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Interactive comment on "Airborne observations of newly formed boundary layer aerosol particles under cloudy conditions" *by* Barbara Altstädter et al.

W. Junkermann

wolfgang.junkermann@kit.edu

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The paper shows some interesting data on elevated ultrafine particle layers under total overcast conditions obtained, using an unmanned aerial vehicle, the UAV ALADINA, flying vertical profiles through the planetary boundary layer and lower free troposphere. The occurrence of ultrafine particles under such meteorological conditions would not be expected following the general description of new particle formation observed elsewhere. The authors nevertheless, claim a clear identification of a new particle formation event. Typically for the production of ultrafine particles via new particle formation or, with the old expression, gas-to-particle conversion, shortwave radiation is suggested to

be required as an initial step to convert first sulphur dioxide into sulphuric acid, which than reacts in the atmosphere often with ammonia or similar compounds producing initial aerosol clusters. This is well established in a large number of publications, see for example for a review Kulmala et al, 2011, and Kulmala et al, 2013 for an analysis of the expected particle generation process. Growth occurs then via (EL)VOC, further sulphuric acid water etc. . The data shown here were measured under conditions without sufficiently high UV radiation levels to produce sulphuric acid in the atmosphere, thus requiring a different production process. Such an occurrence of ultrafine particles under totally overcast conditions has been observed several times and was reported for example over Germany (Junkermann et al, 2016), a paper that is cited in the manuscript. Here these particles were apportioned to primary industrial sources burning fossil fuel. Hence, as several of these elevated sources are located around Melpitz the observations are not really surprising. Anyway, the data analysis leaves several questions

1) The manuscript states that NPF occurrence with subsequent downward transport was clearly identified.

The question: What does 'new' mean within this context? Was the particle formation (cluster production and subsequent growth) really been observed?

How can a nucleation (gas to particle conversion) be identified with instrumentation that is not able to measure the initially formed particles? From the two size fractions the size of the major nucleation or Aitken mode cannot be estimated. It could be anything larger than 12 nm and only shows, that the distribution contains some particles in the range below 10 or 12 nm.

A gas to particle (NPF) event however, should have as a minimum requirement initially the majority of particles in the lowest size bins of the distribution. The number of particles in the size between 7 and 12 nm (or N7-12) has then to be higher than the number of particles > 12 nm before and at the time the growing nucleation mode is

passing this size range. Here, within the data presented the difference between the size bins is marginal, compared to the overall number concentrations indicating rather an aged air mass. Occasionally the N10 is even higher (measurement uncertainty?) than N5., in Fig. 4b and 4c N10 is missing at all.

2) Following Kulmala et al, 2013, it takes several, $\sim$ about five or more, hours for a new particle to grow into the lowest size range of the particles that can be observed with the instruments on ALADINA. Observation of particles at 08:00 UTC thus requires a production at$\sim$ 03:00 UTC or even earlier during the night. These particles have to be generated in a different area, a minimum of five hours upwind, which is according to HYSPLIT at least 70 km. Growth rates are normally smaller or even zero at night. Thus the source could be even further away.

3) Where is the production area of the particles, which are 'clearly' identified as originating from NPF, according to HYSPLIT $\sim$ 70-100 km for 5 h?

4) Why are the meteorological and atmospheric chemical conditions favorable for new particle formation > 70-100 km upwind, during the night and in the elevated layer? Where do the precursor chemical compounds come from? What could be the initial step for nucleation cluster generation at night?

5) Is there probably a source for ultrafine particles upwind that could explain the results? HYSPLIT shows that the air mass has been close to the German-Polish border on the evening of April 3. Here we have at least three power stations that are sources for primary particles in the respective size ranges as well as additional large amounts of sulphur dioxide, ammonia (from the SCR cleaning process) and also internally produced sulphuric acid (see also Junkermann et al, 2016). Size distributions, independence on time of the day (Junkermann and Hacker, 2015) and laboratory results confirm primary emission of 'New' particles (Brachert et al, 2013). Such particle production does not require UV radiation or OH radicals.

A quick HYSPLIT analysis: For June 21 the winds in the altitude of 600 m above

ground come from the northwest passing the power station Buschhaus as the next possible candidate. Further upwind (up to 24 h) along the backtrajectory the industries of Groningen (NL) are located. For westerly to southwesterly winds in the PBL the power stations Schkopau and Lippendorf as well as the refinery Leuna would be possible sources for primary ultrafine particles and further precursors over Melpitz (see also Platis et at, 2015). For size distributions and source strength of such sources see Junkermann et al, 2011; 2016.

6) Page 9, line 20, atmospheric boundary layer conditions were unfortunately not available from the UAS. Instead a model is used to characterize the ABL. However, this model is not able to reproduce the measured vertical structure (Fig. 9). Why are no model data shown for 07:00 to 08:00, the time window before the aircraft measured the pronounced vertical profile? Lateron the PBL rapidly mixed. The vertical structure was visible only within the first 15 minutes of the model 1 h time window.

7) Fine particles: particle number concentrations > 390 nm on April 4 are fairly high. We can compare to particle numbers measured with a GRIMM 1.108 (fraction > 400 nm, second size bin) over Kathmandu (missed approach to Kathmandu international airport) in January 2014, to data gathered during flights in the extremely dusty Mexico City basin during Milagro 2006 or to data from the heavily polluted Po-Valley (QUEST, 2004). The number concentrations there were only about half or less of what has been seen over Melpitz. Under these conditions the condensational sink (CS) would be extremely high and according to most of the published literature a NPF event would be very unlikely. How is this in agreement with the summary / abstract statement that New Particle Formation has been clearly observed?

The summary claims: Further, the NPF event was 5 linked to a condensation sink of larger particles belonging to the accumulation mode at the same altitude. That's not in agreement with the fig. 4, the layers are clearly separated.

8) Fig. 9 and page 11, line 22/23: 'The maximum of total aerosol particle number

concentration was 1.6×105 cm−3 at the altitude of 420ma.g.l. and increased rapidly while descending'.

Not in agreement with the figure. The number concentration below 420 m actually is always lower than the maximum at 420 m and decreased between 08:13 and 08:26 UTC. Probably also a result of changing wind directions ($\sim$ 15 deg within 2 hours)

Summarizing: The paper does not really show evidence or proof for gas to particle conversion respective new particle formation. Though interesting and good to identify ultrafine particle layers the instrumentation used is most probably not appropriate to detect such particle formation events. An SMPS or NAIS would be necessary.

The results are contrary to the general literature about new particle formation and unfortunately, there is no attempt to analyze where the particles observed are produced or originating from. However, the measurements would be in agreement with emissions from a few welll known 'continuous generators for ultrafine particles' e.g. power stations /refineries and the final statement that a thorough meteorological analysis (but, not only in the vertical) is necessary to understand ultrafine particle behavior in the PBL is strongly supported.

References:

Brachert, L., Kochenburger, T., and Schaber, K., 2013: Facing the Sulfuric Acid Aerosol Problem in Flue Gas Cleaning: Pilot Plant Experiments and Simulation, Aerosol Science and Technology, 47, 1083–1091

Junkermann, W., Vogel, B. and Sutton, M.A. 2011: The climate penalty for clean fossil fuel combustion, Atmos. Chem. Phys., 11, 12917-12924

Junkermann, W., and Hacker, J.M., 2015: Ultrafine particles over Eastern Australia: an airborne survey, Tellus B, 67, 25308, http://dx.doi.org/10.3402/tellusb.v67.25308

Junkermann, W., Vogel, B. and Bangert, M., 2016: Ultrafine particles over Germany - an aerial survey, Tellus B, 2016, 68, 29250, http://dx.doi.org/10.3402/tellusb.v68.29250

Kulmala, M., and Coauthors, 2011: General overview: European Integrated project on Aerosol Cloud Climate and Air Quality interactions (EUCAARI) – integrating aerosol research from nano to global scales, Atmos. Chem. Phys., 11, 13061-13143, https://doi.org/10.5194/acp-11-13061-2011, 2011

Kulmala, M. and Coauthors, 2013: Direct Observations of Atmospheric Aerosol Nucleation, Science, 339, 943-946, doi: 0.1126/science.1227385 Platis, A., and Coauthors, 2015: An Observational Case Study on the Influence of Atmospheric Boundary-Layer Dynamics on New Particle Formation, Boundary-Layer Meteorology, DOI 10.1007/s10546-015-0084-y

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