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Atmospheric Chemistry and Physics Discussions

Interactive comment on "New particle formation in the active volcanic plume of the Piton de la Fournaise: specific features from a long-term dataset" *by* Clémence Rose et al.

Anonymous Referee #1

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Processes of formation and growth of secondary aerosols, as well as their contribution to the total number of particles (compared to the contribution of primary aerosols) and to the formation of cloud condensation nuclei, are relatively poorly-known in a tropospheric volcanic plume. This study takes advantage of a year-long dataset collected at the high-altitude atmospheric observatory of Maido (La Réunion Island), which was impacted by three eruptions of Piton de la Fournaise volcano in 2015, to investigate such important processes using a statistical approach.

This rich dataset includes ground-based in-situ observations from three complementary instruments: (1) UV fluorescence analysers providing SO2 mixing ratios indicative

of the presence of the volcanic plume, (2) from a Differential Mobility Particle Sizer (DMPS) providing information on the occurrence of new particle formation (NPF) and particle growth rates in the range 10-600 nm, and (3) from an Air Ion Spectrometer (AIS) informing on early stages of NPF.

The paper is well written, figures are clear and interesting data are presented. Given the year-long dataset, the authors can properly determine the occurrence of new particle formation and the typical growth of particles over periods of time which are not impacted by volcanic eruptions. Therefore, they can better identify the impact of volcanic eruptions compared to background conditions. They show the impact of volcanic eruptions on the occurrence of NPF, the importance of photochemistry and the respective contributions of volcanic primary and secondary aerosols on the size distribution of particles. The volcanic impact on particle growth rate seems less obvious however. Results are of strong interest for the atmospheric science community. Nevertheless, as developed in the following, a more robust methodology would be required in some areas to strenghten the approach and strongly support the results:

- Classification of plume- vs non-plume days :

As the classification is critical for the statistical study, more details and illustrations are missing to describe and validate the classification of plume- vs non-plume days. As SO2 represents indeed a clear tracer of the volcanic plume, a time series of SO2 mixing ratio values at Maido with highlighted volcanic events would be welcome in order to evaluate the amplitude of background variations in SO2 mixing ratios.

As this is the root of the paper, an illustration with AIS and DMPS observations for one representative strong plume day and one weakly influenced plume day before statistical representation of Fig. 1 would be required.

- Selection of plume-days, page 7, lines 19-27 :

If I understand correctly, selected days are considered as 'plume-days' when at least

one of the hourly averages of the SO2 mixing ratio exceeds 1 ppb over the 5 hours of interest each day (between 6 :00 and 11 :00 LT). The volcanic plume was detected during the 5 hours of the time window of interest for only 20 of the 36 'plume-days'. I am wondering if the authors should not restrict their study to these 'fully volcanically-influenced days' ? If not, they should assess the impact of mixing in their study 'plume-days' hours without any volcanic plume. This choice may artificially tend to decrease the difference between plume- and non plume-days.

- Start time of NPF events, page 9 :

Why are the detection and evaluation of the start time made by a visual inspection ? What is the difficulty in automating the detection of a concentration increase in the 1.5-2.5 nm range ? Visual inspection is subject to large uncertainty and raise questions on the accuracy and reproducibility of the obtained results. An illustrative example would be also welcome to see how strong are the AIS/DMPS signals for days only poorly-contaminated by volcanic plumes.

- Particle growth rate, pages 10-11 :

Page 10, lines 3-8 : The authors do not highlight any impact of the volcanic plume on the particle growth rate between 12 and 19 nm. The interpretation of the authors is that it may be difficult to clearly identify the impact of volcanic plumes at the Maido Observatory as the atmospheric dynamics is complex around this site and there may be an importation of growing particles likely transported to this site. The same processes (of imported particles, including potentially biomass burning aerosols as mentionned for CS variations in Sept and Oct) could also bias the observations of J2 and J12 ? The authors should comment on this and propose some solutions to 'clean' data by removing periods with strong influences by other sources of aerosols (urban, biomass burning, etc..).

Clearly higher values of J2 and J12 values are not observed under volcanic influence for the month of Sept. A clear volcanic signature is not identified either for J2 values

for the month of Oct (with also surprisingly very spread J2 values for strong plume days). The authors should describe these discrepancies in the text and provide some interpretations or suggestions of interpretation (impact of biomass burning activities, or others ?).

By contrast, page 10, lines 9-14 : why is observed in May so much increase in J2 and J12 values for plume-days compared to non-plume days ? Is there a specificity of the volcanic events, or of the meteorological conditions occuring in May? Opposite case: why is not observed an obvious distinct behaviour of plume-days in Oct ?

More generally, the authors should discuss the advantages and also the disadvantages or limitations to have data collected at a high altitude atmospheric observatory and the potential biases that may affect the results at such a site (including complex atmospheric dynamics, fluctuating relative humidity, is it easier or not to identify imported species, a less polluted background or not, etc. . .).

- Which is the impact of relative humidity on NPF? As relative humidity is measured at Maido, is there any correlation with NPF? Are observed higher RH values during plume-days (as the Piton de la Fournaise plume may be rich in volcanic water vapour) or not ?

- Impact of condensation sink (CS) page 11 :

Right of Fig. 3 : in the plot of CS vs SO2 mixing ratio for plume-days and strong-plume days, could it be added non-plume days to assess if obvious differences are observed between plume- and non plume-days in this representation ?

Left of Fig. 3 : how do the authors explain the large CS observed in Sept and Oct for non-plume days ?

- Relationship between J2 and [H2SO4], page 12 :

According to Fig. 4, a correlation relationship between J2 and [H2SO4] is not obvious : data points are very scattered, as illustrated by the very low value of R2 of 0.21 and

0.11 for all plume and strong-plume days respectively. In this context, is it meaningful to try to fit anyway a correlation relationship and estimate k and a coefficients?

Moreover, except higher concentrations of H2SO4, data associated to strong-plume days do not seem to present a very different relationship between J2 and [H2SO4] (Fig. 4a). The weak difference in the relationship which is retrieved seems just to result from the influence of 3 points, potentially outliers ? If these points would have been represented in black, and not in yellow, it would be very difficult to distinguish any different behaviour.

Minor comments :

- 'Active volcanic plume' : I do not understand this term. Given lines 31-32 in the introduction, I am wondering if the authors may want to refer to a volcanic plume emitted during an eruption compared to passive degassing emitted out of eruptive periods. If so, please refer rather to 'volcanic eruption plume'

- Page 3, lines 5-8 : 'primary particles are fragment of ash while secondary particles. . .' : Volcanic primary particles do not include only ash particles but also sulfate aerosols, as illustrated by near-source measurements (e.g. refer to first publications on this matter which include Allen et al., 2002 ; Mather et al., 2003, 2004, etc..).

- 'Here we report observations of NPF performed at the high-altitude observatory of Maïdo (2165 m a.s.l., La Réunion Island) between 1st January and 31st December 2015. During this period of time, 3 effusive eruptions of the Piton de la Fournaise, located 39 km away from the station, were observed and documented, resulting in 36 days of measurement in volcanic plume conditions to be compared with 250 "non -plume days'. 250 + 26 = 276 days, what happens with the missing 89 (=365-276) days ?

- There are many references to a study in preparation (Sahyoun et al., in prep) which is presented as an earlier work: has this paper been submitted to a journal with open

discussion where it would be accessible or has it been published since then? If yes, please update so that the reader can have access to this manuscript.

Text :

- Abstract is very long, if possible you should try to shorten it (possibly remove the mention to the correlation relationship between J2 and H2SO4 concentration which does not seem obvious (as developed above)

- Please reformulate these sentences for clarity :

- abstract, Page 1, line 17 : 'as those form the baseline to calculate..'

- abstract, Page 1, line 30 : 'recorded in the different conditions': recorded in the different conditions described thereafter..

- abstract, Page 1, line 26-27 : 'compared to non-plume days, during which condensable species were in contrast transported from lower altitude by the mean of convective processes' : it is difficult to understand the meaning of this sentence if we have not read the manuscript yet

- Page 2, lines 21-22 : 'the radiative forcing. . . still has a large uncertainty'

- Page 11, line 16 : 'loss rate of the vapours' ? What do you mean by 'vapours' ?

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