

Dear Editor, dear Referees,

please find in the following our point-by-point replies to the comments of the Anonymous Referee #1 and François Dulac (Referee #2). Please find in /normal characters/ the Referee's comments and in /**bold characters**/ our replies. The cited text from the manuscript is in /**bold italic characters**/. All mentioned page/line, Sections and figure numbers, are based on the revised manuscript. In general, we have found their reviews and their constructive criticism very useful, and we are enthusiastic about the revised manuscript. In our opinion, the manuscript is greatly improved in clarity and of more general interest than the previous manuscript version.

Sincerely,

Pasquale Sellitto, on behalf of all co-authors

Referee #1

General Comments

Sellitto et al. describes a case study of a small eruptive event of Mt Etna in October 2013. It focuses on the impact that SO₂ and ash emissions from the volcano have upon aerosols and their properties in a plume transported from the crater over a scale of a few hundred kilometres. The study combines quantitative and qualitative in-situ observations of the eruption, satellite observations of the downwind plume, emission estimates of SO₂ and ash from the volcano based on satellite observations, ground-based remote sensing observations of the downwind plume, and modelling of the transport of the plume and its chemistry (simplistically for SO₂) and sedimentation for ash. Using these methods the authors characterise the transport of the plume the temporal evolution of the SO₂ and ash as it moved away from the volcano over the Mediterranean. In some cases, inferences are made about the composition of the aerosol in the plume in relation to sulphate. Finally, the radiative impact of the aerosols and ash are estimated. In relation to the radiative forcing (RF), the authors conclude that it varies strongly with single scattering albedo, and, we can probably assume from this, the aerosol composition as well. From their analysis, the authors infer that sulphate aerosol forms a large part of the aerosol plume composition.

I think that the subject matter of the paper fits within the scope for ACP. I also think that the topic of the paper is interesting and that attempts to characterise the impacts of small eruptions on aerosols and RF are sufficiently novel. To me, the results on the RF were the most interesting. I also think that the authors should be commended for trying to use several methods together for analysing the eruption and its affects. The authors make a great effort to emphasise the complementary nature of these methods, and although I am convinced this approach was useful, I think they could make more effort to more clearly expose the advantages of doing so (see comments below).

Thank you for the kind words.

Despite these largely positive points, I think the study as it is presented currently misses one important analysis step (see comments below), and I therefore would only recommend publication after major changes. I now detail my two major comments:

Major comments

1) This study would benefit greatly from a sulphate aerosol process model to be able to simulate the full process of sulphate aerosol formation and evolution from SO₂ emission, oxidation of SO₂, sulphate aerosol formation, and aerosol losses. As it turns out, the RF estimates are highly sensitive to the single scattering albedo and therefore to the aerosol composition. The authors make it clear that this is the single most important factor in determining the radiative impact of the aerosols. While satellite observations of the single scattering albedo are used in the RF calculations (and these estimates seem reasonable), the evidence in the paper only allows us to infer indirectly what the aerosol composition is and we can only gain limited information about the processes that ultimately control the aerosol composition, properties, abundance and ultimately their radiative impact. Indeed, the methods for assessing the aerosol composition used are either indirect (the single scattering albedo) or inferential (the ash simulation shows fine ash is lost from the plume early in its transport therefore it is assumed that ash plays only a limited role in the later plume composition). In this sense, this study presents a snapshot of the radiative impact of this eruption and we gain only limited knowledge about the underlying processes that could be applied to our understanding of other eruption events or to understanding the global impact of this kind of activity. I therefore recommend that the authors run simulations exploring the formation, evolution and behaviour of sulphate aerosols. One alternative (if this work was considered to be too much effort) would be for the authors to more clearly formulate their inferred conclusions about sulphate aerosols and their impacts into a testable hypothesis (perhaps using a schematic figure) that could be followed up in future studies. It somehow seems disappointing though in this study to have assembled all of this data in a case study and then not to explore the underlying processes using a model. Without these changes one could argue that the paper should be vastly condensed to quickly present the radiative transfer calculations. A lot of effort is dedicated to explaining the FLEXPART simulations, but these model results only give us the vertical distribution of the volcanic aerosol (the RF calculations are insensitive to this distribution) and the inferential evidence about the aerosol composition for the RF calculations. You could argue that the FLEXPART modelling information could therefore be reduced to one or two short paragraphs.

This is a good point. We agree with the Referee #1 that this study, and in particular the attribution of the downwind impacts, would greatly benefit from a chemical/micro-physical model able to simulate the processes leading from SO₂ emissions to sulphate aerosols, and their chemical and micro-physical characterisation. It is worth noting that the radiative forcing is strongly dependent from this characterisation, e.g., the H₂SO₄ concentration and the effective radius of the sulphate aerosols. It is actually the lack of such modelling tool at our institutions, as well as a dedicated sulphate aerosols satellite product, to motivate the synergistic use of the available tools, as reported in the present work. While these two tools, sulphate aerosols chemical/micro-physical modelling and satellite observations are in our agendas for future development (our ongoing work in satellite observations is mentioned in the paper, see ref. [Sellitto and Legras, 2016]), we don't have, for the moment, consolidated products that can be used in this work. At this stage, as stated by the Referee #1, we can only gather indirect or inferential information about the evolution of SO₂ to aerosols. The development of a modelling tool would take months and, as mentioned by the Referee, is virtually impossible for the present work but a necessary step for future developments.

To account for this, we think that an optimal configuration is to mention it in: 1) the

new dedicated sub-section describing the different synergies (see Referee #1, Major Comment 2), 2) in Section 6.3, 3) in the Conclusions. Here are in details the 3 added texts. 1) The last paragraph of Section 2.1 reads: *“It is worth noticing that, for a more complete understanding of the downwind impact of volcanic emissions, two important information layers would be the ones describing the spatial distributions (satellite observations) and the formation/evolution processes (chemistry/micro-physics modelling) of the sulphate aerosols produced by the conversion of volcanic SO₂ emissions. Works are ongoing to provide these further information layers.”* 2) We have modified the last paragraph of Section 6.3 to: *“The unequivocal attribution to one of the two sources (fine ash or secondary sulphate aerosols) is impossible at this stage due to the lack of specific measurements of the chemical composition of the aerosols, or a simulation of the sulphates chemical/micro-physical evolution, as mentioned in Sect. 2.1. Then, we exploit our synergistic approach to infer the prevailing composition of the volcanic aerosol at Lampedusa. Due to the considerations in Sect. 5, we believe that fine ash particles had a very small impact on the measurements at Lampedusa (synergy h) in Fig. 1). Conversely, the discussion in Sect. 4, the comparison of micro-physical aerosol properties at Lampedusa with data in the literature, and the indications of favourable conditions for the formation of new particles through gas-to-particle conversion, suggest that the secondary sulphate particles may be the primary cause for the observed aerosol optical properties and their evolution following the eruption.”* 3) We have modified the second to last paragraph of the Conclusions to: *“Here we want to mention that an analysis based on SO₂ and ash transport modelling and satellite observations, and aerosol optical properties from ground observations, is necessary because of the lack of reliable sulphate aerosol satellite products and chemistry/micro-physics modelling. Dedicated satellite products could allow a direct observation of sulphate aerosols production, life cycle and burden at the regional scale, and chemistry/micro-physics modelling could allow the simulation of their formations and evolution processes. Ongoing studies will provide such kinds of satellite products [Clarisse et al., 2013, Sellitto and Legras, 2015] and modelling tools (the coupling of FLEXPART trajectories with sulphate aerosols box models) are under investigation.”*

Finally, here we want to mention that a more complete analysis of the radiative forcing of Mount Etna's plume and its dependence on the aerosol optical properties has been recently shown in the paper [Sellitto and Briole, 2015], which is now cited and discussed in Section 7: *“The radiative forcing is, then, strongly dependent on the SSA. This evidence has been recently confirmed in a more general context (wider variability of the optical parameters). Sellitto and Briole (2015) have shown how the RFE is more dependent on the SSA than the other aerosol optical input parameters for this study (the asymmetry parameter and the Ångström exponent), especially when sulphate aerosols are dominating (bigger values of the Ångström exponent and the SSA). As the SSA is the dominating factor in the RFE variability, the formation and evolution processes of volcanic sulphate aerosols is very critical for the downwind radiative impacts and a more detailed description of these processes, i.e. with a chemistry/micro-physics model, would be a necessary add in this kind of impact studies, as mentioned in Sect. 2.1.”*

2) The descriptions of the synergistic use of different methods need to be improved. As it is we are told about all of the different methods and how they contribute to the paper, but we are told this information in several different places and one has to piece together this information to get the complete picture. I think it would greatly improve the clarity of this

aspect of the manuscript if the authors presented the information in a table or (even better) a schematic diagram to show what function each method fulfils in the study. As an example, one has to read quite far into the paper to learn that the FLEXPART simulations are used as the basis for the vertical distribution of aerosols in the radiative transfer calculations.

The idea of the joint use of different methods to investigate an eruptive event, from source characterisation to downwind impact, is very central to our manuscript. Therefore, we accept with enthusiasm this suggestion. Correspondingly, in the revised manuscript, we show schematically the synergies and interplay of the different methods by adding the scheme of Figure 1 and a new dedicated section (Section 2.1: “Synergistic use of the different information layers”). In addition, we now make specific reference to the individual identified synergies (list from a) to h) in Figure 1) throughout the text, to guide the reader through our synergistic use of different information layers. *

Please also consider that we slightly change the title to include the radiative modelling that wasn't mentioned before. The title now reads: “*Synergistic use of Lagrangian dispersion and radiative transfer modelling, and satellite and surface remote sensing measurements for the investigation of volcanic plumes: the Mount Etna eruption of 25–27 October 2013*”

Specific Comments

1) The model for SO₂ oxidation and chemical loss is highly simplistic with an assumed calculated constant lifetime and is only adequate. The authors should highlight the weaknesses of this method and should explain how it affects their results and conclusions regarding SO₂ evolution in the plume.

Besides mentioning the expected added-value of a detailed chemistry/micro-physics modelling from the point of view of the sulphate aerosols and the downwind characterisation of the aerosol layer and its radiative forcing (Referee #1, Major Comment 1), we mention in Section 2.3.1: “We then considered a fixed lifetime of 6.7 h (loss rate of about $4.2 \times 10^{-5} \text{ s}^{-1}$) for the sulphur dioxide in our simulations for that day. It must be noted that using a fixed value for the SO₂ lifetime is a simplified approach and more refined estimations (time- and atmospheric parameters-dependent) could be obtained using a detailed chemistry modelling, as mentioned in Sect. 2.1.”

2) Page 31338, line 25. Water vapour is not neutralising unless it contains a base.

We have modified the sentence to “...in presence of water vapour and neutralising species...”

3) Page 31340, line 3-17. As presented here, I found the argument for a synergy of observations and simulations to be poorly motivated. As it is, we are told only about the benefits and disadvantages of satellite observations, but we are not told about in-situ observations or models before we read a conclusion that a synergistic approach is best. This seems disjointed without motivating text related to models or in-situ observations. Can the authors please remedy this problem.

We agree. We have modified this paragraph, that now reads: “...below the detection limit of instruments onboard satellites. Modelling tools can supply further information, provided that input parameters are carefully selected. Unfortunately, many processes (e.g., dynamical, chemical, micro-physical processes) are still characterised by a poor fundamental knowledge. More detailed information on the downwind impact can be provided by ground-based instruments at selected locations, but, in this case, a limited information on the transport and evolution of the emitted gases and particles is available. Thus, a reasonable approach consists in exploiting the synergy of observations and simulations...”

4) Page 31342, lines 8-14. We are told about the method for retrieving the SO₂ and ash emission rates from MODIS observations of the eruption. It would appear that this method implicitly assumes that SO₂ has no loss process and, on the timescale of the emission inversion, that it has an infinite lifetime. This is not stated. The authors should state this, they should justify the assumption, and explain how it affects their results. This assumption should lead to an underestimation of the emission rate since losses are discounted. It is worth pointing out that the emission rate obtained using an infinite SO₂ lifetime is used to calculate the finite lifetime for SO₂ in equation (1) (see comments below), so there is an inconsistency here. This point should be acknowledged and discussed by the authors.

We have clarified this aspect in Section 2.2.1: “This method implicitly assumes an infinite lifetime for SO₂ and ash during the emissions inversion. This could lead to an underestimation of the emission rates.”

5) Page 31345, equation 1. You should mention that the method for retrieving Q will likely lead to an underestimate and that equation 1 will therefore likely overestimate τ .

We have clarified this aspect in Section 2.3.1: “...could be obtained using a detailed chemistry modelling, as mentioned in Sect. 2.1. It must also be noted that, as stated in Sect. 2.2.1, the emission rate Q is probably underestimated and then the lifetime τ could be overestimated. Estimates of the SO₂ lifetime...”

6) Page 31347, lines 7-9. It is not 100% clear from the language whether you have used observed SSA, observed Angstrom exponent, and observed asymmetry parameter. As it is written, it only seems that you use an observed SSA. If this is not the case then please modify the text.

The three are observed parameters (AERONET). We have clarified it in the text.

7) Page 31350, lines 18-20. It is not very clear what the authors mean when they say: “It should be noted that the ground-based spectroscopic emissions measurements were not available for few hours during the main phase of the eruption and, correspondingly, the retrieved emissions rate is likely underestimated.” Do you mean that there were no data at all? Or did you use another data source, i.e., satellite? Or did you infill the data?

The ground-based emission measurements are based on the UV extinction by plume constituents. During an eruptive event, ash emissions can be so important that the plume is too opaque to measure SO₂ with acceptable uncertainties. This was the case for this eruption, during a period of a few hours. It is possible that as ash increased (leading to an increasingly opaque plume) so did SO₂ emissions and then these observations missed part of the magnitude of the SO₂ emissions and,

correspondingly, the emission rates are underestimated. It is important to notice that this aspect has no impact for the FLEXPART simulations, where satellite-based emission rates are used.

8) Page 31351, line 26. When discussing the SO₂ lifetime, it is not clear if you are comparing the model to the observations or visa-versa. Which one has the longer lifetime? In general I found that this paragraph did a poor job of differentiating between talking about the model and observations and it was therefore hard to form an idea about how the two were performing relative to one another.

Our simulations are initialised with MODIS emission rates of Figure 4 and the fixed lifetime of Equation 1. Figure 5 shows the simulation for 26/10 at 12:20 (b), that we compare to the MODIS image at the same time (a). As the MODIS image is a static information, we don't have a specific lifetime estimation from it, so the considerations about mutual differences of (a) and (b) are based on the initialising parameters of the simulations (b).

9) Page 31354, lines 2-4. The authors discuss disentangling the impacts of fine ash and fine sulphate aerosol contributions to the fine aerosol burden. The authors seem to be claiming that knowledge about fine ash will essentially lead to more information about sulphate aerosols. In order to believe this it appears one has to implicitly assume that the remaining contribution to fine aerosol is secondary sulphate aerosol formed from SO₂ oxidation and subsequent sulphate deposition. Without using a process model for secondary inorganic aerosol, how can the authors be confident that this assumption is valid? Please can the authors state this assumption more clearly and justify it. In addition, please can this issue be discussed in terms of its effects on the results and in terms of how it limits the conclusions?

Please refer to the detailed discussion of Major Comment 1.

10) Page 31359, lines 5-12. Please can the authors add a little more explanation about the link between SSA and sulphate aerosols. Presumably higher SSA is indicative of a higher sulphate content.

Please consider that this is already discussed at the end of Section 6.3 (second to last paragraph).

11) Page 31358, lines 16-18. The origins of the information stated to come from Sections 4 and 5 was not very clear. Can this be made clearer please?

We have modified this paragraph, that now reads: “Due to the considerations in Sect. 5 (the size-dependent ash dispersion analysis indicates a limited fine ash component at Lampedusa), we believe that fine ash particles had a very small impact on the measurements at Lampedusa (synergy h) in Fig. 1). Conversely, the discussion in Sect. 4 (the height-resolved SO₂ dispersion analysis indicates that SO₂ concentrations are significantly higher than background levels in the upper troposphere over Lampedusa), the comparison of micro-physical aerosol properties at Lampedusa with data in the literature, and the indications of favourable conditions for the formation of new particles through gas-to-particle conversion, suggest that the secondary sulphate particles may be the primary cause for the observed aerosol optical properties and their evolution following the eruption.”

12) Page 31359, lines 7-10. Can the authors please try to reformulate this sentence as it was quite unclear. "Since it is not possible to quantify the contributions from lower tropospheric aerosols and from volcanic particles to the total AOD, we have used a single aerosol type with the measured AOD, Ångström exponent, SSA and asymmetry parameter in the model setup." Specifically, do the authors mean "the separate contributions...to the total AOD"? And what is the single aerosol type representative of? Presumably, volcanic aerosol?

Yes, the interpretation of the Referee #1 is right. We have slightly changed the sentence to clarify it.

13) Page 31359, lines 15 onwards. The authors discuss different parameters that they have tested that affect the calculation of radiative forcing. The authors have tested the sensitivity to SSA, which is a very interesting test. Similarly to the recommendation for text in the 2nd to last paragraph of Section 6.3, can the authors link the discussion of varying SSA back to aerosol composition. I assume that higher SSA is representative to higher sulphate to ash levels, but it would be clearer if this was stated.

This information is already given at the end of Section 6.3 and is linked to the aerosol composition at the end of Section 7 ("*...supports the hypothesis of the major role of sulphates in our case study*"). Therefore, we think that re-state it here would be partly a repetition. We are oriented to leave the text as it is but if the Referee #1 still thinks that this is a necessary clarification, we will modify this paragraph.

14) Page 31359, SSA discussion and Table 2. I found the results linking the impact of SSA on RFE to be very interesting. I think these results highlight the need to carry out process modelling on the sulphate aerosol. As it is, the conclusion appears to be that the radiative forcing depends strongly on a process that isn't simulated in the model.

As suggested by the Referee #1, and in particular in reference to his Major Comment 1, we have discussed the need of process studies and proposed future work about that throughout the text (for more details please refer to our reply to Major Comment 1).

Technical Comments

All technical comments have been considered and changes have been done accordingly.

Referee #2 : François Dulac

General comments

This work is a case study combining transport and radiative budget modelling, and satellite and surface remote sensing in order to follow the dispersion of a volcanic plume from Mount Etna that was emitted during the ChArMEx Enhanced Observation Period, and its composition in terms of SO₂ and particles. The final objective is an assessment of the aerosol plume impact on the direct radiative budget downwind at Lampedusa Island, in

terms of forcing efficiency at the surface and top of atmosphere. It is found significant, of the order of -55 and -45 W m⁻¹ AOD⁻¹, respectively, and is mainly attributed to secondary sulfate aerosol particles relatively to primary mineral dust and ash particles. Overall, I find that the paper objectives and methodology are sound and relatively clear and that results are relevant for publication in ACP. I have a number of minor comments listed below, among which main scientific issues concern the surface albedo (comment #1), the size distribution (series of comments #4) and the need to further discuss in the conclusion the interest and limitations of this case study in the regional context (comment #9). Putting special attention to the readability of figures given their reduction to ACP format is necessary (comment #10). A list of proposed small corrections is following my comments.

Thank you for the kind words.

Minor comments:

1) I am concerned by the treatment of the surface albedo, which it is expected to impact the aerosol radiative forcing (e.g. Zhuang et al., Atmos. Environ., 2014). Assuming a constant surface albedo throughout the solar spectrum as hypothesized (p. 31347) should be argued. The surface albedo value used from Meloni et al. (2003) accounts for the influence of Lampedusa Island in a marine region of 20 km in radius and, as such, is very specific to the area. This should definitely be made clear in the paper because the reader could think from the abstract and conclusion that aerosol direct radiative forcing results given here apply over sea water. I would expect that there are additional computations of the forcing in order to test the sensitivity of the forcing to the surface albedo. At least a seawater surface adapted to this marine region should be considered (note that sea surface reflectance values at several solar wavelengths for the considered week are available from MODIS at <http://modis.gsfc.nasa.gov/data/dataproduct/Rrs.php>), and possibly a broader range of surface types found in the region (e.g. in Sicily, Malta, Tunisia).

We completely agree with the Referee: the surface albedo is an important parameter that can significantly modulate the radiative forcing. This was already mentioned in the manuscript (“[...]However, the RFE values depend on the day of the year and on surface albedo, and a direct comparison is not possible.[...]”) but we understand and share the concern of the Referee and we decided to perform a few new analyses to explicitly address the sensitivity of the volcanic aerosol radiative forcing to surface albedo. Therefore, we tested how the RFEs and f vary for extreme values of the (wavelength-independent) surface albedo: one value for the sea (0.07) and one for the desert scenarios (0.36). The results of these analyses are discussed in the new Section 7, Page 27, Lines 3-26 (we don't report the text here but please find it in the revised manuscript). The pertinence to produce more refined radiative transfer simulations by considering a wavelength-dependent surface albedo is also mentioned, and we considered it for dedicated future analyses.

2) Page 31342, section 2.1.1: it would be expected to check that SO₂ products from the different sensors (IASI and TES) are coherent with MODIS retrievals.

We have checked our MODIS product with respect to IASI (mentioned in Section 2.1.1) and OMI (mentioned in Section 6.1) and both found consistent as direction and magnitude of the plume, even if a detailed comparison is not feasible due to the reduced spatial resolution of IASI and OMI with respect to MODIS.

3) Page 31343, section 2.1.3: the pixel resolution in the area of interest between Etna and

Lampedusa is of better interest than at the distant geostationary sub-satellite point (0° in latitude and longitude).

We agree. We now mention the pixel dimensions in the area near Lampedusa (4.3 x 3.3 km).

4) Page 31345, lines 22-23: the size distribution discussion is confusing and should be reconsidered.

4.1) The use of a normal standard deviation (σ) that characterizes a symmetric normal (Gaussian) distribution is not appropriate to a lognormal distribution, which is very dissymmetric around its modal (peak) diameter; indeed, the dispersion of a lognormal distribution is characterized by its geometric mean diameter D_g and a unitless geometric standard deviation (σ_g) which is a multiplicative factor so that the dispersion is characterized by $[\ln(D_g) / \sigma_g; \ln(D_g) \times \sigma_g]$.

4.2) Particle size classes 0.1, 0.316, 1, 3.16, 10 and 31.6 μm would be more consistent than 0.1, 0.35, 1, 3.5, 10 and 35 μm to respect a geometric progression that better applies to a lognormal distribution.

4.3) It should be specified in table 1 whether the distribution considered is a number distribution as assumable by default, or a volume (or mass, assuming constant density with size) distribution, which I suspect given the numbers in table 1; D_g of the two distributions have the same σ_g and their respective D_g values are related by a simple relationship; the two values might be provided.

4.4) The geometric standard deviation cannot be 1.0 as stated; this would correspond to a distribution limited to a single particle size with no dispersion at all.

4.5) Using the size distribution given in table 1 and attempting a simple visual fit by a lognormal size distribution with a mode at 10 μm , I end with a geometric standard deviation of 2.0 to fit the peak (see the left plot in figure 1); but the left tail (at small sizes) of the distribution used implies a second mode that can be approached with a modal diameter of 1.0 μm and a σ_g of 2.3, as illustrated below (right plot in figure 1); note, however, that these values are rough estimates of the size distribution (e.g. size 0.1 μm is still not well fitted) aiming at fixing ideas and discussing erroneous statements on the size distribution in the manuscript; a proper fit of the proposed distribution in table 1 would request a σ^2 -based adjustment; assuming that these are volume distributions yields corresponding geometric mean diameters of the number distribution of about 0.125 and 2.37 μm .

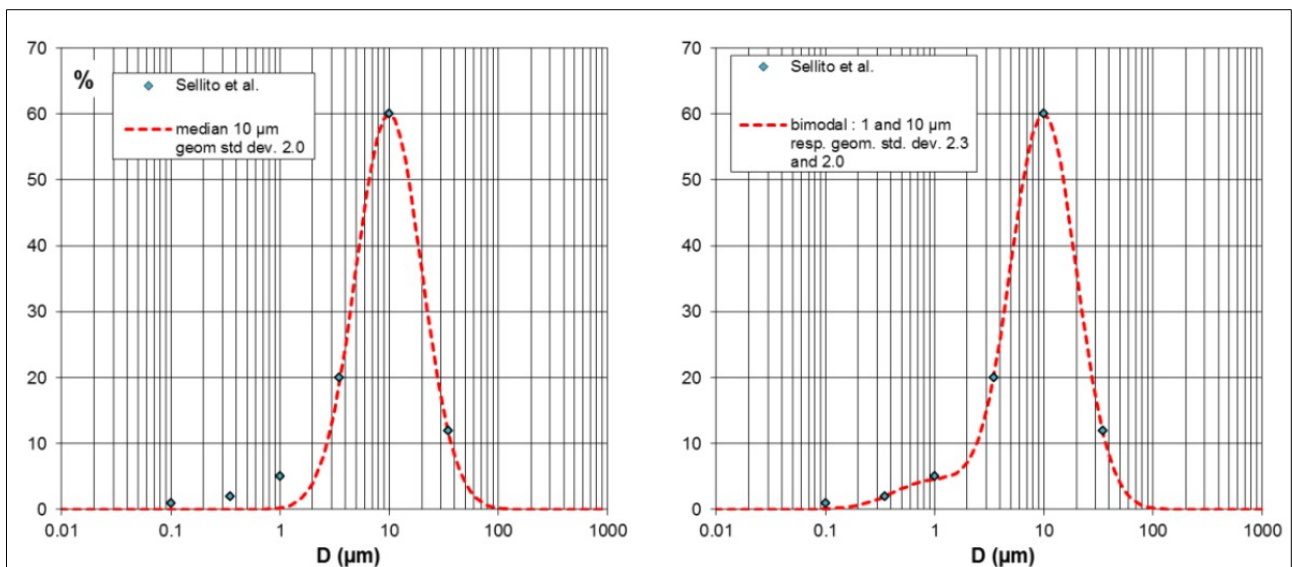


Figure 1: Rough adjustment of the particle size distribution proposed in table 1 of the paper (blue dots), by a monomodal lognormal with a geometric mean diameter of 10 μm (left), and by adding a second mode to fit the tail of the distribution at small sizes (right).

In our work we use a typical ash distribution, as it is produced by the “mk_releases” routine of FLEXPART (acknowledged at the end of our paper). This routine is based on the work of Mastin et al., 2009, and, as discussed in our paper, uses a size distribution consistent with sunphotometric and remote sensing measurements, and deposited ash samplings. In any case, Referee #2 is perfectly right: it is not a monomodal log-normal size distribution, as we simplistically stated. We thank the Referee for this correction and the other precision he made, including the calculations and fits (Figure 1, above). We use these precisions to reformulate this paragraph and to correct the terminology, as well. The paragraph now reads: “Six ash classes are modelled, based on a typical ash size distribution obtained with the *mk_releases.f* routine (courtesy of Nina Kristiansen and Andreas Stohl), based on the work of Mastin et al. (2009). The central radii and the percent population of each class in the ash distribution are listed in Table 1. This distribution can be roughly approximated with a bi-modal log-normal size distribution (geometric mean radii of 1.0 and 10.0 μm , geometric standard deviation of 2.3 and 2.0, respectively), representing both fine and coarse ash particles. This size distribution is consistent with that observed in deposited ash and from airborne (see, e.g., Stohl et al., 2011) or remote sensing observations. Sun photometric measurements (see, e.g., Watson and Oppenheimer, 2000) performed at Mount Etna have shown a 3-modal log-normal aerosol size distribution, with a coarser mode with a mean radius greater than 5 μm , which is partially attributed to ash during relatively weak activity phases. Coarse modes with higher mean radii, greater than 10 μm , are observed in eruptive size distributions, (e.g., at Mount Redoubt) and attributed to ash particles (Hobbs et al., 1991). In addition, a campaign has been conducted at the beginning of October 2013, to characterize Etna’s emissions with in-situ measurements of different gases and aerosols properties. Bi- and tri-modal aerosols size distributions were observed, with the coarser modes identified as ash (T. Roberts, personal communication). These latter modes exhibited log-normal distributions with mean radii and widths similar to the ash size distribution used in our simulations. These direct observations at Etna, for a period of only a few days before our study, justify our ash size distribution assumption, even if coarser particles can be observed during an eruption event”

We mention that, using a mono-modal size distribution, as for the red curve in

Figure 1a above, would lead to stronger conclusions about the relatively small importance of the fine ash component (the fit would indicate a smaller mass fraction for fine ash than what we use), thus our inferred results (the exclusion of fine ash and the bigger importance of sulphate aerosols in the attribution of the downwind impacts, including radiative forcing) are robust with respect to this kind of size distribution variations.

In addition, we have specified in the caption of Table 1 that the size distribution is a mass distribution.

5) Page 31349: what is supporting the hypothesis of a constant wind speed of 18 m s⁻¹; is it assumed constant with both time and altitude? Can you estimate related uncertainties?

The wind speed is assumed constant with both time and altitude, which is obviously a simplified approach. Using measured or modelled (e.g., from reanalyses) variable wind speed is a refinement of the emission rates estimations that we consider for future applications.

6) P. 31352: In section 4, I think it would be better to discuss first the altitude of the plume before discussing simulations. I would sub-title “4.1 Altitude of the SO₂ plume” the section starting from line 13 and shift it early within section 4, in order not justifying a posteriori the FLEXPART simulation hypotheses. Section 4.2 would then start with the presentation of Fig. 3a (presently p. 31350, line 26).

We don't agree with the Referee #2 about this comment. The main reason why we arrange the analysis in this way, is to allow inference about the vertical distribution of the SO₂ plume, after its consistency with satellite observations is verified. We think that this “synergy” is clearer after the add of Figure 1. Therefore, we prefer to leave the structure of Section 4 as it is but we are open to further discussion if the Referee #1 retains his idea of inverting these two proposed sub-sections.

7) The present sub-section 6.4 includes the main results that justify the rest of the study; according to me it would deserve to become a full section (7); this section should be augmented with a sensitivity study to the surface albedo (see comment above); it is also needed (p. 31359) to mention that the stratospheric AOD is considered negligible based on a reference to be cited.

We agree. We have updated the Subsection 6.4 to Section 7. This Section now contains a new paragraph about the analysis of the sensitivity to surface albedo (Referee #2, Comment 1). The stratospheric AOD is indeed considered negligible and a discussion about the reasons of it is included in this Section: “...*is much higher than expected for sea salt aerosols. The stratospheric aerosol optical depth is considered negligible during this event. In fact, while the stratospheric aerosol layer has enhanced particle concentration and optical depth in the period 2000-2010, due to the occurrence of a series of moderate stratospheric volcanic eruptions, this layer has returned to background values by 2013 (a stratospheric aerosol optical depth of about 0.005-0.008, see Fig. 1a of Ridley et al. (2014)). Thus, we assume that the upper tropospheric volcanic particles produced during this eruptive event are dominant...*”.

8) P. 31359: I do not understand the argument that it is better to normalize the forcing by the AOD because there is uncertainty on the volcanic aerosol proportion in the column

(lines 24-25); please reconsider this sentence; according to me, the consequence of such uncertainty is rather a limitation for interpreting the forcing attribution to the various types of aerosol particles present in the column; I find that a discussion on attribution is missing.

What we meant was that we have columnar observations of the aerosol optical properties (AERONET) and, while these values refer to the overall column, we don't have a specific knowledge on the vertical distribution of the aerosols (lower tropospheric + volcanic) and then we cannot attribute the RF to the volcanic component only. The values of the Ångström exponent, the SSA and the asymmetry parameter seem to point at an almost purely volcanic (sulphates) layer but we have decided to stay more cautious by providing a relative (RFE) estimation of the radiative forcing instead of an absolute one. This is a first estimation and, if more objective information about the vertical distribution of the aerosol layer will be available for other future studies (e.g., with LIDAR observations), an absolute estimation of the radiative forcing may be attempted.

9) Conclusions: I would expect that you replace this specific case study, its interests and limitations, in a broader context; I find for instance that we miss a reminder on the AOD range encountered, the frequency of occurrence of this type of event with moderate AOD, the distance from the emission at which the forcing was evaluated, the range of distances impacted by that type of plume; can we extrapolate conclusions on the volcanic sulfur cycle from Etna emissions from this case study? etc.

We agree with the Referee #2 and we added the following sentence at the second to last paragraph of the Conclusion section “*The REFs are very sensitive to the surface albedo and then to the location where these estimations are performed. Nevertheless, even if these estimations are specific for Lampedusa, the assumed surface albedo is very close to a pure marine albedo and then these estimations are representative of a larger sea covered area in the Mediterranean basin.*”, and the following last paragraph at the end of the Conclusions section “*Studies are ongoing to evaluate the frequency of this type of event. Preliminary results show that the area of Lampedusa (south-west direction from Mount Etna) is ventilated by air masses coming from Mount Etna for only about 5% of the time, in the period 2000-2013 (Sellitto et al., 2016). Correspondingly, the impact of volcanic aerosols (from passive degassing or moderate to explosive eruption) in the central-southern Mediterranean is only episodic. A stronger long-term impact is expected at the same distance in the eastern direction (ventilated for about 80% of the time in the same period, considering south-east to north-east quadrants).*”

Please note the new reference (Sellitto et al., 2016) that first presents our (temporally) broader analysis and is matter for a paper in preparation.

10) Compared to the initially submitted version, the reduction in size of figures for matching the ACPD page format had a dramatic effect on their readability: most figures in the paper deserve a significant expansion and in addition most figures from Fig. 3b have far too small characters in their axes and/or legend box; please be careful to this.

We have tried to enhance the readability of the figures. All characters have been enlarged. Following Referee #2's suggestions, Figure 1 and Figure 9 (numbering of the previous manuscript version), which were the most unreadable figures, have been both split into two (now Figures 2 and 3, and Figures 11 and 12). We think that now the Figures are more readable but, if it will be necessary, we will work further

with the editorial office to enhance them.

11) I follow referee #1 to recommend an additional figure presenting a scheme of the methodology that would be very helpful to the reader.

We have added a scheme in Figure 1 (please look at Referee #1, Major Comment 2)

12) Figure 1e: specify in the legend how long in time the trajectories are; plotting only one every two or three trajectories would certainly help the readability; I would find useful adding a figure 1f showing a time altitude plot of the same FLEXPART trajectories as in 1e; then, making a separate figure with 1e and 1f would allow to significantly enlarge the plots and images in Figs1a-d as well.

We have: a) separated the sub-figures of the MODIS RGB images from the FLEXPART simulations, b) added a new sub-figure with time/altitude plots for the FLEXPART trajectories, c) indicated the position of Lampedusa in the MODIS images, d) indicated the time duration of the trajectories. We now have 2 figures out of 1 (Figures 2 and 3 of the revised manuscript) which are far more readable than the previous Figure 1.

13) Figure 9c is independent from 9a and 9b and would better be given in a specific, new figure.

Done.

Technical comments:

All technical comments have been considered and changes have been done accordingly.