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Reply on CC1

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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-AC3, 2021

Comment on the manuscript:

Zeppelin-led study on the onset of new particle formation in the planetary boundary layer by J. Lampilahti et al.

The manuscript describes airborne data from the Zeppelin aerosol measurements during the Pegosos Campaigns in San Pietro Capofiume in 2012 and in Hyytiälä in 2013. The results are highly interesting and could be a useful data set to trace back the appearance and origin of nucleation mode particles in the atmosphere or, to characterize air masses that contain precursor material, as proposed in the abstract. In both cases, in SPC and in HTL the data on the scale of the spatial distribution suggest a significant contribution of horizontal transport on top of the diurnal cycle of vertical convection. An interesting new result is the signature of sulphuric acid found above the MBL before convection mixed a larger MBL volume. Getting into more detail a chemistry transport model would be useful, not included in the current study.

A further well notified result is, that despite transport over major agricultural areas in the center of the eastern Po-Valley, west of Ferrara enhanced nucleation aerosol was not observed. This is a bit surprising after to the results of Kontkanen et al, 2016, who found NPF events each day with only one exception, a paper suggesting that NPF is more a general feature in the Po-Valley.

However, focusing on the current manuscript, the title promises new results on the 'onset of new particle formation in the planetary boundary layer usig an airborne platform (Zeppelin)'

For such an investigation a precise time and location of the data points would be necessary. Unfortunately these data are not presented and on top there are several obvious timing problems clearly visible in the figures:

Fig. 2, Trajectories were calculated for 12:00 UTC (see HYSLIT info in the plots, not for the morning as claimed in the figure caption). Already 2 hours difference might be critical for the wind direction and trajectory. Also, for a process occurring in a diurnal cycle with short lived compounds a trajectory for 72 h does not make a lot of sense.

ANSWER: The information in the HYSPLIT figures is a bit confusing, the different colored trajectories are for different hours of the morning. We updated the figure to address this.

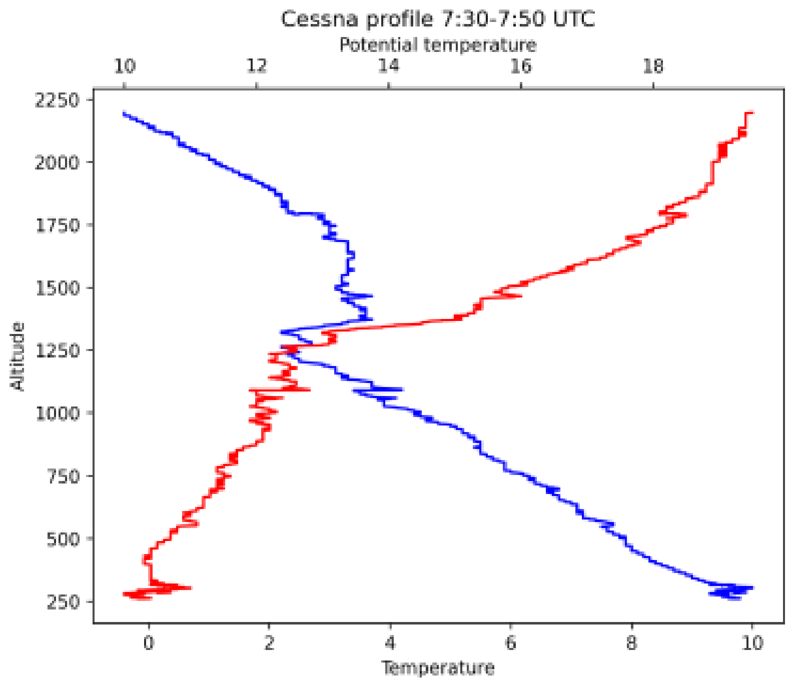
Fig.3, Time axis in the diurnal plots, is claimed to be 'UTC'. The geographical location of

San Pietro Capofiume is 44°39'N,11°37'E. This should result in a peak solar radiation about one hour (1h for 15o) earlier than 12:00 UTC (see also Kontkanen et al 2016, where the same radiation data are presented with the correct timestamp, UTC+1)

ANSWER: This makes sense, even though the data readme says UTC the data is probably in UTC+1 (local time). We shifted the time by 1 hour backwards in the diurnal variation plots where it was needed (meteorology, radiation and SO2 concentration).

Fig. 4, timing problem in the MBL development. Despite less incoming radiation the MBL growth in Hyytiälä is faster than in SPC. A more realistic temporal evolution of the MBL using HYSPLIT data results in a MBL of +1200 m at ~ 09:15 UTC instead of 07:15 UTC. This finally leads to a different interpretation of the HTL data.

ANSWER: The figure below shows the Cessna climb between 7:30-7:50 UTC. The top of the BL indicated by the temperature inversion and stable layer appears to be at 1200 m asl, already before 9:15 UTC. The BL height at 7:15 was determined from a Cessna profile where the airplane climbed only to about 1400 m asl and the top of the BL was not as clear, so we left this first BLH data point from the Cessna out. This does not lead to different interpretation of the results though.



Fig, 6, a) SPC, position data for June 28 are missing, compared to Fig. 9, horizontal position is important. b) HTL Data in Fig. 9 do not support the conclusions in the text. Low wind situation with a good chance to get into self-contamination problems in circle flights. The time spent for half a circle (20 m/sec and ~4 km diameter) is close to air mass transport time across circle. Timing requirements for such a local case study are even more stringent than just using the correct time zone. Even the internal timing of the NAIS or SMPS scanning loop becomes important (Manninen et al, 2016). Is the observation of 3-10 nm particles in agreement with the growth rate and diurnal pattern of > 10 nm particles at HTL in Fig. 3?

ANSWER: Unlike in HTL no clear horizontal variation was observed in SPC on June 28, 2012 on board the Zeppelin. The Zeppelin was measuring inside a small area less than

few km in diameter in order to capture mostly vertical variation. Adding a horizontal Zeppelin track for June 28, 2012 would not bring much new information which is why we do not include it.
We do not think there was a problem with self-contamination on May 8, 2013 since the zone of sub-10 nm particles was observed on board the Zeppelin, Cessna and at the SMEAR II station, separated more than couple of km from each other. Also the particle zone was not more concentrated in the downwind section of the Zeppelin's flight track, instead it appeared to move slowly approximately from soutwest to northeast (see Figure 3 in https://doi.org/10.5194/acp-2019-1013-AC1). The particles were similar size and grew at the same rate with the regional NPF event particles which would not be expected if it was pollution.

Fig. 8, unprecise timing, missing coordinates, comparison with HYSPLIT GDAS temperature and MBL data suggests a profile at ~ 09:00 instead of 07:15, see also Fig 4. Air time of the Cessna (07:00 – 10:00 UTC?) is questionable.

ANSWER: The GPS coordinates for the Cessna flights can be seen in the next figure. The profile includes data between 7-10 UTC, so there is data from 9 UTC also.

Fig. 9, GPS coordinates, altitudes and time missing. a) It's not clear, how and whether are the two patterns in Fig. 9a from Zeppelin and Cessna are coordinated? Wind direction is also changing during the morning by ~ 20 degree within 2 h (HYSPLIT). Fig.9b) Where and at what altitude has the Zeppelin been between 05:00 and 10:00? 5 hours airborne are ~ 360 km, the loop is only 140 km.

ANSWER: The Cessna and Zeppelin flights took place at the same time. The flight patterns were coordinated to minimize downwind pollution interference.
The wind direction is the mean wind direction during the measurement at the SMEAR II station. The change in wind direction probably affected the movement of the sub-10 nm particle zone over the area (again see Figure 3 in https://doi.org/10.5194/acp-2019-1013-AC1). The purpose of this plot is to show horizontal variation. The loop around Ferrara and the return to the airfield was flown at constant altitude at roughly couple hundred meters above sea level. In the HTL case the zone with higher particle concentration was observed at all altidudes inside the ML.

Fig. 10, air mass back trajectories not matching the position of the Zeppelin. The Zeppelin was airborne that day (June 30) for more than 5 hours (see text) flying at 20 m/sec. The trajectories plotted, however, were obviously not calculated for the time, location and altitude of the actual measurement. The spacing is not in agreement with the operational cruising speed of the mobile platform.

ANSWER: We chose trajectory arrival points in the south and north sectors of the measurement area at 400 m asl altitude and calculated the airmass trajectories arriving to these points at 8:00 UTC. The NPF event started at 8 UTC in SPC, at that time the Zeppelin was also measuring about 5 km north of SPC and observed the starting of the NPF event. Then the Zeppelin began to fly north at roughly 200-300 m asl in order to explore the horizontal extent of the NPF event. Almost immediately when moving north the NPF event was lost. To reach the north point of the flight track took roughly 30 min. The point of the trajectory analysis was to see if the south sector where the NPF event was taking place had different airmass origin than the north sector. The current analysis confirms this even though the trajectory end points are not exactly chosen on the flight path. We added information on the trajectories in the caption.

Summary:
A three dimensional airborne study as presented here on a time critical process, 'the onset', requires a clear temporal and geographical identification of data points and a

timely correct trajectory analysis. This is unfortunately missing in this manuscript, or, where it is at least partially available, often and obviously false due to incorrect time settings.

ANSWER: The issues presented were addressed. We think the figures are precise enough to support the conclusions.

Finally the abstract claims: 'In Po Valley we observed NPF that was limited to a specific air mass', however, the air mass is neither specified nor characterized in the text. Air mass origin and composition in the text is speculative.

ANSWER: A full analysis with chemical transport models etc. to more completely characterize the airmass is outside the scope of this study.

The reference list is incomplete. Citations are missing for:
Dada et al, 2017, Dunne et al., 2016; Gordon et al., 2017; Yu and Luo, 2009, Mohr et al, 2019, Pierce and Adams, 2009; Stratmann et al, 2003, Junkermann and Hacker, 2018; Sullivan et al, 2016; Vogel and Elbern, 2021

ANSWER: The bibliography has been updated.

However, there are a few more issues that came up reading the manuscript not addressed but likely worth an investigation with a high resolution chemistry transport model:
Interestingly, NPF is linked to convection rather than to air chemistry, how can this be explained? Convection starts with global radiation, air chemistry with UV radiation, respectively photolysis, about 1.5 h later and subsequently it should also take some time for the fresh clusters to grow into measurable particles (Kulmala et al, 2013).

ANSWER: Nilsson et al. (2001) provides some explanations.

What is the origin of the sulfuric acid (SA) at a time of the day (early morning) when there is not yet sufficient UV radiation to produce OH radicals for sulphur dioxide conversion and why is SA observed first in the 'clean' residual layer? What is the corresponding chemistry? The maximum solar radiation flux for June 30 is only ~ 10% different for SPC/Bologna (896) and Monte Cimone (976). Under conditions with more SO2 pollution, in the boundary layer the formation of sulphuric acid (SA) thus should be more intense.

ANSWER: The pseudo SA concentration starts to increase after sunrise, which was at ~4:30 UTC. SO2 concentration is probably low at the surface due to deposition, but remains high in the RL.

Why are nanoparticles not observed in the north-western loop, north of the fiume, despite an advection of an air mass passing over the areas with intense agricultural activities and more intense ammonia emissions (see the Italian emission inventory, Taurino et al, 2020). Contrary, nanoparticles were observed the same day June 30 in air masses travelling over forested mountains southwest of Bologna. A contribution of the city of Bologna is unlikely based on HYSLIT trajectories for the day as well as a contribution of the Venice area as speculated in the text.

ANSWER: Perhaps in the northern airmass there were also other emissions that prevented the formation of aerosol particles. A more detailed investigation would be needed to fully understand this day.

References:

Nilsson, E. D., Rannik, Ü., Kulmala, M., Buzorius, G., and O’dowd, C. D.: Effects of continental boundary layer evolution, convection, turbulence and entrainment, on aerosol formation, Tellus B, 53, 441–461, https://doi.org/10.1034/j.1600-0889.2001.530409.x, 2001.