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.

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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note that the structure of the article has been modified (especially for 4.1 and 4.6 sections), several figures had been replaced by others and we added new ones.

<u>The new plan is as follow:</u>

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

---------------- <mark>4.1 Dynamics of the NPF events at Maïdo observatory</mark>

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

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

5 Conclusions<mark></mark>

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 #2 comments (RC2):

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We thank also reviewer 2 for the constructive remarks that he did and the improvement propositions that he suggests and we first answer to the major comments.

RC2-1: Does the elimination of 47 days that occurred during the three eruptive periods, along 61 days of missing data, have an impact on your results pertaining to seasonality of NPF. From Figure

5, it appears that a significant number of days in 2015 were not considered during the winter (Feb and March), and there was not overlap with 2014 data for confidence. My concern arises due to the statistics of small numbers considered. Often the months with lower NPF frequency (near or below the annual average) are represented by the months with a more complete dataset (30-31 days). Would you provide more information or statistics showing that you are representing each season with a similar amount of data?

AR2-1: On the figure 5 you can find the numbers of days which were taken into account for the occurrence calculation. For 2015 summer (January to March), 58 on 90 days had been taken into account which represent 64%. For 2015 winter, (from May to November) 145 on 214 days had been taken which represent 68%. As a consequence, the data percentage is quite-similar for both seasons. We are aware that for February and March, the trend could not be confirmed by the 2014 data and the uncertainty is higher. However, we don't think that it could change de bimodal variation.

RC2-2: This is especially relevant since you are contrasting your results with those reported by Rose et al. (2015), and do not see NPF governed by dry/wet seasons. The paper continues with potential explanation for these differences.

AR2-2: Thanks for this remark. We actually moderated our argumentation on the contrast with the results from Rose et al. taking into account the uncertainty on the frequency seasonal variability. We now argue in section 4.4:

"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"

RC2-3: This becomes more evident in Section 4.4. The meteorological variables are discussed in reference to the monthly NPF frequency, monthly averaged GR and monthly averaged formation rates. I would encourage the authors to instead consider robust statistical methods to look for significance (or lack thereof) between these meteorological variables (along with CO) and NPF frequency, GR or formation rates. For example, do you see correlation on the daily level between

NPF frequency and threshold values of gas concentration or state variables? This section is inconclusive.

AR2-3: To respond to your request, we added a new Table 2 in which you can find the R correlation coefficients giving statistically significant results about the relationships between NPF parameters (occurrence, GR and Js) and influencing factors (Ray, RH, T, P, CS_2, CS_{2prop}, CO and BC). Correlations have been calculated with the twelve monthly averages for each parameters. Taking a degrees-of-freedom value of 10 and a risk $\alpha = 0.05$ (95% of confidence), we obtained a lower limit value of 0.576. The highlighted R in the table are above the limit value meaning that the variables are dependent. We especially use this table in the section 4.4 to bring values on the relationships between meteorological parameter and NPF characteristics:

"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."

We also use it in section 4.5 (condensation sink) from line 449 to 451:

"Hence, we actually find a similar seasonal variation between the frequency of CS exceeding a threshold value and the frequency of occurrence of NPF events (also shown by Table 2). This strengthens the hypothesis that there are precursors potentially transported simultaneous to aerosols from lower altitudes."

And in the new section 4.6 (black carbon, the anthropogenic distribution):

"Potential precursors may be of anthropogenic origin and we investigated their potential contribution by using BC as a tracer. Figure 11 shows the monthly mean concentrations of BC over the period January-October 215. The best similarity between the seasonal variations of BC ad the ones of NPF parameters is found for the nucleation rate (Table 2). This may indicate that air masses influenced by a larger contribution of anthropogenic compounds are favourable to more intense NPF events, but not

necessarily more frequent. Indeed, moderately high NPF frequencies are observed for high BC monthly mean values during the spring period, but the high frequencies of NPF event occurrence observed during the autumn period are nor associated to very large BC monthly mean concentrations, and thus other contributors are expected during autumn. The growth rate of newly formed particles is best correlated to CO concentrations (Table 2). Condensable species necessary to form new particles are hence likely different from the ones responsible for their further growth."

RC2-4: Section 4.5 is very interesting, and as the authors note in contrast to previous studies, as low CS is normally associated with NPF or at least you do not expect an anticorrelation. This brings us to the conclusion that you have a precursor potentially emitted simultaneous to aerosols that are increasing CS. This is consistent with your Figure 4 (and previous reviewer's comments pertaining to advection).

AR2-4: Yes, we take into account this comment and reviewer's 1 comment and now include new figure 4b with BC concentration mean diurnal variation for July 6th and new figure 5 with BC concentration mean diurnal variation, N > 100 nm (accumulation mode) and N < 30 nm (nucleation mode) particle concentrations for different seasons in section 4.1 with the corresponding text:

"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"

RC2-5: Why was Figure 4 selected as the example plot? Does the appearance of accumulation mode particles simultaneous to the formation of new particles always happen? This was mentioned in reference to this case, but it is not clear if this happens frequently (during every season) at the site. If so, is this source of accumulation mode particles associated with a specific wind direction? This is stated in the conclusion, but this reader would appreciate more information pertaining to the details of this source

AR2-5: As mentioned in the previous comment, we now provide a diurnal variation of BC for each season, but also show the diurnal variation of the nucleation mode (N_{30}) and accumulation (N_{100}) number concentration of particles for the particular case of the case study, but also for the seasonal averages.

We second answers to the minor comments.

RC2-6 line 21: term "off-season" is unclear to this reader.

AR2-6: We agree that this term is misused and changed it.

RC2-7: line 306: The statement "similar seasonal trends were observed for nucleation frequency in 2014 and 2015", is only observed from May – Dec".

AR2-7: We modified this part, which is now: "As shown in Figure 6, similar seasonal variations were observed for the nucleation frequency in 2014 and part of 2015."

RC2-8: line 331: typing error on "FNP"
AR2-8: It is corrected

RC2-9: Then, you ask a question about the GR_{12-19} which is "Why did you choose to calculate the growth rate over such a small size range (12-19) and few bins? "

AR2-9: The lower limit is defined by the smallest channel that we had on DMPS and the AIS data set was too reduced. The upper limit is close to those which are traditionally used in the literature.

We added a new sentence at line 261: "As the DMPS offers a much more extended data set than the AIS, we applied the method to the DMPS 12-19 nm size range for which the lower limit (12) was defined by the smallest channel of the device."