Review of manuscript acpd-15-31335 entitled "Synergistic use of Lagrangian dispersion modelling, satellite and surface remote sensing measurements for the investigation of volcanic plumes: the Mount Etna eruption of 25-27 October 2013", by P. Sellitto et al.

By François Dulac, 18 Dec. 2015

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 SO_2 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.

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 see surface reflectance values at several solar wavelengths for the considered week are available from MODIS at http://modis.gsfc.nasa.gov/data/dataprod/Rrs.php), and possibly a broader range of surface types found in the region (e.g. in Sicily, Malta, Tunisia).

2. Page 31342, section 2.1.1: it would be expected to check that SO_2 products from the different sensors (IASI and TES) are coherent with MODIS retrievals.

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).

4. Page 31345, lines 22-23: the size distribution discussion is confusing and should be reconsidered.
   - 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) / σ_g; Ln(D_g) x σ_g].
   - 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.
   - 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.
   - 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.
   - 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 χ^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.

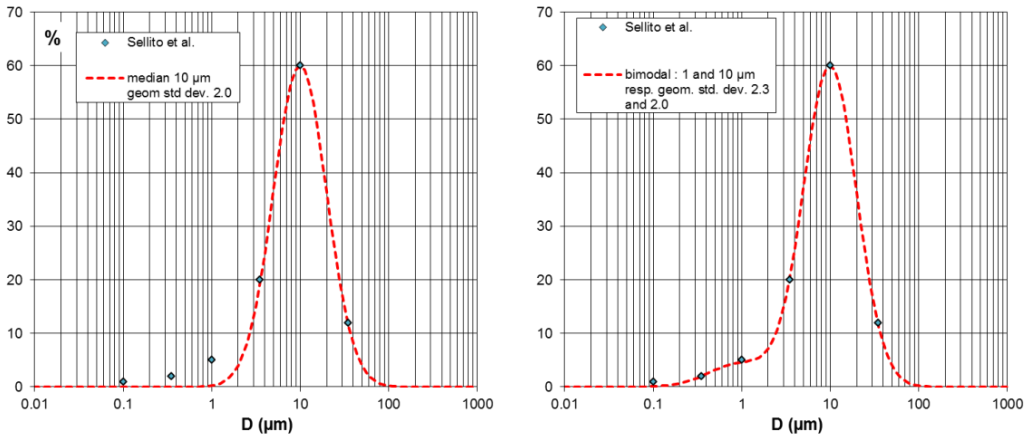

Figure 1: Rough adjustment of the particle size distribution proposed in table 1 of the paper (blue dots), by a monomodal lognormal with a geometric mean diameter of 10 µm (left), and by adding a second mode to fit the tail of the distribution at small sizes (right).

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?

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 SO_2 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).

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.

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.

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.

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.

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

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.

13. Figure 4: reminding the ratio between SO_2 concentration and volume mixing ratio would be useful in the legend since Fig. 4a and 4b use those different units; it is strange that Fig. 4b shows SO_2 above 10 km when none is visible in Fig. 4a; adapting the scales so that the maxima of SO_2 above 10 km in the middle of the transect in Fig. 4b also appears in Fig. 4a would be wise.

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

Technical comments:

Page 31337:
-Line 6-7: “by means of the FLEXible PARTicle Lagrangian dispersion (FLEXPART) model”.
-Line 15: remove “occurred” (or add “that” before).
-Line 16: specify “the column average aerosol particle size distribution”.

Page 31338:
-Line 12: “A number of studies show”.
-Line 19: “changes in”.
-Line 20: specify “aerosol particle size”.
-Line 21: “mainly through gas-to-particle conversion of SO_2”.

Page 31339:
-Line 14: “of its quasi-continuous”.
-Line 19-20: rather shift the first sentence of the new paragraph to the end of the previous one.

Page 31340:
-Line 19: “that occurred on”

Page 31341:
-Line 5: “at the ground station of Lampedusa, one of the”.
-Line 6 and 8 (see minor comments): section 6.4 deserves to become section 7, which means the conclusion section would become section 8.
-Line 7: “of the aerosol plume”.
-Line 14: “onboard both the NASA”.

Page 31343:
-Line 19: “Le Gléau” is missing a space.

Page 31344:
-Line 8: “using two IR, a window, and a sounding channel (13.4, 7.3, or 6.2 μm)” is rather unclear; please reformulate.

Page 31345:
-Lines 6-7 (and possibly elsewhere): use italic style for the symbols.
-Line 13: does this range apply to volcanoes around the world? Can you provide appropriate reference(s)?
-Line 18: “confirm this assumption”.

Page 31346:
-Line 22: “for the resolution” seems more appropriate.

Page 31348:
-Line 1: “A Brewer double monochromator”.
-Line 11: “details). The total aerosol optical depth is separated into”.
-Lines 13-14: “are derived”.
-Line 26: “at about 12:45”.

Page 31349:
-References INGV 2013a and 2013b should apparently be inverted to cite first 2013a in the text.

Page 31350:
-Line 8 “estimates from”.
-Line 17: “suggests”.

Page 31351:
-Line 3: shift this sentence to the end of the previous paragraph.
-Line 4: insert a space between the units “t km”
-Line 4: “South”.

Page 31352:
-Line 9: “tens of km”.
-Line 10: “northwards”.
-Line 14: suppress the paragraph end.

Page 31353:
-Line 3: I'd rather write "It is worth noting".

Page 31354:
-Line 8: "allows us to".
-Line 20: "tens of km".
-Line 20: "North of".

Page 31355:
-Line 2: "central-southern".
-Line 18: "about ±2.0 DU".

Page 31356:
-Line 19: "there is no other case"

Page 31358:
-Line 21"properties and their evolution following".

Page 31359:
-Line 2: "This allows us to".
-Line 28: the ranges of RFE values given in table 2 are worth being also given in the text.
Pages 31359-31360 (and possibly elsewhere):
-use italic style for the ratio "*f*".

Page 31360:
-Line 18: "major" seems to me more appropriate than "important".
-Line 20: Conclusions would become section 8.
-Line 24: "eruption of Mount Etna in late October 2013 by".

Page 31361:
-Line 17: you probably mean "transport modelling".

References:
-P. 31362: Anderson (1986) must be completed.
-P. 31364: in the two references of Derrien and LeGléau, add a space in "Le Gléau"; there is another instance in the reference of Sèze et al. (p. 31370).
-P. 31366: journal is missing in Kerr et al. (1985).
-P. 31367: Mallet et al. is now in press in Atmos. Chem. Phys.
-P. 31368: the two references of McCormick et al. should be inverted to respect the chronology.

Figures:
-Figure 1a: it would be nice to add circles to locate Mount Etna and Lampedusa Isl.
-Legend of figure 3b: trajectory *T* should be introduced here.
-Legend of figures 3b and 5:"Violet-grey to red" and "low to high".
-Legend of figure 7: "Blue to red" and "low to high".
-Legend of figure 8: refer to the preceding figure(s) for the trajectory *T*; a space seems missing between units µg and m^{-3}.