We thank Referee #2 for his comments and suggestions, which we hope will help improving the manuscript. We have addressed the comments point by point below, with separate answers to the different topics/aspects of each of the main comments (1-3). In addition, the errorbars shown on Fig. 9.b were modified, as those did not correspond to the actual variability of N_{50} absolute increase in the original version of the manuscript (former Fig. 8.b).

Comment 1: Section 2.2. The authors should discuss briefly the uncertainties and limitations of the equations 1 to 4 in calculating the particle formation (J) and growth (GR) rates in their data. **(A)** First, these equations have been developed originally for regional NPF, in which formation and growth of particles is assumed to take place relatively homogeneously over large spatial scales. This is apparently not the case in plumes where, among other things, various transport effects on J and GR should be taken into account. **(B)** Second, experimental limitations cause further uncertainties in determining J and GR. For example, using coagulation sink at 12 nm for all particles in the size range 12-19 nm in equation 1 causes some overestimation of coagulation losses, which results in underestimating J12. Also, Calculating J2 from J12 would require knowing GR in the size range 2-12 nm rather than that in the size range 12-19nm. While it is impossible to take into account the above issues to correct the data, the authors should at the very least discuss these issues briefly in section 2.2. If possible, the authors could also estimate whether resulting uncertainties are important or not with respect to their results.

Reply 1:

(A) This is an interesting discussion, as it is true that the abovementioned equations were originally derived to describe regional NPF. However, we do find a stronger increase of $N_{12\text{-}19}$ over time within the volcanic plume compared to non-plume conditions. The resulting increased particle formation rates observed on plume days indicate that particles are formed within the volcanic eruption plume "homogeneously" along the transport pathway to Maïdo. The same reasoning can be done with the growth rate. If nucleated particle appear to gradually "grow" with the typical banana shape, it means that the regional-type nucleation and growth process is taking place along the transport path. Based on these observations, we believe that Eq. (1-4) can be used to describe such events occurring at a sub-regional scale. This aspect is now better addressed in Section 3.1.3: *"Higher particle formation rates observed on plume days indicate that particles were constantly formed in the volcanic plume along the transport pathway to Maïdo, showing that nucleation and growth taking place over a distance of the order of 40 km appears like a regional scale homogeneous process, which can be described with the usual equations (Eq. 1-4) recalled in Section 2.2"*.

(B) In fact, the use of $CoagS_{12}$ for all particles in the investigated size range leads to some uncertainty in the calculation of J_{12}. However, based on Eq. (1), which is recalled below, we believe that $CoagS_{12}$ contributes to an overestimation (and not underestimation) of the particle formation rates.

$$J_{12} = \frac{dN_{12-19}}{\mathrm{dt}} + CoagS_{12} \times N_{12-19} + \frac{GR_{12-19}}{7\,nm} \times N_{12-19} \quad (1)$$

This is now clearly mentioned in Section 2.2: *« Note that the use of $CoagS_{12}$ for all particles in the range between 12 and 19 nm might cause some overestimation of the actual coagulation losses, and in turn lead to high estimates of J_{12}."* Also, we have included a sensitivity study at the end of Section 2.2 to give further insights into the effect of the particle growth rate in the determination of J_2 based on Eq. (2-4). All in all, this analysis reveals only limited effect of the growth rate variability/accuracy over the 2-19 nm in the conditions of our study.

Comment 2: Section 3.2.2. In this work, neither J2 nor H2SO4 concentration were measured directly, but were derived from other measured quantities, resulting in potentially large uncertainties in their values. This has implications which are not mentioned in the paper. **(A)** First, how reliable is the

observed relation between J2 and H2SO4 concentration, and how meaningful is it to compare this relation with those observed in studies were J and H2SO4 concentration were measured directly? **(B)** Second, how meaningful is it compare J obtained here with parameterized J due to binary water-sulfuric acid nucleation as a function of H2SO4 concentration? Does this comparison tell anything about nucleation mechanism?

Reply 2:

(A) We agree with the fact that the use of values derived from indirect calculations may lead to uncertainties, and such limitations should be kept in mind when interpreting the results. Regarding the determination of [H_2SO_4], we were unfortunately not able to evaluate the relevance of the proxy by Mikkonen et al. (2011) in volcanic plume conditions. Hence, in addition to the comment reported in Section 3.2.2 (*"which may stem from a reduced predictive ability of the proxy by Mikkonen et al. (2011) for the highest SO_2 mixing ratios"*), we have included another "warning" earlier in the manuscript, in Section 2.4: *"However, since the relevance of this proxy could not be evaluated in volcanic eruption plume conditions, neither from available measurements nor existing literature, one should keep in mind the potential limits of using such parametrization when interpreting the related results".* Also, this demonstrates the need for additional measurements, as now clearly indicated at the end of Section 3.2.2: *"Such measurements would also allow more detailed evaluation of the proxy by Mikkonen et al. (2011) for the prediction of [H_2SO_4] in volcanic eruption plume conditions".*
Concerning J_2, it goes without saying that the use of Eq. (2-4) naturally leads to some uncertainties in the calculation of J_2. However, these equations have already been approved and used in other studies, such as in the companion paper by Foucart et al. (2018). Moreover, the impact of GR_{12-19} on the results derived from Eq. (2-4), which was rightly questionable, is now explicitly discussed in the revised version of the manuscript.
The results obtained in the present work were compared with that of other studies where measured values were used since those were the only "reference" we had for such comparison, in specific when restricting the literature to the poorly documented volcanic plume conditions. The comparison with Sahyoun et al. (2019) finally gave the chance to highlight the similarity of the values derived from the different approaches (measured *vs* calculated J and [H_2SO_4]) in comparable conditions, thus giving more credit to the values reported in the present analysis.

(B) Comparison with the theory has multiple interests, as mentioned in the paper in Section 3.2.2:
- Investigate whether or not the binary water-sulfuric acid nucleation can explain the particle formation rates reported in plume conditions;
- Show evidence for the ability of the parameterization to provide some reasonable estimates of the particle formation rates based on the knowledge of SO_2 mixing ratios only.

Based on the use of the parameterization, we have also included some additional discussions in the revised version of the manuscript regarding the contribution of the different nucleation mechanisms, charged and neutral (Section 3.2.2): *"As evidenced on Fig. 5.a, the total formation rate of 2 nm-clusters was mostly explained by ion induced nucleation for [H2SO4] below ~ 8×10^8 cm-3, while neutral pathways seemed to explain the observations at larger sulfuric acid concentrations.".*
Finally, the fair agreement between the formation rates derived from measurements and that predicted by the theory gave additional support to our approach based on indirect measurements.

Comment 3: There are a few issues related to the particle growth that need some clarifications. **(A)** First, did the authors consider particle growth from one mode to another when estimating the relative contributions of primary and secondary particles in each mode? This remains a bit unclear when reading the results. **(B)** Second, the authors do not tell what were the typical air mass transport times from the volcano to the measurement site. This is important because for the reported particle growth rates (Fig. 2a), it takes a while before particles formed in the plume are able to growth into the Aitken mode, and for several hours before they can reach the minimum CCN size (assumed >50 nm here) or the accumulation mode. Is it feasible that particle formed by NPF in the volcanic plume reach these sizes

by the time measurements were conducted? **(C)** Third, while I agree with the authors that volcanic emissions are able to boost particle growth by e.g. heterogenous reactions of SO2 on particle surfaces, there seems to be some inconsistences in the storyline: on one hand the authors state that the plume appear not to influence the particle growth (section3.1.3), and on the other hand they state that particle growth in the plume increased both modal (section 3.3.1) and CCN (section 3.3.2) concentrations.

(A) The evaluation of primary vs secondary processes contributing to volcanic emissions were performed via the comparison of the 7:00 LT size distribution in and out of plume to evaluate primary emissions, and via the comparison of the maximum particle concentrations measured for each mode in plume and out of plume (observed between 11:00 and 14:00 LT depending on the modes and conditions) for the secondary particle contribution. The presence of a secondary contribution to the accumulation modes is likely the result of the growth of particles from the Aitken mode, due to the presence of more condensable gases. This is now more clearly stated in the text, and also recalled in the caption of Fig. 8.

(B) We have included a discussion regarding the growth of the particles nucleated close to the vent during their transport to Maido in Section 3.1.3, showing evidence for their ability to reach ~50 nm, i.e. CCN relevant sizes, over the distance of 39 km between the volcano and the station: *"A rough estimate for the transport time of the particles nucleated in the vicinity of the volcano to the Maïdo observatory can be obtained by dividing the distance between the sites by the median wind speed measured on NPF event days: 39 km ÷ 1.8 m s-1 ≈ 6 hours. This indicates that in such conditions, the GR_(12-19) reported on Fig. 3.a were often sufficient (> 8 nm h-1) for the newly formed particles (~ 1 nm) to grow up to CCN relevant sizes (~ 50 nm, see Sect. 3.3.2) during their transport, further explaining the observation of the typical banana shape of the events, both on plume and non-plume days. Similar analysis was repeated with the 75th percentile of the wind speed measured on NPF event days (2.9 m s-1), and, again, the observed growth was often fast enough (> 13 nm h-1) for the particles to reach 50 nm during the corresponding ~ 3 hours 45 minutes trip to Maïdo".*

However, when focussing more specifically on the CCN population in Section 3.3.2, we did not try to isolate the contribution of NPF to the observed CCN increase, as this would have been complex (impossible?), but we reported instead the contribution of secondary particles. Indeed, we did select the days when particles originating from NPF reached CCN-size, and in turn presumably contribute to CCN population, but the concentration increase that we calculated reflected the contribution of secondary processes as a whole, i.e. NPF but also growth of pre-existing particles, as mentioned in Section 3.3.2: *"On event days, the diurnal variation of N_{50} was strengthened due to the concurrent formation of secondary aerosols, i.e. including the formation and growth of new particles as well as the growth of pre-existing larger particles mentioned earlier (Fig. 9.d)"*, *"Following these hypotheses, the contribution of secondary aerosols to the observed CCN population was estimated from the difference between the median of the N_{50} absolute increase observed on event days (i.e. resulting from transport of particles from the boundary layer and secondary aerosol formation) and that of non-event days (resulting from transport only)"*.

(C) It is true that we report higher particle and CCN concentrations in plume conditions compared to non-plume days. However we do not relate those to increased particle growth rates, but rather to increased particle formation rates and to the presence of additional primary particles from volcanic origin during eruptive periods. This is for instance illustrated in the paragraph below, taken from Section 3.3.1: *"This observation was consistent with the enhanced production of particles previously reported for lower sizes in plume conditions, as the increase of the particles concentration in the Aitken mode most likely resulted from the growth of smaller particles originating from the nucleation mode. As already mentioned, the concentrations of the 2 accumulation modes measured at 07:00 LT were both significantly higher on plume days (235 4800 and 100 1300 cm^{-3}, for the first and second accumulation mode, respectively) compared*

to non-plume days (80 and 18 cm^{-3}, respectively), most likely due to additional sources of particles at the vent of the volcano during eruptive periods (see Section 3.2.1)".
Nonetheless it is true that the sentence at the end of Section 3.3.2 was confusing, and was thus slightly modified for clarity: "*the growth of ~~the newly formed~~ more particles to CCN relevant sizes was favoured during NPF events occurring in the presence of large amount of H2SO4 caused by the eruptions*".

Minor/technical issues

Comment 1: Page 7, line 4: "...when global radiation >50 ...". Something is missing from here (was?).

Reply 1: "was" added.

Comment 2: The format of providing the time difference (i.e. 2h10) in section 3.1.2 seems strange to me. Is this a correct way of expressing the time difference?

Reply 2: We are actually not sure about the expected format for expressing a time difference; we anyway changed it to "XX hours and YY minutes" instead of "XXhYY".

Comment 3: Page 10, line 2: "GR12-19 showed an important variability, ...". What do the authors mean by "important" here?

Reply 3: The "important" variability was related to the inter-quartile range; this aspect is now better addressed: "*$GR_{12\text{-}19}$ showed an important variability, as reflected by the monthly inter-quartile ranges, which were on average of the order of 80% of the corresponding medians*".

Comment 4: Excluding the last paragraph of section 4, the text in that section mainly summarizes the results discussed earlier in the paper. As a results, an appropriate title of this section would be "4. Summary and Conclusions".

Reply 4: Changed.

Comment 5: Would it be possible to change the lines and marks with yellow color in Figures into some other, more easily visible color?

Reply 5: The number of figures with yellow lines and/or markers is now limited in the revised version of the manuscript, since, after considering comments from Referee #1, strong-plume events are no longer highlighted in Section 3.3. For simplicity, and in order to optimize both the visibility and the contrast with other colours, we have decided to keep yellow in our colour code. However, we were able to improve Fig. 8 (yellow changed into purple), Fig. 7 (yellow changed into green) and Fig. 4.a (yellow numbers into black).