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Reply on RC2

Janne Lampilahti et al.

Author comment on "Zeppelin-led study on the onset of new particle formation in the planetary boundary layer" by Janne Lampilahti et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-282-AC2>, 2021

This paper describes new particle formation event observed in a rural (SMEAR 2 station) and urban (Po Valley) environment using instruments installed on the ground and in a Zeppelin. As claimed by the authors, "the main goal of the manuscript was to quantify the magnitude of regional to global feedbacks between atmospheric chemistry and physics, and thus quantify their impact on the changing climate". However, I don't think this was achieved within this manuscript. Moreover, it's not clear to me why the authors choose to compare event at both sites. Of course, both environments lead to different situations favorable for NPF. However, the authors are comparing one study case for each environment picked out from short term campaigns. Why these two cases are representative of NPF events in both environment?

ANSWER: The paragraph in the introduction that says

"Here we present NPF measurements on board a Zeppelin airship carried out during the EU supported PEGASOS (Pan-European Gas-AeroSOls Climate Interaction Study) project. The main goal of the project was to quantify the magnitude of regional to global feedbacks between atmospheric chemistry and physics, and thus quantify their impact on the changing climate. The Zeppelin flights were used to observe radicals, trace gases, and aerosol particles inside the lower troposphere over Europe in several locations during 2012-2013"

is referring to the PEGASOS project goals. The main goals of the current manuscript are:

"Here we combine comprehensive ground-based and airborne measurements from the Zeppelin to compare two NPF cases from Po Valley to one case from Hyytiälä. The Zeppelin allowed us to repeatedly profile the lowest 1 km of the atmosphere providing a full picture of what is happening in the BL during the onset of NPF. We will show in which part or parts of the BL the onset of NPF and the subsequent particle growth occurred at the two measurement sites as well as determine formation and growth rates for the aerosol particles."

We removed the sentence "The main goal of the project was to quantify the magnitude of regional to global feedbacks between atmospheric chemistry and physics, and thus

quantify their impact on the changing climate." from the first paragraph so that there would be no confusion.

Detailed measurements during NPF events using the nucleation payload instruments were only obtained on these couple of days. We do not argue that the NPF events we measured represent how the average NPF events occur in the BL, we only point out that the timing and meteorological conditions were typical of NPF events at these locations. Despite this limitation we believe this manuscript gives valuable insight to our understanding on how BL processes can affect NPF at the studied locations.

Major comments

I regret that the authors cite papers mostly from their group. There are other high quality papers out there working on NPF events analysing the vertical extension of NPF event, looking at the link with turbulences...

Figure 5 : I'm not sure I agree with the author's conclusions on that figure. On the SPC side, there are Ultra Fine (UF) particles in the residual layer during the early morning even before the sun rise (4:45). Moreover, during the whole morning the UF particules can be seen at the top of the ML within the ML and in the TL. The NPF onset at the ground is clearly between 5:30 and 6:00. At 5:00, measurements from the zeppelin are showing high concentration of UFP below 200m. It looks like this specific event may be caused by turbulence mostly. On the other side (HTL), clearly the NPF event started at the ground at 7:30. There are no signs of UFP at the ML top but some UFP lies at 2km according to CESSNA measurements. The authors claimed (L. 507/L. 433), based on previous publications, that event occurring at 2km could be linked to turbulence occurring between residual layer and free troposphere and that both events (2km and ground) could be linked but I don't understand how these could be linked... Please do tell ! Moreover, there are no evidence that the interface RL/FT is located at 2km, nor that both events are linked...

ANSWER: There seems to be some confusion with the figure numbers, I believe this is referring to Fig 4.

We agree that in the SPC case turbulent mixing plays a crucial role in the onset of NPF. In the manuscript we suggest that when turbulent mixing starts and the ML begins to grow sulfuric acid, which appears to be concentrated in the RL, is mixed to the surface from the RL. At the surface ammonia and other precursor gases like amines and organics are present and in the resulting mixture NPF starts.

In HTL case we argue that the sub-10 nm particles at 2 km observed from the CESSNA are not linked to the NPF event inside the ML on that day, instead they are probably two separate NPF events. The atmospheric layers above the ML are not clearly visible during the Cessna profile, however observations of such particle layers have been linked to the RL-FT interface over Hyytiälä and therefore we provide it as a possible explanation.

There are no date in Figure 5 label and I think this is missing...

ANSWER: We added the dates in the caption of the Figure (assuming again it is Figure 4)

L 388 : « In the ground-based NAIS data a pool of sub-6 nm particles was present during

the NPF event... This can be seen most clearly between 10:00-11:30 .. "

I believe you were referring to Figure 7b and d at HTL (not HYY as written on the figure). I don't clearly see it from 10:00-11:30 but I do see it during the whole day, right? Again, this rises question about how the events were created. So in SPC, the particle mode is really large always including 3nm particles during the full day suggesting that the event is all over the ML and therefore is not comparable to the HTL event that is observed at the ground and newly formed particles grow and may be then transported into the ML.

ANSWER: It seems that in HTL the formation of the smallest particles might be increased close to the forest canopy, which makes sense since the boreal forest is a source of the organic precursor vapors. Leino et al. (2019) observed increased sub-3 nm particle concentrations towards the surface in HTL even on days when no NPF event occurred. This particle concentration gradient was less pronounced during NPF event days. In HTL the particles could still form throughout the ML, but the formation rate could be slightly higher close to canopy. We changed HYY to HTL in the figure.

L 445 – 452 "The concentrated vertical stripes over the growing nucleation mode in Figure 6b were caused by theAre linked to roll vortices, which are a specific mode of organized convection in the BL. " Could you please take some time to prove it ? As you are comparing two events in different environment I think that you should carefully address how those events appear ...

ANSWER: The location and movement of the particle zone matched that of roll vortices over the measurement area (a boundary layer deep longitudinal zone, less than few km wide, that moved over the measurement site perpendicular to mean wind). This case was analyzed in the paper cited in the manuscript (Lampilahti et al., 2020). In that paper specifically look at Figure 11. More analysis related the Figure 11 was presented in a response to a reviewer (<https://doi.org/10.5194/acp-2019-1013-AC1>: see figures 3, 4 and 5).

Conclusions :

"We compared two different environments where NPF occurs frequently: a suburban area in Po Valley, Italy, and a boreal forest in Hyytiälä, Finland. We aimed to answer in which part of the BL the onset of NPF and the growth of the freshly formed particles takes place and studied the vertical and horizontal extent of NPF. "

Again why choosing two different environments and only two study cases to answer that question ? I would think that more statistical information would be needed ...

ANSWER: We acknowledge that these observation may not represent the typical NPF event at the sites. To make it more clear we rephrased the paragraph to read:

"We compared case studies from two different environments where NPF occurs frequently: a suburban area in Po Valley, Italy, and a boreal forest in Hyytiälä, Finland. We aimed to answer in which part of the BL the onset of NPF and the growth of the freshly formed particles took place and studied the vertical and horizontal extent of NPF."

Minor remarks

P4 L 104 : rephrase : 'Compare from nucleation '

ANSWER: Changed the whole sentence to: "The two ground-based measurement sites that were studied here were San Pietro Capofiume in Po Valley, Italy and Hyytiälä in Southern Finland."

P9 L 272 : "ML started to increase in height" could you please highlight where this is coming from ? Lidar, ceilometer measurement of in flight measurements ?

ANSWER: Yes, changed to: "according to the ceilometer measurements the ML started to increase in height"

P9 L 300 : "no NPF, was observed above the ML" : remove the comma

ANSWER: Done

Figure 6 is hard to read. The Zeppelin measurements were performed at different altitude and it does not appear. Could you please either remove it either include the altitude on that figure ?

ANSWER: The altitude is now included in the figure.

Figure 7 : always add the altitude to any on-board measurements

ANSWER: The altitude is now included in the figure.

Figure 7d : from 8am to 9am there is no black dot. So the GMD is over 30nm ? so there is a clear interruption of this event ! Could you comment on that ? What does that change for your study or for the event in general ??

ANSWER: At the SMEAR II station between 7:30-08:00 we observed the localized NPF event that was linked to the organized convection and moved over the station with the airmass. The black points after 09:00 were linked to the regional NPF event that was taking place at the same time, the mode fitting method could not determine a clear mean mode diameter before 09:00 for the regional NPF event.

References

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