

We thank referee N°2 for his comments and remarks which contributed to improve and clarify the present paper. Our answers to the suggestions are listed below.

SPECIFIC COMMENTS

Comment 1: How do the authors separate CCN formation from freshly nucleated particles and CCN formation from the growth of pre-existing particles during nucleation events? Here, I refer to the very detailed comments of Anonymous Referee #1. There is not much to add.

Reply 1: We agree with the fact that the manuscript needed for more clarity regarding this aspect. We now use a different terminology for section 3.1.1, using the term « CCN production DURING NPF » instead of CCN production FROM NPF. Also, in order to make our approach clearer the methodologies and discussion on the uncertainties on these methodologies are included in the results sections. In addition, several sentences were rephrased throughout the manuscript in order to further avoid any misunderstanding.

Comment 2: How robust is the treatment of advection of different air masses by the selected approach? Are there other observables (e.g., trace gases) available which allow a more robust treatment of particle transport than the simple method deployed in the study? The authors use the hypothesis that similar particle number concentrations are transported to the site during days with and without particle formation events. They state in Section 3.1.2 that at hours outside of NPF events, particle number concentrations were on average similar for event and non-event days. However, what is the variability of the particle background and does it depend on the wind direction where the air masses came from, etc.? In particular the variability of the particle background needs to be presented more quantitatively since this parameter determines the level of uncertainty of the reported CCN increases by NPF. Concerning the structure, the presentation of results in Section 3.1 is confusing. The authors start with a detailed description of CCN production and list all obtained numbers in detail and show the min Fig. 2. Then in Section 3.1.2 they introduce a correction of the presented CCN number concentrations. It is confusing that the CCN production neglecting the influence of advection is shown in Fig. 2 while the more important CCN production from NPF only is not shown but only listed in Table 2. If I understood right, the authors focus on CCN from NPF. If this is true then the way of presenting the data in Section 3.1 should be revised. One possibility is to start with a quantitative analysis of the “particle background” during non-event days, including its variability, introduce then the method for determining CCN production and present finally the CCN production values corrected for particle transport.

Reply 2: We know that the correction that we applied for particle transport is not completely accurate, since it relies on a strong hypothesis which is explicitly mentioned in the text (“similar number concentrations of particles are transported to the site on event and non-event days”), and which might actually not be verified because of some reasons which are now mentioned with more details in the revised version of the manuscript (Section 3.1.2):

“These calculations rely on the hypothesis that the specific environmental conditions on which NPF occurs are not influencing the transport from lower atmospheric layers. In order to further evaluate the reliability of this assumption, wind direction and speed as well as global radiation were investigated on event and non-event days (Figures S3 and S4 in the supplementary material). As previously reported by Rose et al. (2015a), NPF events are favoured during clear sky conditions, when radiation is higher (Fig. S3). Thus, there is likely a bias towards an underestimation of radiative driven transport from lower atmospheric layers due to the fact that cloudy days are over-represented for non-event days. Regarding wind, contrasting directions are also observed between event and non-event days (Fig. S4), with patterns closely related to those observed for the dry and wet seasons, respectively (Rose et al., 2015a). It is worth noticing that winds originating from the more polluted sector of La Paz – El Alto

(south) do not seem to be over-represented neither on event nor on non-event days. However, because of the close proximity of this area, it is complex to further assess how it contributes to CCN concentration from wind direction alone, and we cannot exclude a bias related to the variability of this specific source between event and non-event days. Nonetheless, the particle number concentrations observed at the time preceding the usual occurrence of the NPF events are similar for event and non-event days (Fig 3, S1, S2). Moreover, higher wind speeds are on average recorded on non-event days, that likely lead to an enhanced transport of particles to the site compared to event days, and hence lead to an underestimation of the contribution of NPF to the increase of CCN. In any case, taking into account the contribution of transport when calculating the increase of CCN concentrations after NPF events was never done in the past, and certainly helps approaching a more realistic view of the real contribution of NPF to CCN number concentrations.”

Additional figures are also provided in the supplementary, showing wind speed and direction (Fig. S4) and radiation (Fig. S3)

However, the aim of this correction is not to provide an exact estimation of transport contribution, which, we believe, cannot be retrieved from these measurements, even if including additional parameters, such as trace gases concentrations. Our objective, less ambitious, is rather to go one step further compared with previous analysis published in the literature in order to estimate NPF contributions to CCN number concentrations which are closer to actual values, as mentioned in the text.

We agree that the terminology to describe sections 3.1.1 and 3.1.2. was confusing. We now use the term « CCN formation during NPF » for section 3.1.1 and CCN formation from NPF for section 3.1.2.

We agree that the most important results should be better emphasised for section 3.1.2. For that purpose, an additional Figure (4) is now provided and the text has been slightly changed:

“The contributions of NPF particles to the increase of CCN, all shown on Fig. 4.a. and reported in Table 2 for the different seasons and sizes, hence represent a significant fraction of the CCN increase shown on Fig. 2.a. and reported in Table 1. The contribution of NPF to CCN concentrations are comparable or even higher than those previously mentioned for other stations in the literature, which probably also include CCN sources other than NPF. The relative impact of NPF are estimated to increase the CCN₅₀ number concentrations by more than 250 % during both seasons, and the CCN₁₀₀ number concentrations by more than 100% and 200% during the dry season and wet season, respectively.”

***Comment 3:** How robust is the separation between air masses from the boundary layer and from the free troposphere, and what is the expected impact of air mass history on the occurrence of new particle formation events? This question refers to Section 3.2 which in its current form is difficult to understand. The attempt of the authors is quite understandably to study if NPF events occur preferably in air masses originating from the BL or from the FT. However, doing this requires a clear presentation of event types and characteristics before going into details. Here the authors should restructure Section 3.2, start with a clear presentation of event types and scenarios. One table including all considered cases (with more detail than stated in Table 3) etc. might help. Looking at Fig. 5, there is no big difference between the scenarios, except for S1, S6 and likely S7. The authors may rethink the choice of scenarios in order to get a more precise conclusion from this part of the study. In addition, the expected impact of air mass history should be investigated / discussed.*

Reply 3:

The method used to distinguish between boundary layer and free troposphere air masses performed in the present study is the only one we could apply given the measurements conducted in Chacaltaya.

The impact of air mass history, including occurrence and length of BL contact, has not been investigated due to lack of proxy measurement and modelling tools (such as those used by Tröstl et al. (2016)). However, the fact that air mass history may, in addition to local conditions, influence the occurrence of NPF is now mentioned at the end of Section 3.2.3 in the light of the work by Tröstl et al. (2016) and Bianchi et al. (2016):

“Additional analysis regarding the history of the air mass and BL influence along its trajectory would provide valuable information to even more assess the role of the exchanges between the BL and the FT on the occurrence of NPF and its contribution to the formation of new CCN. Indeed, observations conducted at the Jungfrauoch showed that stronger NPF events (type I) occurred in air masses one or two days after contact with the BL (Bianchi et al., 2016; Tröstl et al., 2016). These results are however based on proxies (CO, NO_y) and modelling tools which were unfortunately not available for Chacaltaya. Nevertheless, our results goes to some extent into the same direction as the work by Tröstl et al. (2016) and Bianchi et al. (2016), at least supporting the major role of BL intrusion (regardless of its kind, before or during the event) to sustain particle growth. Similar FT feeding process from the BL was also shown by Rose et al. (2015b) at the puy de Dôme (France, 1465 m a.s.l.).”

We agree with the fact that the number of scenarios used to assess the impact of BL on the occurrence of NPF was maybe too high. It was thus reduced in order to clarify the message:

” The most frequent scenarios, which include more than 88% of the documented events, are listed, together with their frequency of occurrence, in Table 3. Scenario S1 refers to those days when the first steps of the NPF process were observed to occur in the BL, while scenario S2 refer to the events started in the FT. Scenario S2 is further divided into two sub-classes to distinguish between the events which first steps occur exclusively in the FT (S2.1) from those during which BL dynamics lead to changing conditions in the course of the event (S2.2). Events triggered in the IL are not frequently observed compared to those initiated in the BL or in the FT, and are thus not highlighted in this classification.” (Section 3.2.1)

The description of the scenarios in Table 3 was also detailed.

MINOR COMMENTS

Comment 1: Since the classification of NPF events is crucial for understanding the manuscript, a brief description of types should be given at the end of section 2.2, instead of referring to the references Hirsikko et al. (2007) and Rose et al. (2015).

Reply 1: As suggested, brief description of the event types is now provided in Section 3.1.1:

“First, only those NPF events referred as type I, i.e. with clear particle growth from smallest sizes, were considered; they contrast with type II events, during which the growth is more irregular and may be interrupted in certain size ranges, and bump type events, which completely miss the growth of the newly formed clusters (Hirsikko et al., 2007; Yli-Juuti et al., 2009).”

Comment 2: Please add brief descriptions of quantities J and GR to x axis of Figure 6.

Reply 2: Figure caption was slightly modified:

“Median formation rate of 2 nm particles (J_2) and growth rate in the range 1-3 nm (GR_{1-3}) reported separately for type I events initiated in the BL (scenario S1) and in the FT (scenario S2). Lower and upper limits of the error bars stand for the 1st and 3rd quartile, respectively.”

Additional information is also provided in the text, before the description of Fig. 7:

“In order to further characterize the NPF events in the different atmospheric layers, statistics regarding the formation rate of 2 nm particle and the growth rate (GR) in the size range 1-3 nm as a function of the scenarios were performed for type I events. Growth rates were derived from the particle number size distribution using the “maximum” method from Hirsikko et al. (2005), while formation rates were calculated according to Kulmala et al. (2007).”