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Atmospheric Chemistry and Physics Discussions

# *Interactive comment on* "Aerosol particle formation in the upper residual layer" *by* Janne Lampilahti et al.

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The manuscript describes aircraft based vertical profiles of nanometer sized particles in the range from 1.5 to 400 nm with a main emphasis on the lower particles sizes of 1.5 to 3 nm and 3-20 nm. The authors summarize data from 7 years of flight experiments and conclude that a larger fraction of ground based nanoparticle events (NPE's) (> 40% of observations) occurs in cases when small particles are transported downwards from the residual layer into the planetary boundary layer. A case study is shown as well for one day in May, 2017 to explain the interpretations. A schematic drawing of the proposed processes is included.

General comments



Although the paper is easy to read it's obvious that there is a significant lack of data supporting the conclusions in the manuscript, especially information about the history of the air mass under investigation and potential pollution. The title is misleading. The main message of the title, that this study is an investigation of aerosol formation is not justified. For such an investigation the CESSNA could be a suitable mobile platform but, completely different flight patterns and an upgraded instrumentation would be necessary (see below).

There is a second message behind, not mentioned in the manuscript title: Nanoparticles appearing at the surface as new particles are actually produced elsewhere and advected towards the ground under sunny conditions by a diurnal cycle of vertical transport. This is well I agreement with the observation that solar radiation is a major driving parameter behind the appearance of new nanoparticles at the surface (Baranizadeh et al, 2014). The authors now allocate the place where the particles are formed into the residual layer. However, they neglect that also in the residual layer a horizontal transport process is running, on a larger scale and less, but not completely independent of diurnal patterns. The argument, that particles might be produced elsewhere holds as well for this location / altitude range. Now, where are these particles produced?

The authors claim that nanoparticle formation appears to start in the residual layer, a statement that is not confirmed and that the processes linking transport and nanoparticle appearance are not well understood disregarding a century of atmospheric physics research. They do not discuss the production pathways, the contributing chemical precursors and their sources and the already available literature about gas to particle conversion in the atmosphere (e.g. Mohnen and Lodge, 1969, Gillani et al, 1978).

To investigate the origin of atmospheric nanoparticles, a sound 3D-meteorological analysis is mandatory. In the current manuscript such an analysis is missing. A few vertical profiles of temperature, potential temperature and water vapor are included. For these data scales and units are missing. Horizontal transport, wind (speed and direction), although mentioned in Fig 10 of the manuscript as part of the proposed mechanism for

new particle formation, is not taken into consideration at all. Previous studies on the production and transport of ultrafine particles are not adequately taken into account.

While vertical mixing can be a fast process with a time constant of less than an hour (Georgii, 1956) (a better estimate from the current investigation?) aerosol formation and growth is a process of several hours (Kulmala et al, 2013). Accordingly and in agreement with fig. 10 (from the manuscript) the history of the air mass at least for several hours need to be considered for particles in the size range < 20 nm. It's not important where the air mass is originated on the long term, it's more important, whether and how it is polluted on the way to the observation location and by which chemical or particulate compounds.

In the case study or May 2, 2017, relatively clean, regionally unpolluted air in the morning is compared to a polluted case after noon. The authors assumption of a clean arctic air masses (line 166) for all profiles during the day, a prerequisite for the conclusion that aerosols suddenly appear from gas to particle conversion is thus not valid. Thus, neither a local nor a spatial atmospheric aerosol formation can be derived from these early morning and afternoon profiles.

Fig. 1 shows the results of a HYSPLIT backtrajectory analysis for the may 2 case study and upwind pollution sources with elevated emission altitude ($\sim$ 200 – 400 m). Note the windsheer. The white circle is the 20 km radius range around Hyytiälä, the dotted line the 40 km radius. MBL at 05 UTC was at 260 m over Vaskiluoto, the main source in the area. The sulphur emission rate is $\sim$ 150 kg/h since installation of the desulfuration equipment in 1993 (http://www.energico.fi/ref-vaskiluoto.shtml), compare to 250 kg/h from the power station Karlsruhe and its primary nanoparticle emission (Junkermann et al, 2011a). Paper mills and smelters are similar sources for particulate emissions (Ayers et al, 1979, Bai et al, 1992, Rosenfeld, 2000, Junkermann et al 2011a, Brachert et al, 2013, see also the Finnish national emission inventory, https://www.eea.europa.eu/data-and-maps/dashboards/air-pollutant-emissions-data-viewer-3). Ammonia as a neutralizing compound is widely



used in the fossil fuel industry to suppress NO2 emissions and it is available in high concentrations internally (primary emissions) and also externally as ammonia slip (Li et al, 2017).

The main result of the investigation of the current manuscript is the analysis, that a significant fraction of Hyytiälä nanoparticle events are correlated to enhanced nanoparticle concentrations in the residual layer and are caused by downward transport of this nanoparticle aerosol, driven by thermal convection.

The statements in the conclusions about evidence for NPF and unique thremodynamic conditions (not shown) in line 276/278 about upper residual layer aerosol formation again are speculative and not supported by data. Although only occasionally reported (Kerminen et al, 2018 and literature cited therein) physically also shrinking of aerosols cannot be excluded. The fate of nanoparticles during transport depends on ambient conditions as well as on the presence of other aerosols (coagulation, condensation sink). This information is missing. The statement, that meteorology, but mandatory in 3D and including wind, has to be taken into account for interpretation and analyzing of ground based and airborne data (lines 279 to 281) and the statement that the current results are important for NPF events elsewhere in the world however, can be fully supported.

Questions:

What are the ambient conditions relevant to particle formation or aging in the residual layer, temperature, humidity, water vapor concentration, wind speed and direction, potential condensation sink? Are aging nano-particles in this layer growing or shrinking (Kerminen et al, 2018 and references cited therein)?

40 km is a wide range, see Fig. 1. Where is the GPS-location of the measurements with respect to well known locations of major precursor molecule and/or primary nanoparticle emissions upwind?

What is the flight pattern during ascents and descents? Can this be used to derive wind data from example from GPS when the Lidar is not sensitive enough?

Why are the measurements in the ascend beginning at 200 m, the descend ends at 600 m agl? Teisko, $\sim$ 15 km (alt 158 m) from Hyytiälä (alt 178 m) would be a location for missed approaches and legal low altitude flying. O'Dowd et al (2009) presented profiles nearly to the ground from QUEST 2003.

Are there any indicators for example from the Hyytiälä meteorological tower that can be related to vertical mixing intensity? Potential parameters could be surface temperature or temperatures in the vertical profile. Glider pilots use ground based temperature measurements for a decision when to take off.

Minor comments

Although an SMPS is onboard there is no size distribution presented for the case study. A complete size distribution would be a mandatory information for the interpretation as it carries information about the age of the particles (and potential distance to the source location).

For comparison of airborne and ground based data the same parameters, particle size distributions and not particles on the aircraft and air ions on the ground, should be used.

Whether the vertical profiles within 2 $\frac{1}{2}$ hours in the early afternoon and another flight in the morning are comparable at all remains open, see the HYSPLIT trajectories above. The vertical profiles of the morning flight including air mass history and trajectory need to be included as well.

To investigate, whether the 1.5 nm particles grow into the size range of 3-20 nm and to disentangle between NPF in a sulphur rich environment and primary emissions a better size resolution is necessary (Junkermann et al, 2011a). See there and in subsequent papers size distributions with a finer resolution in the range below 10 nm.



It needs a lagrangian flight pattern to confirm that airmass change is excluded, see Junkermann and Hacker (2015).

The observations in the 3-20 nm range are well in agreement with the patchy structure of particle number and size distributions from previous studies in the area (O'Dowd et al, 2009, Schobesberger et al, 2013, Väänänen, et al, 2016, Leino et al, 2019) as well as the patchwork blanket of power station plumes shown by Junkermann et al (2016). All these studies point towards a significant contribution from local emission hotspots. Chemical analysis from 20 years of particle research at Hyytiälä reveals that sulphur molecules and likely ammonia are among the key substances required for the production of nanoparticle clusters. A recent publication by Hao et al (2018) about measurements at Hyytiälä even requires particulate sulphate in the residual layer mixed downward to explain the observations on the ground.

The sources of such sulfate particles in the atmosphere are well known and typically linked to burning processes (Bigg and Turvey, 1978, Ayers et al, 1979, Whitby et al, 1978). In Finland these sources are mostly located along the coastline, about 200 km or approximately 5-6 hours upwind of Hyytiälä, (Fig. 1, www.endcoal.org). Further examples for primary nanoparticle size, aging and emission rates are shown in the papers of Junkermann et al.

Transport via the residual layer is not an exclusive pattern, veering plumes from wind direction changes in the planetary boundary layer can explain as well the observations without additional aerosols in the residual layer (examples: O'Dowd et al, 2009, for Hyytiälä under conditions with snow covered ground or Laaksonen et al, 2005 (SPC, Italy) Junkermann and Hacker (2018)). In all cases 3D-meteorology is the key for analysis of these observations.

Final comments:

Recent and historic literature is not always taken into account. There are not many airborne studies of nanoparticles, but they should be included.

Time within the manuscript is mixed between UTC and Eastern European Summer Time (EEST) in figures and text.

Fig. 5: Scales for Theta and water vapor are missing. The figure is not really supportive, it suggests a high mixed layer at night although the upper rim of the Lidar data reflect only the vertical range of the measurements. Significant TKE for vertical mixing is restricted only to daylight hours.

Line 166, please take into account: a few hours upwind of Hyytiälä one of Finland's largest pollution source $\sim$150 kg sulphur dioxide / his located, emitting both a large amount of primary particles and a mixture of substances relevant to nanoparticle formation independent on the time of the day.

Fig. 9 should be discussed in terms of the annual variability of meteorology, for example the intensity of convection under typical weather conditions in Finland. The intensity of the vertical mixing process described in the manuscript is dependent on surface conditions (snow until the end of March?) and surface and vertical profile temperatures throughout the year.

Fig. 10 is outdated and needs severe revision. Sources are not always on the ground they can be elevated as well.

Platis et al 2015 should be Platis et al, 2016

Junkermann and Hacker (2018) is cited in the text but missing in the reference list.

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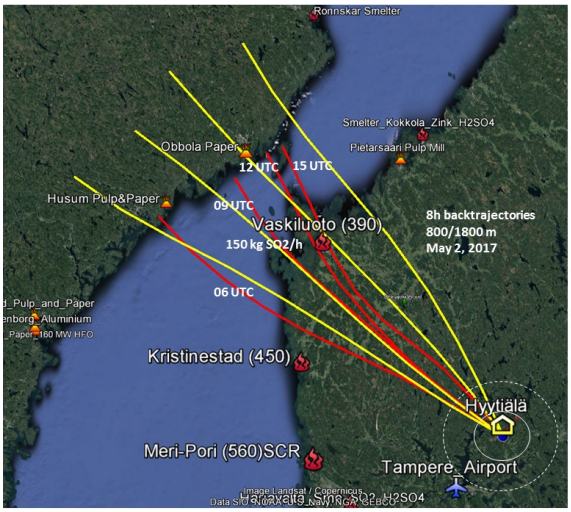


**Fig. 1.** Fig. 1 HYSPLIT analysis (GDAS 0.5) 8 h backtrajectories for Hyytiälä at 06, 09, 12, 15 UTC, at 800 (red) and 1800 m (yellow) and upwind pollution sources