

The authors would like to acknowledge the helpful comments provided by the 2nd referee.

Reply to Referee 2

General comment.

The authors analyze in this paper certain properties of aged volcanic particles observed over Athens, Greece using Raman lidar and sunphotometer measurements. The paper presents very interesting results which are based on high quality data and state of the art algorithms. The results presented provide additional useful information to our knowledge for aged volcanic ash, considering the large distance between the Icelandic volcano and Athens. The paper however would benefit with a better focus and a better structure. It is not very clear what the main objective of the paper is. Do the authors aim to a validation paper for FLEXPART or they use FLEXPART as a tool for the interpretation of their results? Certain parts of the manuscript suggest a validation exercise, while other parts try to use the model for the interpretation of the measurements. To my opinion the authors should avoid the validation character of the paper and focus only on the interpretation, which in few cases could also be clearer, based on less assumptions. In general I suggest that the paper should be accepted to ACP considering my comments for revisions below:

Major revisions have been applied to the manuscript in order to keep its focus clear and straight forward. We fully agree with the reviewer that the weight of the paper is on the properties derived for aged volcanic particles by initializing advanced instrumentation, data synergy and state of the art algorithms and developments achieved within EARLINET. This is now clearly stated in the abstract. The paper is not in any case a validation exercise for FLEXPART, thus all the related parts have been removed. Assumptions that are not supported have been removed. Our detailed answers to one by one reviewer's comments follow hereinafter.

P5317. Abstract: The large range given in the abstract for the effective radius, lidar ratio and the refractive index leaves the impression to the reader that all these values correspond to pure aged ash and the variability could be associated to different age, which is probably not the case. The authors should add a comment here on the cause of this large variability. In addition the authors should avoid to provide a correlation coefficient between LIRIC and FLEXPART which is based on few cases and should rather restrict themselves on a qualitative statement.

The large range found by the inversions for the effective radius and refractive indexes (and correspondingly for the lidar ratio) is mostly attributed to the mixing of the aged volcanic particles with aerosol types of local origin. This point is now stated in the abstract. Moreover, the 30% uncertainty of the inversion should not be neglected for the results reported. The correlation coefficient has been removed.

P5318-5319. Introduction. Since the paper is part of special section, some parts of the description of the volcanic activity could be shortened. In the last paragraph of the

introduction the authors should mention clearly what is the main objective of their study. Is it to characterize pure aged volcanic ash? To determine the mixing of ash with local aerosol sources? To validate FLEXPART?

The description of the volcanic activity has been strongly shortened. In the last paragraph we present the main focus of the paper, which is the study and characterization of aged volcanic ash and its mixing with locally produced aerosol particles.

P5320-5321. Instrumentation (lidar and CIMEL). This part should be drastically shortened, providing a basic description (avoiding too many technical details) and citing the appropriate references.

The instrumentation part has been drastically shortened, according to the referee's comments.

P5324. Equations (1) and (2). It is not clear the way it is written which variable corresponds to the output of LIRIC. Is it $C(z)$ in eq.2? The authors provide eq. 1 as a method to convert ppb to mass concentration, but in the right part of the equation it is missing the quantity to be converted. The authors should also provide a physical meaning of the term ppb when it comes to aerosol concentration. Ppb as a unit suggests a ratio. In line 25 what does pure non-volcanic mean? Please rephrase.

The equations have been revised according to new papers of Tsekeri et al. (2013) and Wagner et al. (2013), so now there are no missing terms. The main output of LIRIC is indeed the aerosol concentration profile ($C_{f/c}(z)$), which is expressed in parts per billion volume (ppbv), or equivalently, the volume of aerosol particles in a unit volume of air ($1000 \times \mu\text{m}^3/\text{cm}^3$). The non-volcanic term has been rephrased to refer to other aerosol types.

P5326: The authors should provide here a short description what is the case study all about. In addition in line 5 the authors mention that for this period no dust was predicted, does this necessarily mean that no dust was actually observed? Please add an appropriate comment.

Dust has been simulated by the dust model BSC-DREAM8b over Athens, but we showed no dust forecasts. However, only the depolarization measurements could provide an indication of dust presence, even though in coincidence with ash presence, the discrimination would have been very difficult due to the polarization sensitivity of both aerosol types. The good comparison of the lidar data with FLEXPART simulations however, which includes only volcanic ash emissions is a second indicator (beside the dust model) of no dust presence over Athens for the period reported.

P5327-5328: It would help the reader a lot if Figs 2,3 and 4 would be merged in a single multi-panel figure keeping the same time-span for the x-axis (which is not the case now),

so that they could be directly comparable. As it is known these figures include different number of days and this brings some confusion.

The Figures have been revised and grouped in one as suggested by the reviewer.

P5328. Line 7. The radiosonde could not show the existence of a dry aerosol layer, could only indicate a dry layer. Please rephrase. Line 20: Is the aerosol-cloud discrimination scheme an automated procedure? Any references?

Radiosonde cannot provide information on the aerosol layers, this is true, and thus this sentence has been rephrased. The aerosol-cloud discrimination has been referenced (Mona et al., 2012) and is a semi-automatic procedure.

P5329 and P5330. Figure 5 is highly confusing. It suggests a multi layer structure (not simply upper and lower troposphere) not discussed or shown earlier. In addition it is risky to associate measured lidar signals around 2 km to ash. A scatter plot would be more helpful if a comparison of the CM is what the authors want to demonstrate. P5330-Lines 8-13. Are these small particles in the lower troposphere associated with the volcano? Any physical explanation? lines 14 to 24 (P5330) there is a discussion on correlations based on an analysis not shown. The authors should either remove this part or if they think that it is essential for the paper they should support this with a different figure 5. To my opinion Figure 6 does not provide anything new in the discussion. The text provided here could assist the description of a revised figure 5.

Figure 5 has been revised including information from Figure 6 that refers to free tropospheric volcanic particle loads only. It is now clearly stated that the particles of volcanic origin can be distinguished only in the free troposphere and not within PBL. A scatter plot is not provided as the demonstration of a direct comparison of a CM is not our objective. In P5330, lines 8-13, the small particles probed in the low troposphere are associated with advected volcanic particles according to the masking procedure. Correlation reports have been rephrased following reviewer's suggestion.

P5331. Since FLEXPART uses only ash as a source the comparison with LIRIC, which uses lidar signals that correspond to the real atmosphere, would help to determine the state of mixing with other aerosol types rather than to validate the model. The authors should make an appropriate comment here.

This idea could be applicable, however we decided to avoid such an analysis since this would rely on the accuracy of FLEXPART in its simulations. We want to keep the focus straight forward and report the measured values for the pure loads of volcanic particles found in the free troposphere.

P5332. The positioning of the LIRIC layers is directly associated with the layers in the lidar signals, so there is no need to repeat here the discussion on the height of the layers. The authors should focus how the concentrations compare and give emphasis on pure

and mixed layers. The discussion about correlation coefficients should be avoided, since they are based on limited cases and they don't have any statistical significance.

The text has been revised to follow reviewer's recommendations. However, we kept the discussion about the correlation coefficients, since this provides additional new information (giving an order of comparison, despite the low statistical significance of our dataset).

P5334. Lines 13-15. The authors probably mean g/m³ and not mg/m³. The discussion of the lower part of figure 10 is highly speculative and confusing. Is surface PM₁₀ based on measurements? If yes such a small variability of surface PM₁₀ could associated too many other factors (local variability, meteorology etc). I would suggest to remove this part if not supported with further evidence.

The units have been checked and reported correctly. PM₁₀ data have been removed; we agree with the reviewer that the discussion was too speculative.

P5335 to 5337. This part of the discussion is very well written but it comes late in the paper. Eventually the authors should merge this part with the previous section, first present this part and then LIRIC, and thus they would avoid describing many times the same layers with a different perspective every time.

We have re-arranged the text in order to fulfill reviewer's suggestions.