Answers to Review3 of paper “First results of the Piton de la Fournaise STRAP 2015 experiment: multidisciplinary tracking of a volcanic gas and aerosol plume”

General and specific comments:

Comments from Referee1. This paper would benefit from reorganization with an aim toward concise communication of the study objectives, methods, results, and interpretation. Study objectives are stated a few times throughout the paper with slightly different levels of detail and emphasis (e.g. p., 3 L19-25, p.2 L31-33, P. 17 L17-19). A careful content and english language edit would help cut down on redundancy, and tighten up the narrative. Attention to consistent use of language, terms, and nomenclatures through the different sections would help the readability. Decide on one spelling for sulfur versus Sulphur, for a single date format, etc.
The paper is interesting and exciting, but is hard to digest in its current format. The introduction could be condensed, as there is extraneous information.

Author's response

We have done our best to improve the English and your help was strongly appreciated (thanks for the supplement comments). We have tried to clarified the general objectives of more interest to the atmospheric community (p2 L31-32), the purpose of the paper (p., 3 L19-25), and deleted the repetition of the paper’s objective in the conclusion(P. 17 L17-19).
We have taken more attention to present a more consistent spelling and format.
The new version of the paper has also been read by a native English speaker.
We agree that the paper will benefit with a re-organisation by separating the methods, results and discussions. It has been done taking into account the recommendations of both reviewers 2 and 3. Now the paper is constructed as follow:
1 – Introduction : The introduction has been shortened.
2 - Description of the 2015 STRAP campaign on Piton de la Fournaise:
We though that it is important to give in this section information about Reunion Island (meteorology conditions and topography), the Piton de la Fournaise volcano characteristics, and to summarized the 4 eruptions of the STRAP campaign. The section 3.1 of the previous version has been condensed.
3 – Methods, models and measurements
We have introduced a subsection named “Campaign management” to summarize the section 2.2 and to point out the location of the main sites of observations.
We agree that most of the affiliations of 2.2 are not necessary in the text; they have been deleted and put in the acknowledgements.
A subsection “Flexpart modelling” corresponding to section 4.1.
A subsection named “Measurements near the plume source”: this part integrates the description of the methods and instrumentation, previously introduced in section 5.
A subsection “Measurements of the physical and chemical properties of the plume” which contains the technical elements and measurement methods introduced in the previous sections 6, 7 and 8.
4 - Preliminary results
The results have been separated into three subsections of results and figures descriptions.

“Simulation of the regional distribution in 2015”: this part corresponds to section 4.2
“Plume geometry and gas emissions at the volcanic vent”: this part corresponds to section 5 excluding the technical elements introduced in the new section 3.
“Examples of volcanic plume distribution and chemical properties”: this part groups the results of distal plume measurements at (sections 6, 7 and 8 of the previous version).
5- Discussion
This new section has been purposed by both reviewer 2 and 3. This section contains the discussion of results previously introduced in the conclusion.
6 – Conclusion
The conclusion has been modified and place the new observations made from measurements presented in this paper into the context of past studies at Piton de la Fournaise and other volcanoes.

Comments from Referee

2. Gas section (section 5) and references to gas measurements. The plots and interpretation in this section could use some revision and clarification
a. In plot 6, I don't see the pulse of SO2 observed at the end of phase 1 (noted in the section text and conclusions). Since Novac data can have strong anomalies due to atmospheric effects, wind, etc., it would be good to corroborate the novac data with emission rates from the mobile DOAS from Sept. 7, 11, 18 to confirm your observation. Plotting all the mobile data on figure 6 seems important.
b. It seems that the data in fig. 6 plot would be much easier to see if it were a scatter plot rather than column plot. E.g. in conclusions “During most of the eruption, SO2 fluxes have been lower than 1.5-2 kt day−1.” It is actually hard to see that the red columns are in that range because of the error bars, which focus your eye on the max error bar value rather than the data points. Or is there some other reason you have it as a column plot?
c. Figure 6. caption: ‘The uncertainty comes from the spectroscopic retrieval, radiative transfer, wind direction and speed, and plume height. This uncertainty is used in the computation of the daily mean values as presented in Figure 5.’ Can you explain how this was done? Both the calculation of the uncertainty, and how it is used to calculate the daily mean values? Or send readers to a reference, if it is published elsewhere?

Author's response

a. SO2 fluxes obtained by portable DOAS and calculated using the Salerno et al., (2009) approach have been added to Table 1. Even if it is tricky to compare average daily fluxes (NOVAC; fig. 5) and multiple daily scans (NOVAC; fig. 6) with single portable traverses, both methods are in reasonable good agreement (Table 1) , as it can be seen in Figure RC3 below:

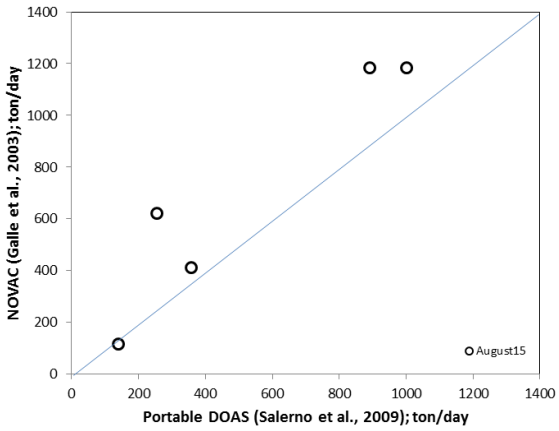

Figure RC3. Comparison of traverse mini-DOAS ('Portable DOAS') measurements with stationary scanning-DOAS ('NOVAC') measurements obtained on August 15 2015, evaluated with the methods mentioned in the manuscript. The correspondence is quite good and within the uncertainty of the measurements.

a, b. The referee points out correctly that Fig. 6 shows large uncertainty ranges and that, based on this figure, the interpretation of changes of activity seems questionable. However, the best estimate of daily SO_2 emission, on which the interpretation of eruptive activity is based, is that of Fig. 5. The reason why they seem to differ, is that Fig. 6 shows the results of individual scan measurements that detected the plume. A careful uncertainty analysis was performed for each individual scan measurement, because of highly changing measurement conditions. For example, a plume can be observed completely above the horizon in one scan, and then decrease in altitude some minutes later affecting the accuracy of the flux measurement. Presenting all scans with their uncertainty makes look the plot dominated by those measurements with large uncertainty (notice that a single day may have up to ~50 scans), but we think the plot actually shows that the uncertainty varies among measurements and that some may indeed be quite large, presenting a challenge for interpretation. To compute a reasonable estimate of the daily mean value and its standard error (shown in Fig. 5), all valid scan measurements within a day are combined, weighting them according to their individual uncertainties, as explained succinctly in the caption of Fig. 6. By these approach, not only the mean value is more representative of the daily emission, but also the standard error accounts for the fact that the larger the number of validated measurements, the more representative the statistic. In any case, we have remake Fig. 6 changing scale and using scatter points instead of columns, for better readability.
c. There is an unpublished PhD thesis (Arellano et al., 2014, Chalmers University of Technology) describing in detail the methodology behind the uncertainty calculation and the computation of daily statistics. Each scan has its own uncertainty analysis based on sampling of distributions of error for each of the variables (column density, wind speed, plume height, plume direction). The daily average is calculated as a weighted mean that favours measurements with lower uncertainty. The standard

error is calculated taken into account the individual uncertainties and of course the number of valid measurements on each day. We did not abound in details in the manuscript to avoid giving too much emphasis to this technique, but included Fig. 5 to show that obtaining daily statistics is not a simple matter of averaging measurements because the quality of the measurements may vary considerably even within a single day.

Comments from Referee

i. The labeling/notations on the 2 FLIR images are inconsistent with each other, and would be better if they were similar (e.g. you might have a single box for the max pixels in the image like for the bottom left image)

ii. Can you say something about the FLIR images, rather than just present them? Are they included to emphasize the less vigorous eruption during May as compared to august? Or is there another point you are wanting them to demonstrate?

iii. The Photo beneath the multi-gas plots detracts from the data plot, and should either stand on its own if you feel it is showing something of importance, or remove it. The plot axis labels cannot be read easily on the MultiGAS plots, and need to be increased in size, and the plots presented in a larger format. Can you explain the trend in the different species, and if you think the concentrations make sense based on the plume traverse? e.g. Should the SO2 and CO2 anomalies be better correlated if they are from the plume, or are the instrument response times contributing to the lack of coincidence of peaks? Might you plot the C/S and H2O/CO2 that are described in the text? It is hard to take away anything from these plots in the current presentation.

iv. Are there some interesting differences in the multi-gas data for the 2 different eruption regimes (May versus August-October)? might you show the data more clearly and completely since the text emphasizes this gas data?

v. Important to add emission rate for the SO2 column amount profile plot. While this profile is interesting for people familiar with the technique, a plot of the mobile doas emission rates for the long eruption seems important in addition to this column amount plot.

Author's response

Fig. 7 has been modified.

i. Two new IR images of the beginning of the August 2015 eruption have now been included with the aim at highlighting the fast evolution from linear to spot source for the plume emission. The text has been modified accordingly.

ii. The MultiGAS figure has been changed; the original image of the results obtained by helicopter flight has been replaced with a typical ground based measurement performed in near field close to the high temperature source; correlated peaks in MultiGAS measurements have been evidenced.

iii. MultiGAS data show relatively moderate change in time, as discussed in the text; however their detailed presentation and interpretation is the topic of a distinct paper (in preparation) which integrates a larger geochemical dataset (bulk rocks; melt inclusion; mineral phase equilibria; gas fluxes and molar ratios)

iv. SO2 emission rates estimated using mobile DOAS have been reported in Table A2.

v. Emission rate has been on the SO2 column amount profile plot (Fig. 7).

Comments from Referee

3. Conclusions

a. The discussion of the preliminary data, and the relationship of the various data sets to each other, deserves its own section.

b. The emission rates for CO2 and H2O are not reported in the paper, although it is referred to in the conclusion. It seems a table with the reported values scattered through ut the paper, and repeated in the conclusions could help the reader (gas emission rate data, Lidar coefficients, LR, particle numbers, etc.). I think such a table could be useful for others looking into plume dispersion and chemistry at their own volcanoes.

4. References – since you refer to radiative transfer a couple of times in the paper, it would be good to add a reference. Kern, C. et. al, 2012 (or other).

Author's response

The discussion of the preliminary data has been now put in a separate section (section 5).

It is complicated to summarize all the observations in one table due to the disparity of measurements types and their duration.

We have chosen to introduce four tables (two in the main text table 1 and table 2 and two in the appendix table A1 and table A2), and one figure for the Maido observatory (permanent observation). It is not possible to summarize in one table all LIDAR measurements (more than 600 profiles). A dedicated paper is in preparation.

We hope that the re-structuration of the paper will be clearer for readers.

The reference to Kern et al., 2010 has been added in the corresponding mention to radiative transfer effects and the reference list.

Minor comments:

Comments from Referee

1. It would be helpful for the maps to have a N arrow and a scale

Author's response

The new Figure 1 have a scale in the bottom part of the picture and a N arrow.

Comments from Referee

2. Since you are reporting SO2 to 1 ppb, you might want to state the sensitivity and resolution of the pulse fluorescence SO2 analyzer.

Author's response

The limit of detection of the SO_2 analyzer is 50 ppt (0.05 ppb). This instrument is used for air quality studies and it is able to measure low level concentrations (e.g. at the free troposphere or rural areas).

The sentence has been modified

Author's changes in manuscript

The new sentence is: “Gas phase measurements of sulphur dioxide were made using a UV Fluorescence SO2 Analyzer (Teledyne, model T100U), which relies on pulsed fluorescence and has a detection limit of 50 ppt (i.e. 0.05 ppb).”

Comments from Referee

3. p. 14 L 16-18. Your use of the terms ‘course’ and ‘fine’ to describe your particle size cut is unconventional for most of us who think of fine particles as PM2.5. Could you quality your description with a caveat like ‘course particles as defined in this study’? Or use some other term to refer to the two size fractions you are discussing?

Author's response

In atmospheric aerosol science fine particles are related to sub-micron-size particles (PM1 with diameter < 1µm). Ultra-fine particles characterize aerosols with a size below the accumulation mode (diameter < 100 nm). The coarse particles have diameter greater than 1 µm.

The AIS and the nanoCPC system installed at the Maïdo observatory are able to count particle number at 5 nm (nucleation mode). We agree that we are not in the range of particles usually observed in volcanic sources (e.g. supermicronic size - coarse mode for ashes). This is the reason why we have given precise size information in the paper, as follows:

- ultrafine particles (Dp < 100 nm)
- fine particles (Dp < 1 µm)
- coarse particle (Dp > 1 µm)

Author's changes in manuscript

New sentences are:

“Meanwhile, there was a moderate increase in coarse particle concentrations (particle diameter Dp > 1 µm).”

“It is very likely that the particles in the volcanic plume were generated by oxidation of volcanic SO_2 and subsequent particle nucleation or by condensation of volatile compounds onto pre-existing fine particles (Dp < 1 µm).”

“The morning advection of a relatively wide range of ultrafine particles (Dp < 100 nm) to the Maïdo station indicates that nucleation and early growth takes place already at the vicinity of the crater, and continues within the plume at least up to the Ma\"ido station.”

Comments from Referee

4. P. 14 L22. Figure 10 suggests SO2 is west of the vent, so the text is confusing since it states ‘east’.

Author's response

It was an error (thanks). The sentence has been corrected in the new version.

Comments from Referee

5. P. 14, L33. Do you mean ‘volcanic aerosol-free air masses’? Otherwise, it is confusing - since particle size distribution in aerosol-free air masses doesn’t make sense.

Author's response

We mean “volcanic aerosol-free air masses”. Thanks for this remark. Is has been corrected in the new version.

Comments from Referee

6. The red text on figure 1 is not legible. Can you use a color that more strongly contrasts, and with better resolution?

Author's response

You are right. The colour of the text has been changed to white and the size of the figure increases.

Comments from Referee

7. P. 16 L 7-8. Can you reorganize this sentence so that it is clearer? You could start the sentence with 'Examples of the evolution. . ..' And omit the first 5 words.

Author's response

The sentence has been modified.

Author's changes in manuscript

Examples of the fast growth of cluster ions to larger sizes can be followed on the SMPS size distributions up to 50 nm on 1-2 September, and 100 nm on 20-21 May.

Comments from Referee

8. Figure 2. Might you Label the contour lines with elevation, for people not familiar with the topography? Fig.1 helps, but you could help your reader out by labeling it in fig. 2.

Author's response

It has been done for figures 2, 3, 8, 9 and 10. Now dotted lines represents in red the topography at 2000 m asl and in black the topography at 1000 m asl.

Comments from Referee

9. P. 16 L21-22. The wording of this sentence is unclear as you seem to be calling the sulphuric acid the precursor gas.

Author's response

The word precursor is deleted.

Author's changes in manuscript

The sentence is now:

"Due to its low saturated vapour pressure under typical atmospheric temperatures (Marti et al., 1997), the common assumption in the scientific community is that the sulphuric acid is the main gas responsible for the nucleation processes."

Comments from Referee

10. Can you mention the double maxima modelled in fig 9 bottom left in the final sentence of section 6? Or is it explained somewhere else? What might cause that?

Author's response

This double maxima is related to the modification of wind intensity above the vent. So the volcanic air mass loads different quantity of volcanic pollutant during its passage above the emission aera.

Author's changes in manuscript

The new sentences are: "On 2 September 2015, the plume was forecasted to be located north-west of the volcano. Two maxima were modelled by FLEXPART (above

the OVPF and above the Maïdo area) in relation with the evolution of the wind intensity above the vent."

Comments from Referee

11. For plots, state in captions or axis label if altitude is agl or asl

Author's response

It has been corrected.

Comments from Referee

12. Is the 6.8 kt/d SO2 data point noted in the conclusion (and in the earlier text) on the plot?

Author's response

Figure 6, which shows individual scan measurements shows these values, as discussed in the text. See above for changes done on this figure for better readability.

Technical comments:

Comments from Referee

1. Identify acronyms with first use. While some sections do a good job of this, the Introduction needs attention. The subsequent sections don't have to repeat it, but watch for how the different authors use the acronyms so there is consistency throughout the paper.

P. 15: ASQUA, ACTRIS – are these defined somewhere?

Author's response

ASQUA, VACC are deleted. ACTRIS is now defined.

Author's changes in manuscript

"The quality of the DMPS measurements was checked for flow rates and relative humidity according to the ACTRIS (Aerosols, Clouds, and Trace gases Research InfraStructure Network) recommendations (Wiedensohler et al., 2012)."

Comments from Referee

2. L26 p.3 –Do you mean topography rather than morphology?

Author's response

Yes, it has been corrected.

Comments from Referee

3. L14-15 p. 4 – suggest revision of sentence: The Observatoire Volcanologique du Piton de la Fournaise (OVPF/IPGP) manages the monitoring networks on the island, allowing the observatory to follow eruptive and specific volcanic events, and to describe their time and space evolution.

Author's response

It has been modified.

Comments from Referee

4. L 17 p. 4- replace Internationals with International

Author's response
Thanks, this sentence has been deleted in the new version.

Comments from Referee
5. P. 12-13, look carefully at the use of the word ‘aerosols’ versus ‘aerosol’ in this section.
Author's response
We have corrected it in the text.

Comments from Referee
6. P. 13 – both UTC and local time are provided in the discussion which is helpful. Consider doing this in key sections where you are describing a process that is dependent on diurnal orographic meteorology.
Author's response
It has been done.

Comments from Referee
7. Global replace of ‘pick up’ with pick-up or ‘pick-up truck’
Author's response
In the new version we have changed “pick up” by “pick-up truck”.

Comments from Referee
8. Caption for fig. 10 – recommend clarifying sentence 2. “The flight path is coloured as a function of the measured...”
Author's response
Thanks, the sentence of the caption has been modified.

Comments from Referee
9. P.16 L26. This sentence needs to be clarified. ‘. . .because it depends whether the volcanic plume arrives at the station.’ Do you mean it depends on ‘when’ it arrives? Or ‘when and if’ it arrives?
Author's response
We agree that the sentence was unclear. The new sentence is “Unlike other parameters, for instance anthropogenic pollutants, the SO_2 concentration variation is not periodic because it depends on whether the volcanic plume is advected to the station or not

Comments from Referee
10. P. 16 L33-34. This sentence needs reorganization and grammar corrections.
Author's response
The sentence has been rephrased as “For the case of 20 May, it is possible that newly formed particles are grown by condensation to sizes above the detection limit of our instrumentation.”

Comments from Referee

11. P. 17 L2-4. This sentence needs to be rewritten, as it is very hard to follow.

Author's response

The sentence is now written as: “Then the variability of the correlation between the new particle formation rate and sulphuric acid will be further studied for other case studies. This will allow to derive, for the first time to our knowledge, a parameterization of nucleation rate specific to volcanic plumes.”

Comments from Referee

12. Global replace ‘researches’ with ‘research’

Author's response

Thanks, it has been done.

Comments from Referee

13. Figure 14. It would be kind to your readers to label the DMPS and AIS panels more clearly. Also, might want to make scale label and caption consistent (chose either cm-3 or #/cm)

Author's response

The Figure 14 has been modified. We have chosen cm^{-3} to be consistent with the rest of the paper.

Comments from Referee

14. Alternate wording suggestions have been included in a pdf version of the manuscript for many technical issues, but will not take the place of a through English language edit.

Author's response

Thanks for this work. It was very helpful. We have done our best to improve the English and the new version of the paper has now been read by a native English speaker.

Please also note the supplement to this comment:

http://www.atmos-chem-phys-discuss.net/acp-2016-865/acp-2016-865-RC3-supplement.pdf

Comments from Referee

(Table A1): the numbering of this table seems unusual.

Author's response

This numbering came from latex Copernicus package (it is automatic). So it is the format asked by the ACP. Table 1, 2 are numbering for the main text whereas A1, A2 corresponds to appendix.

Comments from Referee

“One explanation could be attributed to a subsidence...” : grounding? or sinking? subsidence generally refer to solid surfaces

Author's response

In meteorology the term “subsidence” is largely used to refer to downward transport of air masses.

Comments from Referee

“One can also notice a fresh crossover of aerosols plume starting at 11 UTC, credibly coming directly from the vent." : introduction?

Author's response

We are not sure to understand this comment placed in the supplement document.

Author's changes in manuscript

We purpose to rephrase as: “The LIDAR backscattered signal increases from 2.2 up to 2.6-3.5 (au) between 0 and 500 m agl at 10:30 UTC (14:30 *h* LT) until the end of the measurement period. This shows the passage of a freshly emitted aerosols plume likely coming from the vent.”