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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Investigation of nucleation events vertical extent: a long term study at two different altitude sites" *by* J. Boulon et al.

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The authors are grateful the referee #3 for his relevant comments and remarks which significantly contributed to improve the quality of the paper. In this revised version, we have tried to respond to most comment modifying the document accordingly. Specific anwers to each comments that we received are listed below.

Referee's comment (RC): The terms new particle formation and nucleation are both used here. What is the definition of new particle formation here? Nucleation is only part of the NPF process and should not be used as a synonym. The authors should

very clearly define these terms and use them carefully.
Author's reply (AR): The different terms used throughout the manuscript do not refer to the same processes. Aerosol nucleation refers to the first steps of new particle formation, which can be observed only using an instrumentation detecting nanometer-size particles. When the instrumentation detection limit is higher than the nanometer scale, the term "nucleation" can not be used anymore and it has to be replaced by "new particle formation". This is the reason why in the "Introduction" the term "new particle formation" was used when describing previous studies which were not conducted with intruments with a sufficiently low size range detection. Other occurences in the manuscript are used to avoid the systematic term "nucleation and subsequently growth" since when the nucleation occurs, it is followed by a new particle formation.

RC: There is definitely a difference between measurements on a mountain and airborne measurements in a similar altitude. All vertical mixing processes are of course influenced by the topography, i.e. the mountain. This should be mentioned in the introduction and discussed later. Such measurements on a mountain are the only way to realize long term measurements in high altitudes but cannot fully replace airborne measurements.
AR: This is right, high altitude mountain based measurements cannot replace airborne studies and do not pretend to replace them. Both are complementary and usefull to understand different processes. Mountaineous area represent mixing zone between different atmospheric layers due to the topography. They lay at the interface between the low and the high tropospheric media. Although there might be some bias at mountain sites due to the distorsion of the air flows because of the topography, they provide statistical information that can not be obtained from airborne studies. The bias due to the topography can however be indirectly evaluated by analyzing the LIDAR vertical profile in regard to the in situ measurements. We observe that nucleation is always promoted at high altitude when the LIDAR indicates that the PBL height is

at the puy de Dôme level or lower. This can not be a coincidence (over 157 cases) and this indicates that the phenomenon is occuring above the PBL or at the interface between the PBL and the free troposphere. However, the influence of the topography on possible air mass distortion is now mentioned and discussed in the paper.

RC: The climatic conditions should be described in more detail. Only winter temperatures are mentioned, whats about summer?
AR: Summer temperatures are in the range $5 - 25$C. This is now mentioned in the text.

RC: Figure 1 should be better explained, in the color plot the unit of height is missing, the labels on the axes are mostly too small.
AR: Figure 1 labels were enlarged to help the reader. Furthermore, we add a better description of this figure in the text and in the figure caption.

RC: The PdD station is explained in detail but I did not find anything about the measurement technique at Opme. Please explain the instrumentation at both sites.
AR: The Opme site is poorly equiped compared to the PdD station. In addition to basic meteorological parameters monitoring such as temperature, relative humidity and pressure which are continuously measured, the site was equiped with an AIS or NAIS depending the period of the year for ultrafine aerosol size distribution measurements. This description was added in the text.

RC: Water-vapor-to-dry-air mixing ratio can be named much shorter: water vapor mixing ratio means the same and is more commonly used in atmospheric science.
AR: Corrected.

RC: The acronym FT has not been explained so far. Of course, most readers know it but it should be introduced again (or I did not see it..)

AR: This is right. Corrected.

RC: I do not really understand the procedure to obtain the PBL height. The figure does not help much without any explanation. A more detailed figure caption should be given. What is an injection layer? I never heard this word before. If you really want to use this term, please define what it means. It is not commonly known.
AR: The PBL height procedure presented in the manuscript is based on the assumption that the PBL height is characterized by a break in the particle concentration (higher in the PBL, lower in the FT). Thus we can infer that the light diffusion signal will be discontinuous between those two layers. As a consequence, the PBL height can be estimated as the height where the light diffusion regims starts to change, as illustrated in the figure 2. From this procedure, it is possible to define the lower limit of the PBL height and the upper limit where the FT should start, and a small layer in between. This latter layer was first named "injection layer" but since this term is ambiguous we changed it in "interface layer" to avoid misunderstandings.

RC: Page 8257, line 12, please explain "particular local environment".
AR: By "particular local environment", we meant that the PdD station is surrounded by specific characteristics (for instance vegetation types). This is now better explained in the text.

RC: Page 8258, lines 2 - 5, there are some numbers given with uncertainties, such as formation rates for charged and neutral particles as 0.071 +/- 0.087: I think, 3 decimal digits are not reasonable in this context. Please rethink the number of digits given here!
AR: This is right, we now limit the number of digit to two.

RC: Page 8259, line 16/17: What do you mean by "the model cannot describe local air mass motion such as topographical effects..." I think the model contains information about topography.
AR: Yes, the topography is included in the model, but with a resolution such that the topography is "smoothed" and does not reflect the reality. This is now mentioned in the text more explicitely.

RC: Page 8260, line 19: The acronym "PdD" was used for the first time here? If yes, it should be defined.
AR: This is right, we now defined the acronym "PdD" in the "Measurement sites" section.

RC:Figure 5: the labels are too small and the quality of the figure might be improved. What is shown there? The caption says negatively charged particle size distributions and particle size distributions...
AR: Sorry for this error due to a mixed of different versions of the paper. We corrected the caption which is now: *Average temporal evolution of the negatively charged particle size distribution for the puy de Dôme (upper panels of grey boxes) and Opme (lower panels of grey boxes) in case of "O" events (upper left box), "P" events (lower left box), "D_S" events (upper right panel) and "D_D" (lower left box).*
We enlarged the labels and increase the resolution of the figure to help the reader.

RC: Page 8261, line 24: What do you mean by cloud presence? Simply clouds yes or no, means if the station was in-cloud or not? Or anything else?
AR: We wanted to mean "in-cloud". The term "presence" is not appropiate and "cloud presence" is now replaced by "in-cloud" conditions.

RC: Page 8263 line 12: what is meant by: the presence of condensing vapours is less a limiting factor... This sentence should be rewritten in a clearer form.
AR: Contrarily to the JFJ, the PdD is surrounded by a coniferous forest which is known as a source of many volatil organic compounds. The VOC emmitted can lead to the formation of condensable vapors. Thus we made the assumption that at the PdD, the condensable vapor concentration is likely to be higher than at the JFJ. and hence that its presence may not be as limitant as at the JFJ to trigger the nucleation. The text is now:
"As a consequence, it is likely that, at the PdD station, the condensable vapor concentration is not as a strong limiting factor to trigger the nucleation as at the JFJ station, because it is always present at relatively High concentrations due to the proximity of the vegetation. In this VOCs enriched environment, a low CS becomes the main condition for nucleation to occur."

RC: Page 8264, line 12 ... when the nucleation is onset. This is not a correct sentence to my understanding, please rewrite. AR: We changed "when the nucleation is onset."
"As for the 12 August 2007, the two sites always seem to be located within the planetary boundary layer when the nucleation is onset."
changed to
"In case of "O" type events, Opme and PdD seem to be both within the PBL at beginning of the nucleation process."

RC: Page 8264, line 17 Where are the values for the nucleation period? I do not see it in the figure 8?
AR: During the nucleation period, between 09:00 and 11:00, $RH = 100 \pm 0.04$% and $LWC = 0.07 \pm 0.05 \mathrm{g.m^{-3}}$ on average, which indicates that the PdD station is mainly in "in-cloud" conditions when the nucleation is detected at the Opme site. This has been added in the text.

RC: What do you mean by "same atmospheric layer"? The development of the boundary layer should be discussed here in more detail. I think, differences between the two sites are strongly connected with differences regarding the PBL, i.e. if the station is within the mixed layer or above. I think this is the key for any difference.
AR: We made the assumption that within a given atmospheric layer (PBL, FT), the evolution of thermodynamical properties and the value of θ_e are homogeneous. We used the expression "same atmospheric layer" two times in the text. The first time is when we analyze "O" type events:
"This could indicate [i.e. the difference between the θ_e at both site but the θ_e correlation] that the two sites are not in the same atmospheric layer but in two different layers which are connected or in the same inhomogeneous layer".
changed to:
"This could indicate that both sites are not located within the PBL, unless this layer is strongly stratified thus inhomogeneous."

The second time we used the expression "same atmospheric layer" is in the analysis of the D_X and D_S cases.
Two different sub-cases could be pointed out: (i) the two sites are located within the same atmospheric layer when the nucleation is triggered (47 events), and, (ii) the nucleation is triggered at one site and then through atmospheric mixing, the nucleating air parcel is transported to the other site (40 events).
changed to:
Two different sub-cases could be outlined: (i) the nucleation is triggered at the same time at both sites (47 events), and, (ii) the nucleation is first detected at the low elevation station (Opme) and after a variable time delay, the event is detected at the altitude site (PdD) (40 events).
And:

the two sites are not exactly located in the same atmospheric layer. However, and contrarily to what is observed on "P" cases, the two sites are strongly connected: the puy de Dôme station is located inside the injection layer while Opme station is within the lower boundary layer.
changed to:
The LIDAR signal analysis (Fig. 12) confirms what is deduced from the analysis of the temporal evolution of θ_e. As seen in Fig. 12, the air parcels at the two sites are strongly connected: the PdD station is located inside the lower part of the interface layer while Opme station is within the planetary boundary layer. Moreover, during those cases, the PBL is not very polluted and well mixed. The nucleation events occur during the PBL development when the PBL influence on the interface layer is maximal.

RC: Page 8265, line 25 What do you mean by:..change in atmospheric properties... Please clarify!
AR: The "change in atmospheric physical properties" that we mention in the text in the "P" events section is refering to the sudden variation in the equivalent potential temperature time evolution θ_e. So it refere to what could influence this thermodynamic tracer: T, P and RH. We change "physical properties" into "thermodynamical properties (T, P, RH)".

RC: It would be great to see if the occurrence of nucleation events at both sites depends on the season, i.e. the typical boundary layer height. This should be discussed by the authors as well.
AC: This is right. Unfortunatly, as it is mentioned in the manuscript, due to many discontinuities of the measurements at the Opme station, a seasonnal variability analysis can only be performed for the PdD station. Furthermore, not enough LIDAR data are available to produce a seasonality of the boundary layer structure in relation to nucleation events. But measurements are still on progress and this will be investigated

as soon as the data set will be statistically relevant.

RC: Conclusion: First, this is more a summary and this should be mentioned in the caption. What numbers are given here? Mean + stdev? Or error? This should be explained, maybe also earlier. Again, the given accuracy here is not reasonable, e.g a boundary layer height of 1453+/-520.
AC: All the numbers given in this section are mean values associated with their standard deviation. We add this precision in the text. We change the accuracy boundary layer height, thus giving a 10-m resolution as suggested in the comment. The conclusion has been modified and a discussion was added.

RC: Here, also the relation to the boundary layer development should be discussed. This is really important to understand the variation at a high altitude site. Furthermore, the special case of a mountain site in relation to airborne measurements should be mentioned and discussed. NPF processes occurring at e.g. inversion layers cannot be studied at mountain sites.
AC: We added a discussion in the conclusion section.
"In the puy de Dôme measurement area, the PBL development is less sharply defined than over flatlands due to complex topographic effects on atmospheric dynamics. The boundary layer development must be analyzed for each type of event: "O", "P" and "D_x". Since no LIDAR data were available for "O" type events, only "P" and "D_x" cases are analyzed. During "P" events, the puy de Dôme is located in the upper part of the interface layer, between the FT and the PBL, while the Opme station is located within the PBL. During those cases, the PBL is relatively clean (based on the LIDAR signal) and well mixed. The nucleation events occur during the PBL development when the PBL influence on the interface layer is maximal. On the contrary, it was found that in "D_S" cases both sites are likely to be located in the planetary boundary layer or in the lower part of the interface layer. This latter result highlights that the nucleation occurs

within the whole lower tropospheric column, from the interface layer to the ground level. During the non-simultaneous events, the D_D cases (24.8% of total observed events), the PBL is much more polluted and the nucleation primarily occurs at low altitude before being detected after a various time delay at the high altitude suggesting a transportation of the nucleation process. In those particular cases, the nucleation process at the high altitude station is strongly linked to the PBL development since the nucleation is detected at the high altitude site when the PBL extend over the puy de Dôme measurement station.

The vertical extend of nucleation and growth events observed in this study exhibit some similarities with the one detected during previous airbone studies. During the EUCAARI field campaign, Crumeyrolle et al. (2010) have shown that the vertical extension of the new particle formation events do not exceed the top of the boundary layer and that most of the nucleated particle events observed have not been formed at the surface but must have nucleated elsewhere in the boundary layer suggesting that the nucleation process could be enhanced by the elevation. In a similar manner, during the IMPACT campaign (Wehner et al., 2010), authors have demonstrated that the nucleation likely occurs at high altitude, in turbulent zones of the residual layer.
Our findings demonstrate in a statistically relevant approach that the nucleation process is more frequent at high altitude site and occurs twice as frequently as actually detected in the PBL. Different assumptions can be made in order to explain why nucleation is more frequent at the high altitude site. The lower temperatures at higher altitudes increase the saturation ratio of condensable species (for a given concentration). Furthermore, LIDAR data show that the particle concentrations and hence the condensational sink decrease with the altitude. Although we do not know about the vertical gradient of condensable vapours concentrations, the two predicted gradients tend to favor the nucleation and growth processes. Another possible reason why nucleation is more frequent at the PdD station might be mixing processes at the interface between the PBL and the FT. At this interface, turbulence might be increased

due to the different properties of the two distinct atmospheric layers. As already proposed by Wehner et al., this turbulent mixing could lead to local supersaturation of condensable vapor which in turn could enhance the nucleation process. In addition to this statistical analysis, different vertical scales of nucleation have been pointed out: (i) the nucleation only occurs at high altitude above the mixed layers, between the PBL and the FT in the upper interface layer; (ii) the nucleation occurs at the same time along the whole the PBL and in the interface layer and (iii) the nucleation is triggered within the low PBL and then transported to highest altitude during the PBL development. Those observations suggest that the nucleation and subsequently growth process could have a very large vertical extent, at least up to more than 800 m. However, this work only provide a lower boundary value since no measurements were available above the puy de Dôme station. Thus complementary airborne studies are needed especially to characterize what happen in the free troposphere when the nucleation is triggered in the planetary boundary layer and in the interface layer. Furtur campaigns would allow to check if the nucleation extends over the planetary boundary layer–free troposphere limit or if the phenomenon is vertically constrained as suggested by the work of Crumeyrolle and co-workers."

RC: Page 8268, line 20/22 and later: nucleation should be replaced by new particle formation. In fact, nucleation is not really observed here, because this is just the first step and influences the very small particles only.
AC: Instruments we used to track the nucleation events, AIS and NAIS, measure charged and neutral particles from 0.8 to 42 nm in NTP-conditions. Thus we think it's justified to use the term nucleation since recent studies confirm that the size of the critical cluster sould be around 1.5 nm (Vuollekoski et al., 2010). All event we present here are nucleation and subsequently growth events.

RC: Page 8268, line 7: talking about atmospheric composition is a bit challenging

for the presented study, because there are not many measurements available. This should be clearly stated. Reading atmospheric composition I expect very detailed measurement of chemical species.
AC: Unfortunately we only have few measurements of the atmospheric composition (see the measurement site section). We did not pretend to give an exhaustive description and we try to connect what we measure in term of composition at the station with the studied nucleation events. The term was probably not appropriate and we renamed the section "Relationship to other atmospheric parameters".

RC: There should be a bit more discussion. Why is the NPF more frequent at the high altitude site? At least some speculation should be possible. This is really necessary to add because otherwise the conclusions contain mainly a description and summary.
AC: Different assumptions can be made in order to explain why NPF is more frequent at the high altitude site. The lower temperatures at higher altitudes increase the saturation ratio of condensable species (for a given concentration), . Furthermore, LIDAR data show that the particle concentrations and hence the condensational sink decrease with the altitude. Although we do not know about the vertical gradient of condensable vapours concentrations, the two precited gradients tend to favor the nucleation and growth processes. Another possible reason why nucleation is more frequent at the PdD station might be mixing processes at the interface between the PBL and the FT. At this interface, turbulence might be increased due to the different properties of the two distinct atmospheric layers. This turbulent mixing could lead to local supersaturation of condensable vapor which in turn could enhance the nucleation process. This phenomenon has been already observed in the recent paper of Wehner et al. (2010). These authors have shown that new particle formation likely took place in turbulent zones of the residual layer. This discussion has now been added to the text.

AC: We corrected all mentioned the typos and check all the manuscript and captions.

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