Interactive comment on "High occurrence of new particle formation events at the Maïdo high altitude observatory (2150 m), Reunion Island (Indian Ocean)" by Brice Foucart et al.

Manuscript published for discussion on 27 September 2017.

The authors would like to thank the reviewers for their very constructive and informative comments. These comments and suggestions have helped us to improve the quality of our manuscript, including strengthening our conclusions with additional evidences of likely precursors. Below we have responded to each of the reviewers comments. **The reviewers comments are in bold** and our responses are in normal text. Please, note that the structure of the article has been modified (especially for 4.1 and 4.6 sections), several figures had been replaced by others and we added new ones.

The new plan is as follow:

Abstract

----- 1 Introduction

----- 2 Characteristics of the Maïdo observatory

---------- 2.1 Geographical location and networks

---------- 2.2 Large and local scale atmospheric dynamics

---------- 2.3 Potential gas-phase precursor sources

---------- 2.3.1 Sulfur dioxide (SO_2) and sulfuric acid (H_2SO_4)

---------- 2.3.2 Ammonia (NH_3) and amines

---------- 2.3.3 Volatile Organic Components (VOCs)

---------- 2.3.4 Phytoplankton

---------- 2.3.5 Biomass burning

---------- 2.4 Instrumentation used

----- 3 Calculations

----- 4 Results and discussion

---------- 4.1 Dynamics of the NPF events at Maïdo observatory

---------- 4.2 Nucleation and frequency analysis

---------- 4.3 Particle formation, growth and nucleation rates

---------- **4.4 Meteorological parameters and onset of NPF**

---------- **4.5 Condensation sink**

---------- **4.6 Black Carbon as a tracer of the anthropogenic contribution**

5 Conclusions

Acknowledgments

Figures modifications:

The Figure 3 has been modified: We added the BC dataset

The Figure 4 is now composed of two figures with **a)** July 6th 2015 diurnal variation of negative ions (1-10 nm) and (10-700 nm) aerosol particle size distribution (note the different concentration scales for ion number and particle concentrations) and **b)** the BC concentration variation in $ng.m^{-3}$

The Figure 5 (DMPS spectra for 31 January and 25 March) has been displaced to the supplementary as **Figure A2**. **The Figure 5** is now "The average diurnal variation during winter and summer of the **a)** BC concentration, **(b)** number concentration of particles which diameter is larger than 100 nm (N_{100}) and **(c)** number concentration of particles which diameter is smaller than 30 nm (N_{30})".

We associated **Figure 11** to Figure **10** which is now composed of **a)** "The monthly CS_2 and event frequency" and **b)** "The monthly CS_{2prop} exceeding the average". The Figure 11 has been replaced by a new one.

We added a new **Table 2** which deals with "The R correlation coefficients giving the relationships between NPF parameters and influencing factors".

References modifications:

We added three new references which are:

- Mirme et al., 2007 at line 219
- Kulmala et al., 2001a at line 250
- Hermann et al., 2015 at line 304

Response to Anonymous Referee #1 comments (RC1):

Received and published on 11 November 2017,

R1-General comment: "The work includes the experimental observation and some analytical analysis for NPF events in Reunion Island. My major comment is that several statements in this manuscript are just speculations. Without further supporting evidences, the conclusions claimed here are very shaky."

AR-General comment: We thank reviewer 1 for his comments on the paper and have prepared a modified version of the manuscript that take these comments into account. In this version, we clearly state that we start with enumerating possible new particle precursors possible sources, and then bring some new evidences of some of these sources likely contributing to the process, the others staying only as possible contributors. We agree that this paper do not contain any precursor direct measurements, but it lays as a first description of the new particle formation occurrence in this area of the world which is very poorly characterized. We now avoid any speculations and we modified our conclusions.

The point-by-point response to **the detail comments** can be found below.

RC1-1: line 69, "at low or high altitude?"

AR1-1: We now specify high altitude

RC1-2: line 130, "Unfortunately, H_2SO4 was not measured" highlighted. Lines 143-144, "the major causes of SO2 emissions are connected with human activities: agriculture, power plants, sugar exploitation and road traffic" highlighted, line 155, "no-direct measurements highlighted"

AR1-2: It is directly linked to the major comment and no comment was associated so it was not clear what action should be taken by us on these portion of the manuscript.

RC1-3: line 169: "Format needs to be corrected"

AR1-3: We harmonized the format to "Yu et al. (2017)" and "Kirkby et al. (2016)".

RC1-4: lines 249-250: "according to Pirjola et al. (1999) Equation (2): it is not the definition of CS used in Pirjola's article" and "the unit is not 1/s (based on the formula)".

AR1-4: It is true that this equation is not exactly the same than in Pirjola 1999. In fact it can be found in the following reference.

Kulmala, M., Dal Maso, M., Mäkelä, J., Pirjola, L., Väkevä, M.,Aalto, P., Miikkulainen, P., Hämeri, K., and O'Dowd, C.: On the formation, growth and composition of nucleation mode particles, Tellus B, 53, 479–490, 2001a.

Pirjola was the precursor of the idea that was revisited by Kulmala (2001). We now refer to Kulmala 2001a (updated in the reference list).

We believe that there is no mistake concerning the equation unities. $\boldsymbol{D_{vap}}$ is in the unit of square divided by time ($m^{-2}.s^{-1}$), r is a length (nm to m conversion), $N(r)$ is a particle number for a volume ($part.m^{-3}$). At the end, CS is in s^{-1}.

RC1-5: line 256: "drained".

AR1-5: This a mistake and we replaced it by "dried", as proposed.

RC1-6: lines 265-266: "Kulmala et al. (2007)" highlighted, "The GR is in the unit of hr, and the J is in the unit of second. Please unify"

AR1-6: We agree that there are non-homogenous unities relative to time. By convention and for clarity, we do not put unities in the equation. However, the Kulmala 2007 is the good reference and the exact equation can be found in the supplement of their article.

RC1-7: lines 265-266: "What is the unit of the number 7 list here? Based on the closure of unit here, it should be dimensionless. But why is it? And what does it st and for?"

AR1-7: We made a typing error in that part of the equation. GR_{12-19} has to be divided by ΔDp which is the difference between the upper and the lower channel. Here, this is 19 – 12 = 7. The equation had been corrected in the manuscript and it is now:

$$J_{12} = \frac{dN_{12-19}}{dt} + CoagS_{12} \times N_{12-19} + \frac{GR_{12-19}}{7\ nm} \times N_{12-19}$$

RC1-8, line 272, Eq (5): "CoagS12 is in the unit of 1/s, based on the second term used in Eq (4). If so, the unit of the exponent term would be in length (eg. nm). However, this make no sense for physics."

AR1-8: We think again that there is no mistake concerning the equation unities here. J_2 and J_{12} should be in part.m^{-3}.s^{-1}. As a consequence the exponent term should be without unity. It is right because d2 is a length (m), multiplied by $CoagS$ (d2) which is a time (s^{-1}) divided by a GR_{12-19} which is a length divided by time (m.s^{-1}).

However, we made a typing error here. We replaced the d1 in the exponent term by d2.

$$J_2 = \frac{J_{12}}{\exp\left(-\gamma \times d_2 \times \frac{CoagS_{(d_2)}}{GR_{12-19}}\right)}$$

RC1-9, line 276: quote missing in the Eq (7).

It is done:

$$m = \frac{\log\left(CoagS_{(d_{12})}\right) - \log\left(CoagS_{(d_2)}\right)}{\log(d_{12}) - log(d_2)}$$

RC-1-10, line 291: "probably" highlighted, with the comment: "lots of uncertainty".

AR1-10: We do not have the means to evaluate if the station is in the free troposphere or not during nighttime, but we now provide indications that this is likely the case. We replaced the 28 August 2015 Figure 4 by the 06 July 2015 one (Figure 4a) which is not impacted by volcanic plume, we add the corresponding diurnal BC concentration variation (4b), and we modified the end of section 4.1 as reported here:

"The initiation of the formation of new particles at 06:00 UTC (10:00 LT) is followed by the appearance of accumulation mode particles. Further growth of the newly formed particles is generally accompanied by the simultaneous growth of the accumulation mode particles, starting around 07:00 UTC (11:00 LT), that are likely representative of the updraft of boundary layer air to the station. We computed the diurnal variation of black carbon (BC), a good indicator of any anthropogenic, hence boundary layer, influence. The corresponding diurnal variations of BC ($ng.m^{-3}$) is shown on Figure 4b. BC concentration clearly increases from 06:30 UTC (when ignoring early sharp peaks that may due to local contamination), which is half an hour later than the occurrence of the cluster mode particles. Hence we can hypothesise that boundary air convection to the site is a trigger for NPF events, most particularly when the interface BL/FT is sampled. At 07:00 UTC, as the accumulation particle concentration increase from 2000 to 8000 $part.cm^{-3}$, the BC concentration also increase to reach 630 $ng.m^{-3}$ at 09:00, when the BL is fully sampled at the site. At the end of the afternoon, the accumulation mode particles concentrations drop to less than 1000 $particle.cm^{-3}$ and BC concentrations drop to very low values. Most high altitude stations are strongly influenced by free troposphere air during nighttime regardless the season, but mostly during wintertime (Venzac et al., 2008; Rose et al., 2015a). This is also true for stations located in complex terrains such as Jungfraujoch station in the Swiss Alps (Herrmann et al., 2015) and at the Chacaltaya station in the Andes (Rose et al., 2015b). These are indicators that the station lays in the free troposphere at night. The Aitken mode particles present during nighttime at the station are hence likely present in the free troposphere and are sampled at the site in subsiding air masses (Tulet et al., 2017).

These features can also be observed on average, both for the summer and winter seasons. BC average diurnal profiles (a), together with the average diurnal variation of the number concentrations of particles larger than 100 nm (N_{100}) (b), and the number concentration of nucleation mode particles with a diameter smaller than 30 nm (N_{30}) (c) are shown on Figure 5. We observe that, on average, BC concentrations increase in the morning at the same time as N_{100} and N_{30}, confirming the influence of the BL on the occurrence of NPF events at the scale of the season. Moreover, we can note that during winter, BC concentrations are higher during nighttime (from 16:00 to 06:00 UTC) than during summer. This observation is also true for N_{100} with higher values from 17:00 to 02:00 UTC during winter compared to summer. We assume that during winter, trade winds favour the large scale remote primary particles transport to the Maïdo station"

Herrmann, E., Weingartner, E., Henne, S., Vuilleumier, L., Bukowiecki, N., Steinbacher, M., Conen, F., Collaud Coen, M., Hammer, E. and Jurányi, Z.: Analysis of long-term aerosol size distribution data from Jungfraujoch with emphasis on free tropospheric conditions, cloud influence, and air mass transport, J. Geophys. Res. Atmospheres, 120(18), 9459–9480, 2015.

"Hermann et al., 2015" has been added to the reference list.

RC1-11, lines 305-306: Sentence "As shown in Figure 5. Similar seasonal trends were observed for the nucleation frequency in 2014 and 2015" highlighted with the comment: **"It is not very convincing to make the conclusion of seasonal trend based on only one and half year observation."**

AR1-11: Many studies reporting on a seasonal variation for NPF do not have more than one year and a half of data (Rodríguez et al., 2005; Suni et al., 2007; Rose et al. 2015; Berland et al., 2017; to mention only a few). We believe that one year and a half of data allows to derive a seasonal variability, especially when this seasonal variability is reproducible from one year to the other for the common months.

Berland, K., Rose, C., Pey, J., Culot, A., Freney, E., Kalivitis, N., Kouvarakis, G., Cerro, J. C., Mallet, M., Sartelet, K., Beckmann, M., Bourriane, T., Roberts, G., Marchand, N., Mihalopoulos, N. and Sellegri, K.: Spatial extent of new particle formation events over the Mediterranean Basin from multiple ground-based and airborne measurements, Atmospheric Chem. Phys., 17(15), 9567–9583, doi:10.5194/acp-17-9567-2017, 2017.

Rodríguez, S., Van Dingenen, R., Putaud, J.-P., Martins-Dos Santos, S. and Roselli, D.: Nucleation and growth of new particles in the rural atmosphere of Northern Italy—relationship to air quality monitoring, Atmos. Environ., 39(36), 6734–6746, doi:10.1016/j.atmosenv.2005.07.036, 2005.

Suni, T., Kulmala, M., Hirsikko, A., Bergman, T., Laakso, L., Aalto, P. P., Leuning, R., Cleugh, H., Zegelin, S. and Hughes, D.: Formation and characteristics of ions and charged aerosol particles in a native Australian Eucalypt forest, Atmospheric Chem. Phys. Discuss., 7(4), 10343–10369, 2007.

Please note that we also changed the sentence at line 325: As shown in Figure 6, similar seasonal variations were observed for the nucleation frequency in 2014 and part of 2015.

RC1-12, lines 311-312: "Because of a lack of knowledge about the potential gas precursor variation at Reunion Island, it is quite difficult to explain the event frequency variation with respect to the sources" highlighted, with the comment: **"This is very critical. In this manuscript, several statements are just speculations but not conclusions. There are several missing measurements, such as gas precursors, SO_2 etc., hence the insight provided here is very limited."**

AR1-12: We removed this sentence and we now reformulated this section to investigate only how physical parameters may influence the NPF frequency of occurrence and moved this part to section 4.4 where it belongs better. We also added the **Table 2** which gives more statistic details about the existing relationships between the main characteristics of the NPF events and the influencing factors. From line 391 to line 413:

"We also computed in Table 2 the existing relationships between the monthly average meteorological parameters and the ones of the main characteristics of the NPF events. We observe that radiation is highest between September and November (272.19 $W.m^{-2}$ on average), coinciding with one period of high NPF frequency (Fig. 5), but not with the maximum frequency of occurrence (March to May), nor with any high values of the GR or J_2/J_{12} (Figs. 7 and 8). As a consequence, no correlation is observed

between radiation and the NPF variables. Hence, the availability of light for photochemistry is not the only parameter influencing the NPF frequency, nor the formation rates or growth. The temperature averages are higher from November to April (14.02 °C). We find a significant (at the 95% confidence level) anti-correlation between temperature and the nucleation rate and GR. As mentioned earlier, this parameter would partly influence the VOC emissions (Yu et al., 2017) since it is one of the conditions for vegetation development and the decomposition of organic matter. Thus, the anti-correlation would indicate little influence of biogenic precursors from the vegetation on the intensity of the NPF and on the growth of the nucleated particles. However, the seasonal temperature variations are similar to the seasonal variation of the NPF event frequency even if the correlation is not significant. The relative humidity values are typical of an inter-tropical island with peaks in summer, between December and March (76.79% on average), and the lowest values obtained in July and September. Cloudy conditions were previously shown to inhibit formation of new particles, by scavenging newly formed clusters (Venzac et al., 2007). They might also decrease photochemical processes at the origin of the formation of condensable species contributing to the growth of clusters to stable particles. At Chacaltaya, Bolivia (5200 m a.s.l), Rose et al. (2015b) reported high frequencies during the southern winter, which coincide with the dry season. For the Maïdo station, frequency variations are not fully synchronized with the dry or wet periods as defined in Fig 9b. However, there is some uncertainty both in the dry/wet season segregation and with the exact identification of maxima/minima in the seasonal variation of the NPF frequency. When considering relative humidity, we do not find any link between RH and the nucleation frequency (Table 2) but a significant anti-correlation with the formation rate: low RH values correspond to the July-August-September nucleation peak. Figure 9b shows that the appearance time of the ultrafine particles seasonal variation is well correlated to the sunrise."

RC1-13, lines 341-343: "on average, the GRs were enhanced during the wet period, which is not in agreement with the present study, as we find high medians during the dry period (22.82 nm.h-1 averaged from July to November)" highlighted with the comment **"Any explanation or comment for this?"**

AR1-13: Several reasons may explain these discrepancies, such as the topography of each station relative to cloud formation during the wet season, or the seasonal variation of condensable species responsible for the particle growth. However, we do not have enough information on the respective factors that may induce such a discrepancy and we could only introduce more speculation that we want to avoid.

RC1-14, lines 360-362: "The peak in July is correlated to the Somalian phytoplankton bloom, which indicates a possible influence of a marine source on the NPF intensity during this month" highlighted

AR1-14: Yes, we agree that this is speculation, and do not mention this coincidence anymore in the discussion and the conclusion.

RC1-15: "arbitrarily" is highlighted, with the comment: "Why arbitrarily? Will selecting different values result in different conclusion? Need justification of this threshold value".

AR1-15: The value chosen is not totally arbitrary. We started our investigation by choosing the median CS over the whole year as a threshold value. We then increase and decreased this threshold so we have a reasonable seasonal variation of days exceeding this threshold values. Choosing other threshold values would have led to less pronounced seasonal variability, but would not have changed the shape of the seasonal variability. We added new sentences at lines 442 - 447:

"We started our investigation by choosing the median CS over the whole year as a threshold value. We then used an iteration process to fix the threshold so we have a reasonable seasonal variation of days exceeding this threshold values. Choosing other threshold values within CS_{2ev} and CS_{2noev} medians range would have led to less pronounced variability but would not have changed the shape of the seasonal trend".

RC1-16, line 441: "modelling methods" highlighted, with the comment: "What kind of modelling methods mentioned here? "

AR-16: It would be helpful to perform back-trajectories analysis (of the flexpart type) with a fine resolution to take into account the local and complex atmospheric dynamic at the Maïdo observatory. We could also use mesoscale atmospheric models such as Meso-NH, but this would need to include all potential sources of precursor gases and a good nucleation parameterisation. We now mention the models that would be useful in the text at lines 480 – 484:

"To complete this work, it would be valuable to have direct measures of the cluster ion composition that would provide indication of the anthropogenic, vegetation or marine sources contributions to nucleation at the site. In addition and although they are complex, modelling methods such as a detailed back-trajectory analysis should be used to understand the origin of the local air masses and source contributions at the Maïdo observatory."