**Atmospheric
Chemistry
and Physics
Discussions**

Interactive comment on "Physical and optical properties of 2010 Eyjafjallajökull volcanic eruption aerosol: ground-based, LIDAR and airborne measurements in France" *by* M. Hervo et al.

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General comments: Hervo et al. describe a method where ground-based in-situ measurements are used to characterize volcanic ash aerosols at Puy de Dome (1.5km a.s.l., France). Using these measurements, the mass extinction ratio and the lidar ratio of the aerosol is calculated. In the next step, these parameters are used to calculate the mass concentration profile from measurements of a nearby backscatter lidar. The vertical and temporal development of the ash layers and their mass concentrations are intercompared in two case studies, whereby the li-

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dar profiles, ground-based in-situ measurements, aircraft in-situ measurements, and FLEXPART simulations are considered. The manuscript contains interesting and original work. However, the description of the instruments and the methods is often too superficial, the uncertainties of the measured parameters are not properly accounted for and discussed, and the calculation of the lidar ratio seems to be flawed (see below). Therefore, I recommend to publish the manuscript in ACP only after substantial revisions.

General answers: Following the recommendations of the reviewer the uncertainties have been calculated and discussed for the most relevant parameters. The Lidar ratio has been calculated using 2 different methods and the results were compared on 2 cases to analyze the effect on the non spericity.

Specific comments: [page/line]
24632/18: "dominance" might be replaced by "presence": If the supermicronic particles were dominant with respect to Angstrom, the Angstrom would close to zero.
Author: Corrected

24632/24: Are the uncertainties of your method really this low? If you write the uncertainty in the abstract, please specify clearly what it covers. Otherwise such low uncertainties are misleading.
A: The uncertainties presented are the standard deviation of the data retrieved during the main event and we have replaced the term "uncertainty" with the term "variability". An evaluation of the systematic errors in the mass retrieval is now presented in section 2.5.

24636/3,4: Please add details about the "TEOM-FDMS 8500C", for example cutoff diameter, measurement principle, etc... I think it is a very important instrument in your study.
A: The following description has been added to the text: "Total mass concentrations

of aerosol particles are measured with the commercial instrument: Tapered Element Oscillating Microbalance-Filter Dynamic Measurement System 8500C (Grover et al., 2005;Ruppecht et al., 1992). The TEOM-FDMS uses intermittent sampling through a filter to account for the mass of semi-volatile material lost due to volatilization. The resolution of the TEOM-FDMS given by the constructor is 2.5μ.m-3$\pm$1% of the measurement." "All the in situ instruments presented in this paper measure the dry aerosol properties and are sample after the inlet with a cut-off diameter of 30μm"

Section 2.1: In addition, the description of relevant details of the other instruments would also be very helpful.

A: To our opinion, now, the most relevant details are mentioned in the manuscript. We invite the reader to look at the corresponding cited manuscripts for further information.

24636/10: Please call this parameter "Angstrom exponent for scattering" or "scattering Angstrom exponent" throughout the manuscript because the default meaning for Angstrom exponent is for extinction.

A: Corrected

24636/15: An Angstrom of 1 means that large and small particles are mixed and that they are almost equally relevant for the extinction (if 2 is assumed for small and 0 for large particles). Nevertheless, large particles might be dominant with respect to particle volume if the Angstrom is around 1. If "dominated" is meant with respect to particle volume, this should be mentioned here.

A: Corrected

24637/3,4: "... Angstrom exponent is equal to unity (i.e. the absorption is independent from wavelength variation... ": This is wrong. The Angstrom exponent is zero for wavelength independent properties.

A: Now the wavelength dependance is taken into account. The absorption is calculated at 355nm instead of 637nm using an Absorption Ångström exponent of 1. The

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corresponding uncertainties are comment in next paragraph.

24637/8: How do you calculate the mass extinction factor at lambda=355nm? Do you use Angstrom assumption? Do you assume wavelength-independent properties? And 24638/20: How do you account for the difference in wavelength (lidar 355nm vs. maap 637nm)?

A: The following sentence is now included in section 2.1 "The scattering properties are calculated at 355 nm using the scattering Angstrom exponent (α). According to previous studies the uncertainties on the scattering coefficient can be consider of 7% (Anderson and Ogren, 1998;Anderson et al., 1999). After propagation (Wagner and Silva, 2008),the value of the error in the calculation of α was determined ($\Delta\alpha$=0.32).Extrapolating the scattering coefficient at 355nm, from measurements at 450nm and the Angstrom exponent, generates an error of 19% with the following equation:

$$\Delta\sigma\text{sca}(355)/\sigma\text{sca}(355) = \ln(637/450)\Delta\alpha + \Delta\sigma\text{sca}(450)/\sigma\text{sca}(450)$$

The absorption coefficient is calculated at 355nm with an Absorption Ångström Exponent (AAE) equal to unity (i.e. the absorption is directly proportional to the wavelength variation: $\sigma\text{abs}(355\text{nm})\approx 637/355*\sigma\text{abs}(637\text{nm})$). AAE is correlated with aerosol composition or type: for most urban industrial site, the AEE value is close to one but can increase until 2.5 for others kind of particle (Russell et al., 2010). Using an AAE of 1 ($\sigma\text{abs}(355)$=1.79*$\sigma\text{abs}(637)$) instead of 2.5 ($\sigma\text{abs}(355)$=4.31*$\sigma\text{abs}(637)$) implies an important error on the absorption coefficient by a factor of 2.41. Nevertheless, as the single scattering albedo is relatively high (0.97$\pm$0.01) during the measurement period (i.e. the scattering coefficient is significantly higher than absorption coefficient) the absorption impact on the extinction is limited. Indeed the extinction was calculated for both AAE of 1 and 2.5 in order to estimate the error generated by such an approximation, when using the AAE of 1 instead of 2.5, the extinction was underestimated by only 4.2% on average with a 10.4% maximum on the period considered in this study.

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The same error will be propagated to the mass extinction ratio η and to the Lidar mass concentration."

24637/5-9: Do the instruments measure "dry" or "wet" aerosol properties? AND 24638/20-21: Please explain in more detail how the refractive index is inverted. Is the measured size distribution also "dry"?

A: Comment added in section 2.1: "All the in-situ instruments presented in this paper measure the dry aerosol properties".

24638/14-21: It is well-known that the particle shape is highly relevant for the backscattering by aerosol particles (e.g. Mishchenko et al., 1997, doi:10.1029/96JD02110) and consequently also for the lidar ratio. As you mention, volcanic ash particles are nonspherical, thus I'm surprised that you use Mie theory for calculating the lidar ratio.

A: We included the following paragraph into the text: "We performed a sensitivity study of the impact of using mie calculation on non-spherical particles for calculated the Lidar ratio on another date, when depolarizing particles where detected (δ>40%) and photometer measurements were operational (28/06/2011). The Lidar ratio retrieval using the synergy between Lidar and photometer (Raut and Chazette, 2007;Cuesta et al., 2008) was 75 Sr-1. The difference with the Lidar ratio computed with the procedure described in the present paper was not more different than 18%. The same procedure was proceeded on the 11/07/2011 when no depolarizing particles were detected. The difference in the Lidar Ratio was 5 %. The estimates of the Lidar ratio derivate from LIDAR/Sun photometer is expected in the range 15%-25% (Nehrir et al., 2011;Dubovik and King, 2000). Consequently, using a Mie code instead of the most classical method (Lidar/Photometer synergy) is not the main factor of uncertainties. At least the Lidar ratio retrieved with our procedure is close to the Lidar ratio retrieved with the most accurate methods describe previously."

24642/2,3: Why is the asymmetry factor lower in case of volcanic ash aerosols

(large particles) than for other aerosols (small particles)? Above, you mentioned that the asymmetry parameter is mainly a function of the size distribution. But then, your results are in contradiction with Andrews et al. (2006), who showed that the asymmetry parameter increases with increasing particle size.

A: The asymmetry coefficient was calculated from a simple parameterization and might not represent the complexity of this parameter. Since the results are in opposition with the Ångström exponent and as we are less confident in the calculation of g than α, we choose not to publish the g variations in this publication.

24642/22: What is the "1.8μm particle concentration measurement channel"?

A: The sentence is replaced by: " The volume size distribution show a clear maximum at 1.8μm"

24645/3-4: The real part of 1.65 is a bit high for volcanic ash. A comment on this refractive index would be useful. AND 24645/4: Using spherical particles and a refractive index of 1.65+0.005i, it is hardly possible to get a lidar ratio of 52sr at lambda=355nm. Mie shows that the lidar ratio of any single particle in the range from about 0.4μm to 4μm diameter is lower than 50sr. For 0.6μm to 2μm particles, the lidar ratio is on average lower than 5sr. Their backscattering is on average more than 10 times stronger than for particles with lidar ratio 52sr! Only particles around 0.3μm diameter (or larger than 10μm) have a lidar ratio substantially larger than 52sr. The 0.3μm particles must be at least a order of magnitude more relevant for the extinction at lambda=355nm than the larger particles in order to explain a ensemble-average lidar ratio of 52sr at lambda=355nm. Please check whether the 0.3μm particles are really that dominating or whether your Mie calculations are flawed.

A: Our Mie calculation to calculate the Lidar ratio were indeed performed without taking into account the wavelenght dependance of the nephelometer truncature correction. The refractive index has been recalculated following your suggestions. The value of

the refractive index has slightly changed to 1.52+0.008i ±0.03 +0.002i (the variability represents the standard deviation during the main event from 16:00 to 17:30). With a refractive index of 1.52+0.008, the LIDAR ratio of a 2μm particle is 74 Sr-1 and 34 Sr-1 for 0.3 μm, instead of 7 Sr-1 and 13Sr-1 with a refractive index of 1.65+0.005i.

24645/4: If it turns out that 0.3μm particles are really dominating, the applicability of your mass extinction factor is questionable because the factor then critically depends on the presence of these 0.3μm particles. Are they really present in the layer at 3000m a.s.l. where the conversion factor is applied?

A: The mass extinction ratio increase only when the optical properties are the most influenced by large particle (Ångström coefficient <1). that's why we are confident in applying this mass extinction ratio to the layer at 3000 a.s.l.

24645/4: How is the uncertainty of the lidar ratio derived?

A: Again, those are not uncertainties but variabilities calculated as standard deviations on the mean Lidar ratio

24645/8: How is the uncertainty of the extinction derived?

A: idem

24645/9: 461*1.42=655 not 700

A: corrected

24645/9: Would it be possible to estimate the uncertainty of the mass extinction factor and to consider it for the mass concentration? If you can not estimate the uncertainty of the mass extinction factor, this should be mentioned and discussed.

A: We included the following paragraph into the text: "The uncertainties on the mass extinction ratio has been calculated following this equation:

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$$\Delta\eta = 1/(\sigma abs + \sigma sca)*(\Delta M + M*(\Delta\sigma abs + \Delta\sigma sca)/(\sigma abs + \sigma sca))$$

Where $\Delta\eta$ are the uncertainties on the mass extinction ratio, ΔM the error on the mass measurements M given by the TEOM constructor, $\Delta\sigma sca$ and $\Delta\sigma abs$ respectively the uncertainties on the measured scattering and absorption coefficients at 355nm(σsca and σabs). Now the uncertainties calculation are described in the text and are represented on the figure 5."

24645/9-11: A "consistency" check of the extensive properties (extinction, mass concentration) with results for a ash plume at a large spatial and temporal distance (over one month!) might be a bit far-fetched.

A: "consistent" has been replaced by : same order of magnitude

24645/14-15: How was the mass extinction factor derived here? It is lower than used at 3000m a.s.l. On what information is the height dependence of the mass extinction factor based? This should be explained.

A: We did not applied any altitude dependance on mass extinction ratio. We are aware that the in situ volcanic ashes detected at the PdD is an approximation of the ashes located at 3000m. This is now better stressed in the paper. We still believe that it is good approximation considering the mass extinction ratio found into the litterature

We consider that at 3000m a.s.l. the mass extinction factor is mainly influence by the volcanic particles, whereas in the planetary boundary layer different type of particles are mixed. Consequently, ηvolcano was used at 3000m a.s.l and the "real time" mass extinction ratio was use in the planetary boundary layer

24645/15: Why is the relative uncertainty of the mass concentration higher than for the extinction, while it was about the same for mass and extinction in line 8-9?

A: Again, we choose to show the standard deviation and not the relative uncertainties.

Indeed, the relative uncertainty for the mass concentration (given in section 2.5) is constant, whereas the standard deviation can bring more information

24645/15-24646/3: In Section "instrumentation and modelling" it was not clearly described what aerosol parameters were measured or modeled as "dry" or "wet". This should be elaborated in more detail. Otherwise the results and discussions are not easy to follow.

A: Comment added in section 2.1: "All the in-situ instruments presented in this paper measure the dry aerosol properties".

24650/23-26: Miffre et al. assumed a diameter of 10μm, which is 5 times larger than the 2μm found in your study. This difference in particle size would result in about 5 times higher mass concentrations in Miffre et al. than in your study (if all other parameters are the same). Thus, the diameter of 10μm from Miffre et al. does not explain the differences, rather it makes the difference of mass retrievals less understandable. Please try to explain the difference more consistently.

A: The difference between the mass concentration calculated by Miffre et al.; and the one found in this study is mainly induced by a different LIDAR backscatter measured: the LIDAR ratio and the mass extinction ratio are nearly equal (55 against 52 Sr-1 and 1.44 against 1.57g.m-2). This backscatter difference might be simply due to the inhomogeneity of the volcanic plume

Fig. 4: There is a sharp decrease of the volume distribution between 2.5μm and 3.5μm. Is this realistic or an instrument effect?
A: Indeed, the OPC often show a sharp decrease between 2.5μm and 3.5μm. This decrease might be accentuated by the wiggled and non-monotonic shape of the scattering cross section function at these diameters (Bukowiecki et al., 2011). However Bukowiecki et al. show that for a reasonable range of refractive index, the volume concentration can change, but not the diameter of the maximum concentration. Moreover, the OPC has already detected particle with a modal diameter $\geq$ 3.5μm.

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Technical corrections:

A: All the technical corrections were corrected