

## ***Interactive comment on “Multi-wavelength Raman lidar observations of the Eyjafjallajökull volcanic cloud over Potenza, Southern Italy” by L. Mona et al.***

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The paper has been reviewed following the precious referees comments. On behalf of all authors, I would like to thank the referees for the comments and suggestions, that contribute to improve the quality of our paper.

In the following, the referees comments are repeated first and then our answer to each comment is reported. When necessary the modified paper text is reported too. The revised paper is attached as supplement.

Anonymous Referee #1

C10026

The paper presents aerosol lidar observations taken during the eruption phase of the Eyjafjallajökull volcano in Southern Italy. The paper is interesting to read because it gives valuable insights in the properties of the volcanic ash cloud in Southern Europe. In contrast to Central Europe, this region was not heavily affected by the ash cloud during the first eruption phase in April 2010. Consequently the observed ash particles travelled a long way and can be described as aged aerosol. The use of an advanced Raman lidar technique allows for a distinction between Saharan dust particles and volcanic ash, which were observed in similar heights on the same days. The paper presents good science but it needs to be rewritten at several places because some descriptions are too detailed and the style is too narrative. I recommend publication after the necessary changes have been applied

Specific major comments

page 12764, line 6/7: The difference between ‘multi-wavelength Raman lidar measurements’ and ‘EARLINET measurements’ is not clear for the reader. You may omit this.

The authors would like here to underline that the methodology for the volcanic layer identification is of course based on the multi-wavelength Raman lidar measurements performed during the volcanic event, but it is essential the contribution provided by multi-year measurements performed at CIAO. The sentence has been rephrased.

A methodology for volcanic layer identification and accurate aerosol typing from the multi-wavelength Raman lidar measurements has been developed taking advantage from the long-term lidar measurements performed at CIAO since 2000.

page 12764, line 14: ‘In the April-May period ...’ You may omit this sentence. It is not important here.

OK

page 12764, line 22-28: This is too detailed. Try to describe it in a short way in the abstract.

C10027

The sentence has been rewritten as follows: The study of these intensive parameters indicate the presence of volcanic sulfates/continental mixed aerosol in the volcanic aerosol layers observed at CIAO. Differences observed in correspondence of the two maxima in the volcanic aerosol load indicate the presence, besides sulfates aerosols, of some aged ash.

page 12765, line 2-8: This is a typical sentence that makes the paper difficult to read. It is too long and therefore difficult to understand.

The sentence has been rewritten as follows:

This medium-sized eruption (Petersen, 2010) caused an enormous disruption to air traffic across western and northern Europe, because it injected ash directly into the Jet Stream and from there in the northern Europe free troposphere.

page 12765, line 18-25: This sentence is too long

The sentence has been rewritten as follows: Almost the whole European continent was affected by the arrival of the volcanic cloud. Volcanic particles were observed in UK, Germany and France from very low altitude up to the upper troposphere for almost the whole 2010 Eyjafjallajökull eruptive period (Pappalardo et al., 2010a; Emeis et al., 2010; Flentje et al., 2010; Schumann et al., 2011). The cloud reached Italy and Greece starting from 19-20 April, after passing the Alps (Pappalardo et al., 2010a). In May 2010, the volcanic cloud was transported over the Iberian Peninsula moving then towards East, reaching again Italy and Greece (Pappalardo et al., 2010a).

page 12766, line 7/8: Matthias et al. present reconstructions, too, but they use a different type of model (Eulerian model) than Stohl et al. (Lagrange model).

Yes these are different models. Here we underline that observational data can be used for the evaluation of different kind of models lagrangian or eulerian. However, we re-wrote the sentence in the revised version of the paper on the base of the other reviewers comments (see following answer).

C10028

page 12766, line 8: What is meant by 'border conditions'?

Our observations are typically related to very long (in time and space) path of the volcanic cloud. This leads to small quantity of observed volcanic particles. On the other hand, the observational site is far away from the source. These 2 aspects make our observations interesting for evaluating the models capability at the extremes of their operability, i.e. for low aerosol concentration and at far distances from the volcano.

In order to make it more clear and avoid confusion, we rewrote the sentence:

Firstly, the large distance from the volcano and the low amount of aerosols reaching this area make the observations of the volcanic cloud in Mediterranean region useful and necessary for the evaluation of different models (e.g. Matthias et al., 2011; Stohl et al., 2011) at the extremes of their operability, i.e. for low aerosol concentration and at far distances from the emitting source.

page 12766, line 13-16: How could this affect the Mediterranean eco-system? I do not immediately see which effects this might be.

The effects of aerosol deposition on the sea are indeed not well understood. Atmospheric aerosol deposition can be an important source of nutrients and trace metals to the open ocean that can enhance sea productivity and carbon sequestration and thus influence atmospheric carbon dioxide concentrations and climate. However the response depends on specific components in aerosols. Aerosol additions enhance growth by releasing nitrogen and phosphorus, but not all aerosols stimulated growth. Toxic effects were observed for some aerosols (Paytan et al., PNAS,2009). There is actually poor knowledge of the effects of volcanic particles on sea. Kockum et al., Chemical Geology 2006 reports that "owing to the mixing of acid ash leachate with fresh water, aluminofluoride complexes persist in aqueous systems with low turnover rates, and could be toxic to both plants and animals." and "In addition, the deposition of volcanic aerosols may enhance biological activity in the marine environment by providing micronutrients.

C10029

page 12767, line 21-24: The 3+2 measurements include the 1064 nm backscatter don't they? How can they then be used to retrieve the 1064 nm backscatter? Additionally, you need to explain what is meant with 3 backscatter and 2 extinction measurements. It is not clear that they are taken at different wavelengths.

In order to make it more clear and avoid confusion, we rewrote the paragraph: This allows us to measure directly the lidar ratio (extinction to backscatter ratio) vertical profile both at 355 and 532 nm. Additionally, the aerosol backscatter at 1064 nm is retrieved through an iterative procedure (Di Girolamo et al., 1999), with a lidar ratio profile selected on the basis of the lidar ratio profiles measured at 355 and 532 nm. Summarizing, aerosol backscatter coefficient profiles at 3 wavelengths (355, 532 and 1064 nm) and extinction profiles at 2 wavelengths (355 and 532 nm) are simultaneously measured at CIAO. This ensemble of measurements will be referred as 3+2 measurements in the following.

page 12768, line 1-9: This paragraph can be omitted, it is not necessary to understand the paper.

OK removed

page 12768, line 19 - page 12769, line 20: This is described in too much detail. It is not important why you couldn't perform measurements in this or the other case. It is sufficient to describe what you have.

Lidar measurements were not possible for low clouds or rain. The presence of rain instead of low cloud could affect the deposition of the particles to the ground. In addition washing out could be referred to as a background condition before the arrival of other materials. In this sense distinguishing between low clouds or rain could be interesting. However it is true that both these aspects are out of the aims of this paper, therefore the paragraph has been shortened in the revised paper:

From 15 April, when the first alert was sent, lidar measurements were performed at

C10030

CIAO whenever the absence of low clouds and rain permits them. During 19-22 April period the arrival of volcanic ash over Northern and Central Europe and, after that, a feeble transport of ash beyond the Alps were forecast. In 25-30 April period, desert dust arrived over Southern Europe followed by a change in the wind direction with air masses coming from North-Eastern Europe, potentially transporting material emitted by the Eyjafjallajökull volcano over Western Europe and then over Italy and Greece. This situation lasted for the following days, when Saharan dust intrusions over Southern Europe also occurred. A possible arrival of volcanic cloud over Northern Italy was forecast for 8 May. Accordingly, lidar measurements were performed from 8 May, 20:00 UTC till 11 May, 02:00 UTC. CIAO ran lidar measurements from 12 May, 12:00 UTC, till 15 May, 01:00 UTC, when a shower forced a sudden stop. The last measurements performed for the Eyjafjallajökull volcano eruption started on 18 May, 06:00 UTC, and continued until 19 May, 11:00 UTC.

page 12769, line 21: In this Methodology section, some of the results are already explained and this leads to some confusions with the Results section. In particular fig. 6 is doubled in fig. 7. You should avoid this, shorten the Methodology section to what is really needed to explain the methods and then give the details in the Results section.

One of the main result of the paper is the developed methodology (as reported in the abstract and in the summary section) and a detailed discussion of it is essential for the reliability of the other results (mask and volcanic layers optical properties) reported in the last part of the paper. Each one of the treated points (layering identification – clouds identification and aerosol typing) is a critical issue. As reported in the first part of this section, there are different automated methods, such as that used for the CALIPSO retrieval, based on modeled aerosol properties that rely on the idea that the whole range of possibilities in terms of optical properties had already been measured and characterized for each aerosol class. Of course this is not the case in particular for tropospheric volcanic aerosols. Indeed, a detailed analysis is needed both for the identification and typing of aerosol layers and the investigation of aerosol mixing

C10031

processes. Therefore it is here important to explain the developed methodology into needed details.

However, this section has been shortened in the revised version in order to make the paper more readable. In particular: - introducing methodology part was shortened and included in the general introduction - the whole section in general has been revised to shorten it - the description of 13-14 May situation has been shortened and moved to Results section - the figures 3 and 4 have been removed - conclusive part of the aerosol typing section was moved to the results section following the reviewer suggestion - Fig 6 was removed to avoid duplication

page 12771, line 24/25: This feeble layer is almost not visible in Fig. 1. Besides that, this paragraph is misplaced in the Methodology section.

The figure contrast was modified in order to make the feeble layer more evident. As reported above, the discussion of how quantitative aerosol masking is obtained from quicklook available almost in near real time during the volcanic event (like figure1) is an essential part of the paper. On the other side, no lidar experts are used to consult these quicklook images (Level 0 uncalibrated data) since they were provided and widely distributed already few hours after the measurements themselves. It is therefore essential explaining which kind of information (mainly qualitative) can be obtained from Level 0 data and what instead is more quantitative and assessed (also in terms of uncertainties) like aerosol masking reported in the following.

page 12772, line 9-15: I do not see the argument why it is better to use backscatter profiles instead of the range corrected signals. The normalization has to be done, too.

In principle, it is true, the backscatter profiles, as the range corrected signals, need for a calibration procedure, but there is a big difference between the 2 quantities. RCS also after a normalization is still a system dependent quantity expressed in arbitrary units. The aerosol backscatter profile instead is system independent and more important it has a physical meaning in absolute sense. This makes this methodology exportable

C10032

in absolute to other lidar measurements, like for example all EARLINET lidar observations performed during Eyjafjallajökull volcanic eruption. In addition, EARLINET quality assurance program guarantees for the high quality of the backscatter profile retrieval procedures. Finally, the availability at the observational site of simultaneous backscatter profiles at 355 and 532 nm retrieved by elastic Raman technique (self calibrated technique) and the long term database of the aerosol backscatter profiles at 355-532 and 1064 nm furthermore reduce uncertainty related to the calibration of elastically retrieved backscatter profile. As reported in the following, besides the backscatter gradient, statistical error on aerosol backscatter and the scattering ratio are used as parameters for identifying particle layers. In this context the use of RCS would lead to an underestimation of the error and to not quality assured quantity.

page 12772, line 16: 'an ill-posed procedure'. What does it mean? Is it of importance here?

This is a very technical details and therefore not strongly needed here. We removed it: However, since the derivative is highly sensitive to fluctuations, a smoothing procedure is typically needed.

page 12772, line 24/25: '... evaluated by using the Rayleigh criterion ...' This is too specific and cannot be understood even if the reference is given.

OK removed. The whole paragraph was shortened:

A second-order Savitsky-Golay filter is applied on the differential, because of its effectiveness in preserving vertical structures (Pappalardo et al., 2004). The number of points is progressively increased as the signal noise increases, with 1000 m as fixed maximum of the effective vertical resolution (Pappalardo et al., 2004).

page 12773, line 25-27: What is meant here if you say the 'models run every 6 hour'? The models run continuously with a time step of a few minutes over the whole period, concentration fields are stored every hour.

C10033

The sentence has been revised in the new version:

A temporal average of 1 hour is chosen in order to be able to draw a direct comparison with models that typically provide data every hour (e.g. Matthias et al., 2011).

page 12774, line 1-3: 'The iterative procedure reported in Di Girolamo et al. (1999) is applied for the 1064nm backscatter retrieval, with lidar ratio values selected on the basis of the 3+2 measurements performed at CIAO.' This has been said before.

OK removed.

page 12774, line 8-19: From Figure 2 it is difficult to believe that you can distinguish different layers above 7 km. To me this looks too noisy to be sure about these layers.

By-eye layer detection typically works well, but it is not objective and it can fail for not well defined and/or feeble layers. For example for the case under investigation, the layers above 7 km cannot be distinguished by-eye. Here an objective criterion has been established for identifying if a gradient corresponds or not to a layer: statistical error on the final product, i.e. the aerosol backscatter coefficient, lower than 30%. For higher statistical error (however lower than 50%), layers are identified as regions where the scattering ratio values is higher than climatological threshold plus the statistical error itself. As reported in the paper, the identification is performed on each individual backscatter profile, but a final check on the resulting layering temporal evolution allows us to disregard false layer identification. The layers above 7 km reported in Figure 2 are identified through gradients and scattering ratio criteria. The statistical error is lower than 30% for the lowest of the 3 layers and around 40% for the others. In addition the further check of the temporal evolution of the layers indicates that these signatures are present not only at this hour but also at the following ones. Therefore these are objectively aerosol layers distinguishable from the measured backscatter profiles. In order to make more clear that only the applied methodology allows the identification of these small thin layers (not distinguishable by looking the profile plot) we rewrite the sentence:

C10034

At upper levels, the applied methodology allows the identification of thin and sparse layers as exceeding the threshold on the scattering ratio. This is an indication of the presence of a low amount of aerosol at these altitudes. The further check of the temporal evolution of the layers indicates that these signatures are present not only at this time but also at the following ones. All these elements allow for an objective identification of these features as aerosol layers distinguishable from the measured backscatter profiles. At altitude higher than 12 km a.s.l., longer integration time, or a time series analysis, could allow us to better describe the upper level particle layers.

page 12774, line 20: Couldn't you also use the depolarization measurements to distinguish clouds from aerosols?

Particle depolarization is strictly related to the shape of the particles, so that in general it assumes high values within ice clouds and very low values in water clouds. Aerosol depolarization ratio is high however also for large irregular aerosols like dust particles. Also large volcanic ash particles are expected to have high particle linear depolarization ratio. Therefore even if generally important parameter for aerosol/cloud discrimination, the depolarization does not provide an unambiguous aerosol/cloud discrimination. However, the cloud identification treatment is here twofold: the cirrus cloud identification and the removal of low (mixed and water) cloud from the backscatter profile retrieval. The identification of cirrus cloud is typically performed after the backscatter profile retrieval and therefore the depolarization was also used, if available. This has been added in the paper:

Cirrus clouds are identified mainly on the basis both of their temporal dynamical evolution (Mona et al., 2007), the high particle linear depolarization ratio and the almost neutral backscatter spectral dependence, due to the large size of hydrometeors.

page 12776-12780: this paragraph needs to be shortened drastically. You describe in very much detail, what has been observed. This is not necessary to explain the method. You may do it later in the Results section.

C10035

As reported above, the developed methodology is a itself result of the paper. In the revised version of the paper this section is written in such a way that readers interested only in the resulting mask and optical properties could also skip it, while who is interested in the method itself would find in this section all necessary information, but avoiding too much details.

page 12775/76: You describe three different trajectory products but you use only one. It is not important in this context what is available besides the product you use here.

All three trajectory products were used: HYSPLIT choosing appropriate time and levels and DWD and NASA/Goddard backtrajectories, as reported in the paper, as additional check.

page 12778, line 23-26: Why can you be sure that volcanic ash has different properties than Saharan dust?

It is not sure at all. Different aerosol types could have also similar optical properties. Volcanic particle and Saharan dust are two types of aerosol that indeed contain a large variety of particles for dimension, shape and composition. As a whole however it can be assert that volcanic particle are more absorbing than Saharan dust; ash and dust are typically large in dimension, while sulfate coated particles are smaller. Overlap for some optical properties could be observed. Therefore only a complete characterization of the aerosol optical properties, in conjunction with all ancillary information available, can provide a reliable typing of the detected aerosol. In particular the temporal behavior of optical parameters concentration independent, like Angstrom exponent, is effective for identifying the changing of optical properties in a layer, or the arrival of a layer with different properties.

In the specific reported case (13 May around 05:00 UTC) we found: - uncertain situation with presence of dust and/or volcanic cloud (backtrajectory analysis) - typical value obtained in 10 year of measurement of Saharan dust for an identified layer (in agreement with backtraj. anal.) - a sudden change in the Angstrom values indicating

C10036

the arrival of particles with different properties - the backtrajectories indicate the arrival also of volcanic cloud, so this change is due to the arrival of them over a Saharan substrate

In order to make it more clear the text has been revised:

For these layers the mean  $\text{Å}^{-1}$  value is 0.2. The significant change in the Angstrom exponent indicates the arrival of particles with different properties. According to the air mass backtrajectories these altitudes are likely affected by the arrival of volcanic cloud. This indicates a mixing between dust and volcanic particles.

page 12778, line 11-12: 'The feeble feature extending between 3.4 and 4.3 km a.s.l. is characterized by ...' I cannot see a value for the Angstrom exponent at 6:00 UT in Fig.5.

There was a mistake in the plot. It was corrected in the revised figure.

page 12779, line 24/25: If you classify mixed aerosols it would be of interest to know which types are mixed. How many types in total can you distinguish in your classification? Are there other types which were not observed during these days?

Multiwavelength Raman measurements plus additional information provided by backtrajectories analysis and/or models and satellite images allows for a detailed characterization of the aerosol typing. There are many papers in literature reporting EARLINET experience in this. Just as example, cases with Saharan dust mixed with maritime aerosol are identified in Mona et al., 2006. As reported in the current manuscript the methodology here described was developed starting from this multi-year experience gained within EARLINET. For the current paper, different distinct sources are considered: volcano eruption, Saharan dust storms, forest fires, continental pollution. In addition it is considered that within the PBL aerosol produced by very local mechanisms are present eventually together with long range transported aerosol arriving at low altitudes or intruding the PBL from upper altitude levels. All possible mixings among

C10037

these types of aerosol were taken into account. For the area and period under investigation the following mixings were observed: local + dust, local + volcanic, and, dust + volcanic. Description of observed mixing is reported in 4.1 section. The following sentence regarding the mixed situation was added in the revised version of the paper.

All possible mixings among these types of aerosol were taken into account.

page 12781, line 3: What is SEM analysis?

OK explicitly written now in the paper. Scanning Electron Microscopy

page 12781, line 14-18: It is not of interest here what other obligations you have.

OK removed.

page 12782, line 21-25: Wouldn't you expect that the volcanic ash is not hygroscopic? Eventually formed sulfate particles may be connected with the ash and change the hygroscopicity, but do you have any indication that this was the case? Additionally, most particles, even hygroscopic ones, will not be largely affected by water uptake at 50 % RH.

Yes, volcanic ash is expected to be not hygroscopic, and sulfate particles, that could be a result of the ash modification during the transport over Europe, are sensitive to relative humidity conditions. The study of the optical properties observed in the volcanic layers, reported in section 4.2, together with co-located RH measurements permits to identified 2 different behaviors probably related to cases with the presence of volcanic sulfates/continental mixed aerosol and cases in which, besides sulfates aerosols, there was some ash affected by the aging through the European continent. In particular, Angstrom exponent and lidar ratio are investigated in terms of RH dependence. Different studies, experimental and theoretical, demonstrate that these quantities are highly sensitive to RH also for values lower than 50% (e.g. Ackermann, Jtech 1998; Ferrare et al., JGR, 2001, Muller et al., JGR 2007).

page 12784, line 14-15: '... while Ångström exponents are smaller': How small? Small  
C10038

compared to what?

The sentence has been revised: Lidar ratio values are in between those observed in correspondence of the first arrival on 20 April and for after 21 April, while Ångström exponents are smaller than values typically observed in the previous days and a mean particle linear depolarization ratio of 16%, similar to 20 April case, is observed.

page 12786, line 1-3: This sentence is not well formulated. The emissions cannot be directly observed at Potenza. If you observe high concentrations this does not necessarily mean that the emissions were high because only a small fraction reaches the measurement station. You may reformulate this.

OK the text has been revised:

A lidar ratio of about 40 sr at 355nm increasing with the relative humidity up to 60–70 sr, and a ratio of lidar ratios of about 0.8 was observed at CIAO on 19–20 April and 13 May 2010, dates corresponding to larger amount of aerosol emitted by the Icelandic volcano with respect to the other days under investigation.

page 12786 line 3 and later: How can I interpret the ratio of lidar ratios? It would be good if you would say something about this quantity, what it is and how it can be used to characterize aerosol particles.

The investigation of this parameter is relatively new. As reported in the paper, observations performed in different regions and for different aerosol types shows that the ratio of lidar ratio at 2 wavelengths is particular effective for the typing (Muller et al., 2007). General comment on this quantity is reported at pag 12785 lines 7-10 and specific comments about values assumed by this quantity are discussed at page 12786 line 8-10, 15-17.

page 12787 line 12: This paper of Pappalardo et al. may be cited if it is submitted and may be included in the final version of the paper as accepted. Otherwise you should omit this reference.

OK, we removed it for the moment.

page 12787 line 18-23: How do your observations compare to other lidar and sun photometer observations of the Eyjafjallajökull ash cloud?

In the revised paper we added few lines about the comparison with AOD values obtained for this volcanic eruption from lidar and AERONET measurements. Previously reported comments (lines 18-23) have been removed. The current text is reported in the following:

These values are significantly lower than the peak values up to 0.7 at 532 nm observed over Germany in the volcanic layer during this event (Ansmann et al., 2010) and the moderate columnar AOD around 0.3-0.4 and 0.5 observed over Iberian peninsula (5-11 May) and Cabauw (17-21 May), respectively, for almost direct transport (Toledano et al., 2011; Ansmann et al., 2011). The low values observed at CIAO are related to the larger distance from the Eyjafjallajökull volcano and to the dispersion of the volcanic cloud during its path across Europe.

page 12787 line 28 - page 12788 line 4: This is again one of the sentences that is quite hard to get. You may rearrange it and explain a bit more clearly what is meant.

OK we re-wrote the sentence in the revised version:

Typically high  $S_{uv}$ , particle linear depolarization ratio increasing with RH and values of the ratio of lidar ratios greater than 1 are measured in the volcanic aerosol layers at CIAO. These values suggest the presence of volcanic sulfates/continental mixed aerosol. Different intensive aerosol optical properties are measured at CIAO in correspondence of the maxima in the observed volcanic aerosol: lidar ratio increasing with RH (from 40 to 70 sr for RH from 20 to 70%) and ratio of lidar ratio values below 1. These values indicate the presence, besides sulfates aerosols, of some ash affected by the aging through the European continent.

Figure 2: How are the 'layers' above 7 km distinguished? Can you really identify differ-

C10040

ent layers?

See answer reported above

Figure 4: What is seen in Fig a,b,c,d? Better start with 'HYSPLIT backtrajectory analysis during the transition regime between Saharan dust observation and the volcanic aerosol arrival on 13 May 2010.'

The figure was removed in the shortened Section3.

Figure 6: The maximum investigated altitude is not visible for many hours: why? If you classify 'mixed aerosols', could this be specified a bit more? Can you show which types are mixed, e.g. Saharan dust and volcanic aerosols?

The maximum investigated altitude for this case is typically around 21 km. Figures report the aerosol masks up to 10 km, because we are here focused on tropospheric aerosol. However the 50% threshold fixed for the applied methodology is typically reached for the period under investigation around 10km. Mixed aerosol types are discussed in the paper (see also answer reported above). In addition, in the revised version of mask figures, mixed layers are reported with different colors depending on the mixed aerosol type.

Figure 7: This is not a nice figure, it is too small, it looks not well arranged and it includes a figure that was already shown (fig. 6). You might think about having a separate figure for each episode and/or putting some figures in an appendix.

Figure 6 has been removed. Figure 7 instead is now split into 2 separate figures so that each mask is now more readable.

Figure 8: You might use different colors for the different days/heights to facilitate a comparison of the different aerosol properties for one observation. The labels are too small and difficult to read in this figure.

The figure has been re-edited following reviewer suggestion.

C10041

Minor comments / expressions which are difficult to understand

page 12764, line 15/16: doubling of observed'

OK

page 12764, line 20: are discussed

OK

page 12765, line 12: omit 'has been'

OK

page 12765, line 21: Emeis et al., 2011

OK

page 12765, line 28: Gasteiger et al., 2011

OK

page 12765, line 9/10: doubling of occurrence/occurring

OK

page 12766, line 21: state-of-the-art

OK

page 12766, line 21: for ground based

OK

page 12766, line 14: studies

OK

page 12766, line 26: of the PEARL set-up and the retrieved products

OK

C10042

page 12769, line 22/25: the term 'in order to' is used quite frequently, maybe you can try to avoid it at some places or find something else

OK

page 12770, line 9: 'for the provision' or 'for providing'

The sentence has been re-written in the revised paper:

This kind of typing algorithms aims to provide reliable results in near-real time

page 12771, line 19: 'seems to occur' this is not a good expression for a scientific text. You have to be more precise: Did it occur? Could it be seen? If yes, write it down clearly.

OK

page 12771, line 19: 'Measurements started ...' The sentence may be omitted.

OK

page 12772, line 9: With respect to ... . Please check this through the whole document.

OK

page 12772, line 21: see my comment page 12771, line 19

OK

page 12773, line 4/5: 'the aerosol backscatter scattering ratio' This not a nice term. Couldn't you name it 'scattering ratio' and explain what it is.

OK

page 12780, line 14: Which models?

Models used for identifying the potential sources are reported in the methodology (typing) section.

C10043

page 12783, line 5/6: 'As reported above, the measurements stopped on 23 - 24 April due to rain.' This is not important.

Here the information that it was rained can be important for understanding that particles previously transported were removed from the atmosphere. The sentence has been re-written in this sense:

On 23-24 April, it was rained for almost all the day and on 25 April a strong dust event was observed.

page 12785, line 1: better: 'Optical properties of volcanic aerosol'

OK

page 12785, line 2: You may list the intensive properties that you can determine and that are investigated here.

Changed in the revised version:

The dependence of intensive properties retrieved by lidar (backscatter-related Ångström exponent at 532/1064 nm, extinction and backscatter-related Ångström exponents at 355/532 nm, lidar ratio at 355 and 532 nm, and linear particle depolarization ratio) as a function of the relative humidity measured by the co-located microwave radiometer is investigated (Fig. 6). In particular, backscatter-related Ångström exponent at 532/1064 nm,  $\alpha_{\text{Å}}$  (Fig.6a) and lidar ratio at 355 nm,  $S_{\text{uv}}$  (Fig. 6c) are preferred to Ångström exponent at 532/355 nm and lidar ratio at 532 nm, respectively, because of the larger availability of these data.

page 12785, line 16: You need to introduce  $S_{\text{uv}}$  and later  $S_{\text{vis}}$

OK

page 12786, line 14: 'For all the other cases ...' this sounds like a lot, but aren't there only 2 other cases?

C10044

OK

page 12786, line 18-23: The sentence is too long, you may rearrange it in two or three separate sentences. '... no simultaneous Raman lidar ...' is not a good expression.

The sentence was changed accordingly to referee comment:

At this stage the aerosol size distribution for the cases reported in Table 1 and Fig. 6 cannot be appropriately investigated on the basis of co-located AERONET measurements because only few AERONET data are available for the presence of clouds. Moreover, the Raman lidar (night-time) and AERONET (diurnal) measurements are not simultaneous, and the observed high variability in the aerosol content does not permit to use AERONET inversion for furthermore investigating the aerosol layers identified through the lidar measurements.

page 12787, line 4/5: better: These measurements can be a reference point for the testing of atmospheric transport models. The observations are taken far from the source and the amount of volcanic aerosol reaching the area is low.

OK

page 12787, line 8: in detail

OK

Figure 4: 'highly noisy' is colloquial. Better describe it with 'large statistical errors'. What about error bars between 1 and 4 km?

Modified in the new version:

Mean values are reported as squares for backscatter related Ångström exponent at altitude levels where statistical errors is larger than 30%.

Anonymous Referee #2

This is an interesting paper about the observations of volcanic ash made after the

C10045

eruption of the Eyjafjallajökull. Also, a method is described how to present the data such that the evolution of the plume over one location can be followed by the creation of masks. A comment by another reviewer on the discussion paper is already published on the web, which I have read. I agree fully with the comments made by this reviewer. There are a few additional comments that I should like to make, which may sometimes overlap (partially) with the comments already published

#### General comments

The text should be shortened by removing parts that provide unnecessary details. In particular:

- pp 12768. The description of MUSA is superfluous, as no data is used. Probably the system should not be mentioned at all, as it is irrelevant to the content of the paper.

OK removed

- Pp 12775. Sec 3.3. Aerosol typing. In my opinion the lengthy description on pp 12776-12779 are actually not part of the methodology, but belong to the results section. Also, the text is too extensive, so I should suggest shortening it, by concentrating on the methodology, without going too much in detail about specific cases. Move actual results to the next section.

One of the main result of the paper is the developed methodology (as reported in the abstract and in the summary section) and a detailed discussion of it is essential for the reliability of the other results (mask and volcanic layers optical properties) reported in the last part of the paper. Keeping in mind the relevance of the developed methodology, this section has been shortened in the revised version taken into account the reviewer's comments. In particular: - introducing methodology part was shortened and included in the general introduction - the whole section in general has been revised to shorten it - the description of 13-14 May situation has been shortened and moved to Results section - the figures 3 and 4 have been removed - conclusive part of the aerosol typing

C10046

section was moved to the results section following the reviewer suggestion - Fig 6 was removed to avoid duplication

- Pp 12785. Sec 4.2 I find the statement that the particle linear depolarization ratio decreases with increasing relative humidity counterintuitive. Unfortunately, I do not have a copy of the paper by Sakai et al.. My reasoning would be that humidity would tend to smoothen irregularly shaped particles, thereby achieving the opposite: lower depol with higher humidity. This is also mentioned in the abstract.

It is for sure that increasing RH the depolarization ratio of a particle decreases. The reviewer is right, the sentence is misleading. What authors meant here is that it can happen to observe situation in which, even in absence of dust particles, both RH and depolarization ratio are high. This kind of situation can be related to the presence of sulfates (Sakai et al.,) The paper Sakai et al., reports humidity, backscatter and depolarization ratio measured by Raman lidar at Nagoya in 1994-1997. A total of 332 tropospheric profiles were considered. They found that in some conditions high depolarization ratio in correspondence of high relative humidity. In that paper a detailed discussion on depolarization ratio behavior with RH as a function of aerosol chemical compositions is reported. Their discussion is based on laboratory and experimental field measurements reported in literature. They discussed their result about the observation of high depolarization ratio in correspondence of high RH and found that this could be related to the sulfate presence. This is mainly based on the following aerosol classification (reported in Sakai): - Water-insoluble particles like mineral dust, with high depolarization independently of RH - Water-soluble droplets characterized by almost zero depolarization ratio independently of RH - Water-soluble deliquescent particles (eg NaCl, (NH<sub>4</sub>)SO<sub>4</sub>) : they exist as solution droplets (low depolarization) above the deliquescence point (DRH), and as crystals (high depolarization) below the efflorescence point (CRH). Between DRH and CRH, the phase (and therefore the depolarization ratio) depends on the RH history.

Based on Sakai results we can assume that our observation of a high particle linear

C10047

depolarization ratio for high RH could indicate the presence of sulfate aerosols for the considered period.

The sentence has been revised:

The particle linear depolarization ratio shows higher values in correspondence of higher RH, that could indicate the presence of sulfate aerosols for the whole period (Sakai et al., 2000).

Also the final part of the abstract has been modified, because the sentence reported in the previous version was misleading:

The study of these intensive parameters indicate the presence of volcanic sulfates/continental mixed aerosol in the volcanic aerosol layers observed at CIAO. Differences observed in correspondence of the two maxima in the volcanic aerosol load indicate the presence, besides sulfates aerosols, of some ash affected by the aging through the European continent.

Figures should be changed for proper readability. In particular:

- Fig 3. Trajectory plots are too small to read the height scale.

This plot has been removed in the revised version.

- Fig 4. Trajectory plots are too small to read the height scale.

The figure has been removed in the revised version of the paper to shorten the section on Methodology

- Fig 6. Legend is not readable

The figure has been removed in the revised version of the paper to follow Reviewer 1 suggestions

- Fig 7. Annotation of axes for subfigures not readable. Legend too small.

The figure has been completely re-edited in the revised version of the paper.

C10048

- Fig 8. All panels are too small.

The figure has been completely re-edited in the revised version of the paper.

Comments to the text

The text needs a fair bit of editing and error corrections. Below is a list of examples.

- pp 12764 - line 5. The statement "both of the multi-wavelength Raman lidar measurements and EARLINET measurements performed" is unclear. EARLINET promotes the Raman measurements. Therefore I assume the contrast between Raman and elastic measurements is meant?

Authors would mean that multi-wavelength Raman measurements performed during the event are used, but the experience and climatological values obtained from our measurements performed since May 2000 within EARLINET are an important added values at our volcanic related measurements. To make it more clear, the sentence has been modified in the revised version of the paper:

A methodology for volcanic layer identification and accurate aerosol typing from the multi-wavelength Raman lidar measurements has been developed taking advantage from the long-term lidar measurements performed at CIAO since 2000.

- Pp 12765 - line 1. Replace "a small Iceland"s ice cap" by "a small volcano under Iceland"s ice cap"

OK

- Pp 12765 - line 8. Replace "airspace" by "airspace"

Removed in the revised version.

- Pp 12765 - line 12. Replace "has been" by "has"

OK

- Pp 12765 - line 15. Replace "have been" by "were"

C10049

OK

- Pp 12765 - line 15. Replace "accordingly" by "according"

OK

- Pp 12770 - line 3. Replace "warm colors" by "yellow, orange and red"

This info has been added.

- Pp 12770 - line 9. Replace "This kind of typing algorithms is highly performing for the providing of typically reliable results in near-real time" by "This kind of typing algorithms aim to provide reliable results in near-real time"

OK

- Pp 12770 - line 14. Replace "On the contrary" by "In contrast"

OK

- Pp 12771 - line 16. Replace "atmosphere" by "atmospheric"

OK

- Pp 12771 - line 18. Replace "May morning" by "May in the morning"

OK

- Pp 12771 - line 24. Replace "feeble" by "tenuous"

OK

- Pp 12771 - line 25. Replace "falling down" by "descending"

OK

- Pp 12771 - line 26. Replace "intense" by "dense"

OK

C10050

- Pp 12772 - line 16. What do you mean by "ill-posed"? Presumably, ambiguous results are expected, however, this is not the same as what is usually meant in mathematical terms.

This is a very technical details and therefore not strongly needed here. Following the reviewer1's suggestion we removed this detail.

Please also note the supplement to this comment:

<http://www.atmos-chem-phys-discuss.net/11/C10026/2011/acpd-11-C10026-2011-supplement.pdf>

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Interactive comment on Atmos. Chem. Phys. Discuss., 11, 12763, 2011.

C10051