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Discussions

Interactive comment on "New Particle Formation and impact on CCN concentrations in the boundary layer and free troposphere at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia" *by* C. Rose et al.

Anonymous Referee #1

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Title: New Particle Formation and impact on CCN concentrations in the boundary layer and free troposphere at the high altitude station of Chacaltaya (5240 m a.s.l.), Bolivia

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The manuscript describes new particle formation (NPF) events observed at the Chacaltaya station in Bolivia and their contribution to the cloud condensation nuclei (CCN) population. The results are based on measurement done with the SMPS and NAIS where the CCN are particles with different sizes (50, 80 and 100 nm). The manuscript

also highlights the importance of studying this mechanism in order to better understand the influence of NPF in the CCN population. I consider that the data set is quite valuable (long term and remote location), however, the authors don't take full advantage of such data set. I therefore recommend the publication of this manuscript only after major revision.

I actually have three major concern listed here:

1- The CCN increase during NPF is confused with CCN increase due to NPF. The authors tried to differentiate these two points correcting for the transported CCN, but this is not enough, especcially because they don't correct for the growth of pre-existing particles. Since this is the major point of the paper I believe should be more solid. 2- The discrimination between FT and PBL is weak. The authors don't consider the history of these air masses which previous research has shown to be of paramount importance. 3- The section 3.2 is vague and beside some small details I don't see the take home message of this section.

Introduction: The introduction is well written; however, I think that some major studies have been forgotten or omitted. Recently, studies conducted at the Jungfraujoch appeared in Science and JGR-A. These studies should be mentioned as a comparison, especially in terms of CCN production, would be extremely valuable (Herrmann et al. 2015, Bianchi et al. 2016, Tröstl et al. 2016)

Page 2 Line 29: The reference Yli-Juuti is only about one site (Hyytiälä). I would recommend to consider the Manninen et al. (2010) EUCAARI paper which reports findings from 12 European sites.

Page 2 Line 30: Reference is needed.

Page 3 Line 17: "....However, observations to validate these predictions are scarce, especially for the FT...."

It is true that little information is available in this specific field of research. However, a new study by Tröstl et al., (2016) has just been published in the Journal of Geophysical Research (Atmospheres) which (among other things) investigates the contribution of new particle formation to the CCN concentration in the Alps in some detail. I think a comparison to this work (Alps vs Andes) would be quite interesting, especially considering the general scarcity of similar research.

Page 4 Line 1,2: The authors underline the vicinity of the site to a city like La Paz that has a large population and is assumingly rather polluted. This fact seems weirdly underused in the study. Why not determine La Paz air masses to find out what (if any) effect polluted air masses have on NPF and CCN production? A backtrajectory analysis might actually be quite interesting.

Section 2.2 Indirect method for the estimation of the NPF contribution to the CCN production I'm not sure that this method fulfilled what the authors claimed. I agree with the fact that the CCN increased during NPF is not just a pure coincidence since the NPF precursors certainly also facilitate growth. However, it's not possible to distinguish the CCN formed by the NPF events with the growth of pre-existing particles during the same time. An easy way to fix that somehow would be to assume that ALL pre-existing particles to become CCN before any new particles. I.e. the number of particles below 100 nm before the event must be subtracted from the CCN100 you are now using and so far for the other sizes. This still would not account for, say, 90 nm particles that are transported to the site during NPF and grow above the threshold and contribute to CCNmax but it would be better than the current approach.

Page 4 Line 28: I found CCNhigh and CCNlow quite confusing. I would rather prefer to use the size of the particles, therefore, I would call them CCN50 and CCN100.

Page 5 Line 1: "...The CCN production during an event was obtained from the comparison of the CCN concentration Ninit prior to and the maximum CCN concentration Nmax during the event..."

I agree with the authors that this is the CCN production during an event. However,

the authors treat this as the CCN increase due to NPF. As already mentioned, CCN increase DURING and BECAUSE OF NPF are not the same thing. This distinction demands for clarity of concept and language whenever the topic is discussed. The manuscript in its current form, however, conflates those things. Besides the need for exact language, I actually think the issue can be addressed (to an extent) as lined out above.

Page 5 Line4 "....tinit, when nucleated particles reach the threshold size...."

I don't think is possible to know that nucleated particles reached CCN size instead of larger particles that simply have grown above the threshold. The respective figure 1 actually shows that t_init isn't found as the text claims: if the text was true, then t_init_100 should be well after t_init_50 because growth takes time. In the figure, however, all t_init are the same. That means t_init is really just the time when CCN numbers start to increase. But that increase doesn't likely come from NPF. Figure 1 illustrates a further problem with this claim: t_init_100 is roughly 1.5...2 h after nucleation onset. If those were really newly nucleated particles, we would need growth rates of 50 nm/h. I find that hard to believe as such numbers have never (to my knowledge) been reported in the literature for atmospheric nucleation.

Page 5 Line 14: The authors acknowledge only partially the previous point. They say that the particles can be transported during that period but they still don't mention that small particles transported there can then grow to the threshold and being considered as formed by NPF. They also correct the transported particles by comparing NPF days with non-NPF days. This assumption is valid only if the physic dynamic is the same. If the NPF is triggered by the wind convection might be that during nucleation (more wind) the particles transported up there are more. This point needs further investigation or at least being commented.

Section 2.3 Method to assess the influence of the boundary layer in Chacaltaya.

To my understanding, this method only takes into account the local PBL influence at the time of nucleation. It does little to actually describe the air mass in which nucleation occurs. Bianchi et al. (2016) have shown that strong PBL contact 1-2 days before NPF is crucial in the case of the Alps (Jungfraujoch). While conditions are certainly different in the Andes, there is no reason to believe that local wind conditions could accurately describe an air mass and its history which is what one must do to get a handle on PBL influence. There is a good body of literature dealing with the assessment of PBL influence. Much of it has been summarized in recent papers by Bukowiecki et al. (2016) and Herrmann et al. (2015).

Page 8 Line 14: "...when particles reached the lowest activation diameter, i.e. 50 nm, they systematically grew up to at least 100 nm..."

This statement is stronger than what the data seem to support. We don't know for certain that those are not pre-existing particles that simply did a bit of growing above the considered threshold, or do we?

Page 8 Line 16: "...aerosol particles originating from NPF event and reaching CCN sizes..."

Yet again the same problem that I think it should be fixed. I haven't seen any evidence that all those new CCN come directly from NPF, and, indeed, I find it highly unlikely: as long as there are pre-existing particles their chances to add to the CCN concentration are MUCH higher than the chances of newly formed particles.

Page 9 from Line 15 to Line 32: The paragraph lacking a message.

First the authors give us comparisons to sites that are hardly comparable to a 5000 m peak, and then they tell us in the last few lines that those comparisons are more or less pointless and I actually i would agree because these sites are just different and the comparison does not provide useful information. A comparison to Tröstl et al., (2016) might be more interesting, especially since those results are quite different.

Page 10 Line 8:

Sunrise is typically a well-defined point in time and not a process that has an onset.

Section 3.1.2 Correction for the contribution of particles transported to the site

I'm a bit concerned regarding this method to correct the contribution of particles transported. As mentioned earlier, this method is valid only in case every day we have the same physics and nucleation only depends on the vapors present. However, if nucleation is triggered by the wind coming up the valley than during nucleation we would have more transportation of big particles and therefore the correction method is not ideal. Would be nice to know what are the differences (Wind direction, wind speed etc..) during nucleation and during no nucleation where these background values is taken in account.

Section 3.2 How layering influences growth to CCN-sizes I do understand the need of knowing where the nucleation events take place and especially if this lead to a big production of particles in the free troposphere. However, I believe that dividing in 10 scenarios is a bit over exaggerated and probably not quite realistic. I think it would be better if the authors can simplify this section. I don't think that selecting more than 3 scenarios is feasible. In addition to that the split into different scenarios seems ill-conceived since most scenarios are quite irrelevant with very little occurrences. This might all be a nice exercise in data analysis but the text fails to tell us what the actual results are. What do we learn in this section apart from some minuscule details? This section has the feel of filler material and needs to be improved with a fair amount of actual substance.

Page 12 Line 4: "...regarding the location of the station in the tropospheric layers..."

I wonder if this is actually relevant at all? The NPF events are mainly driven by the air mass history and not so much by the atmospheric layer when the event begins.

Page 12 Line 8: "389 NPF events"

Is that a different data set?

Minor edits:

Figures: In general no need to state Chacaltaya at the end of every captions.

Figure 1: Why Tinit is not before the nucleation but already a after the start of the event? Please also describe the figure, Particle size distribution measured by. . ... and so on.

Refererences:

Bianchi, F., et al., (2016), New particle formation in the free troposphere: A question of chemistry and timing. Science, 10.1126/science.aad5456

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Herrman, E., et al., (2015), 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., 10.1002/2015JD023660

Manninen, H.E., et al., (2010), EUCAARI ion spectrometer measurements at 12 European sites – analysis of new particle formation events. Atmos. Chem. Phys. 10.5194/acp-10-7907-2010

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