General Comments

Sellito 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). 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:

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.

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.

Specific Comments

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.

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

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.

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.

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.

Page 31347, lines 3-5. You have explained that you use a “moderate volcanic aerosol layer” in the radiative transfer calculations. However, it is not clear in the text what the properties are of this layer, nor is it clear why the choice of this particular layer is appropriate. This choice should be described in more detail and it should also be adequately justified.

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.

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?

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.

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?

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.

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?

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?

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.

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.

Technical Comments

Abstract, line 2. I recommend to not use "/". Do you actually mean "and".

Page 31339, line 5. Change to "…from **a** few days…".

Page 31340, line 1. Change to "…helps **the** understanding **of** the contribution…".

Page 31340, line 20. Change to "characterization of such a kind of eruptive **event**,…"

Page 31340, line 28. Change to "…and at **a** relatively large distance…".

Page 31342, line 1. Change to "…of the volcanic plume **is** most opaque…".

Page 31344, line 1. Change to "…are separated into two classes…".

Page 31344, line 8. Change to "…high thin clouds using two IR **channels: a window and a sounding channel**…".

Page 31345, line 17. "**An** SO2 point…".

Page 31346, line 1. "…with **a** mean radius…".

Page 31346, line 8. "…with a mean radius and width…".

Page 31350, line 24. "We therefore study **in** more **detail** the emissions…".

Page 31353, line 3. "…**the** south west…"

Page 31353, line 23. "…towards **the** south west…"

Page 31361, line 5. "…sources is of particular importance.."