is described in more details in the supplementary material (see also "Response to Referee # 1"). To summarize briefly, different initial guesses for inversion code were performed on a multidimensional grid of the input parameters using a number of input files.

The following text has been added to Section 4.1.

Details of the software package, we used in this works, as well as of the code application are described in the supplementary material. To summarize briefly, different initial guesses for the inversion code were performed on a multidimensional grid of the input parameters using a number of input files. The minimum and the maximum sizes of particles as well as the spherical/non-spherical partitioning ratio belong to the set of assessed parameters in addition to the refractive index and the size distribution.

3. Is Maxwell Garnett mixing rule really applicable to this type of aerosol particles? How the applicability was estimated and what is the accuracy of estimated air voids?

The effective-medium approximation (EMA) along with the Maxwell Garnett mixing rule have already been used in a number of works to calculate optical properties of porous particles (see, e.g., Voshchinnikov et al., 2007; Kylling et al., 2014). The question of EMA applicability is discussed in details by Mishchenko et al., 2016.

Our estimation of air voids is given as an interval of values, that is, about 18 to 35 % in terms of the total volume. That interval represents errors (accuracy) of our estimations.

The following text has been added to Section 5.

The effective-medium approximation along with the Maxwell Garnett mixing rule have already been used in a number of works to calculate optical properties of porous particles (see, e.g., Voshchinnikov et al., 2007; Kylling et al., 2014). The question of EMA applicability is discussed in details by Mishchenko et al., 2016.

4. In Table 1., the residuals seem too high for "optically" spherical. It would be interesting to look at the dependence of angular measurements fit as a function of scattering angle.

The right panels of Figure 4 show the measured (solid red circles) and the reconstructed (solid black circles) angular scattering intensities (ASIs). The reconstructed (retrieved) ASIs were computed from the retrieved size distribution.

In other words, the dependence of angular measurements fit as a function of scattering angle is shown by the solid black circles at each right panel of Figure 4. It is seen that the measured ASIs are well fitted by the retrieved phase functions.

The following text has been added to the corresponding paragraph.

In other words, the measured ASIs are well fitted by the retrieved phase functions.

References (additional to the acp-2016-183 discussion manuscript).

Kylling, A., Kahnert, M., Lindqvist, H., and Nousiainen, T.: Volcanic ash infrared signature: porous non-spherical ash particle shapes compared to homogeneous

spherical ash particles, *Atmos. Meas. Tech.*, 7, 919-929, doi:10.5194/amt-7-919-2014, 2014.

Mishchenko, M.I., Dlugach, J.M., Yurkin, M.A., Bi, L., Cairns, B., Liu, L., Panetta, R.L., Travis, L.D., Yang, P., and Zakharova, N.T.: First-principles modeling of electromagnetic scattering by discrete and discretely heterogeneous random media, *Phys. Rep.*, 632, 1–75, doi:10.1016/j.physrep.2016.04.002, 2016.

Voshchinnikov, N., G. Videen, and T. Henning: Effective medium theories for irregular fluffy structures: aggregation of small particles, *Appl. Opt.* 46, 4065–4072, doi:10.1364/AO.46.004065, 2007.