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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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We thank the Referee for the comments. Please see our answers below.

Comment: A number of previous studies, i.e., Nilsson et al. (2001), Stratmann et al. (2003), Stanier et al. (2004); (Wehner et al., 2007), and (Platis et al., 2016) suggested that enhanced turbulent mixing, related to the growth of daytime convective boundary layer and the lift of the inversion could cause downward mixing of the particles, which had already grown in size. In addition, there have been several recent studies that point out direct evidence for NPF occurring aloft, in the interface between the shallow convection and inversion (Chen et al., 2018; Größ et al., 2018). By using turbulence statistics and the boundary layer dynamics (Meskhidze et al., 2019) and (Zimmerman

et al., 2020) quantified the frequency of the residual layer and the ground level nucleation events and assessed their contributions (relative to other sources) to the near-surface fine particle number budgets during different seasons. The authors don't seem to acknowledge many of these studies. That leaves the impression that the residual layer nucleation and the particle entrainment into the mixed layer is a novel mechanism for explaining the appearance of >10 nm-sized particles at the near-surface layer. I would encourage the authors to clearly discuss how their research builds upon these prior studies and highlight the similarities.

Answer: In order to put this study into context we added the following background to the Introduction:

"NPF has been observed in various environments and at various altitudes inside the troposphere. The majority of NPF observations come from ground-based measurements (Kerminen et al., 2018; Kulmala et al., 2004), which can be argued to represent NPF within the mixed layer (ML). Measurements from aircrafts show that NPF is also common in the upper free troposphere (FT) (e.g. Clarke and Kapustin, 2002; Takegawa et al., 2014). Entrainment of particles formed in the upper FT was identified as an important source of CCN in the tropical boundary layer (BL) (Wang et al., 2016; Williamson et al., 2019). Measurements from high-altitude research stations also demonstrate that NPF frequently takes place in the FT, in these cases NPF was often observed in BL air that was transported to the higher altitudes (Bianchi et al., 2016; Boulon et al., 2011; Rose et al., 2017; Venzac et al., 2008).

When studying the vertical distribution of NPF in the lower troposphere one has to consider the evolution and dynamics of the BL. Nilsson et al. (2001) found that the onset of turbulent mixing correlated better with the onset of NPF at ground level than with the increase in solar radiation. The authors gave several hypotheses to why this might be. One hypothesis was that NPF starts aloft, either in the RL or in the inversion capping the shallow morning ML. As the turbulent mixing starts, the newly formed particles would be transported down and observed at the ground-level.

Many observations have supported the hypothesis put forward by Nilsson et al. (2001). Größ et al. (2018), Meskhidze et al. (2019) and Stanier et al. (2004) reported positive correlation between the onset of NPF at ground level and the breakup of the morning inversion due to beginning of convective mixing. Chen et al. (2018), Platis et al. (2015) and Siebert et al. (2004) used in situ airborne measurements and observed that NPF started during the morning on the top of a shallow ML capped by a temperature inversion at a few hundred meters above ground. The particles grew to detectable nucleation mode (sub-25 nm) sizes aloft, and when the ML began to grow due to thermally-driven convection, the particles were mixed downwards and observed at the ground-level where they further continued to grow in size. Stratmann et al. (2003) observed newly formed particles inside the RL disconnected from the shallow ML or the inversion that capped it. Furthermore, Wehner et al. (2010) observed that NPF inside the RL was connected to turbulent layers. On the other hand, Junkermann and Hacker (2018) attributed their observations of elevated ultrafine particle layers at few hundred meter altitudes in the RL to flue gas emissions from stacks with subsequent chemistry taking place during air mass transport over long distances.

The hypothesis proposed by Nilsson et al. (2001) was based on observations done in Hyytiälä, Finland, which is a rural site surrounded by boreal forests and with very clean air. However, the supporting evidence comes from measurements done in more polluted environments in Central Europe and USA. Airborne measurements done over Hyytiälä have not found NPF on top of the shallow morning ML or within the bulk of the RL, instead the NPF events seem to start within the ML (Boy et al., 2004; Laakso et al., 2007; O'Dowd et al., 2009). This might be because in the more polluted environments the RL and/or the shallow ML contains high enough concentrations of precursor vapors from anthropogenic sources, so that NPF can be initiated in the morning inversion and/or within the bulk of the RL. Interestingly, though, observations from Hyytiälä using a small instrumented airplane have frequently found nucleation mode particle layers above the ML at a much higher altitude range of ∼1500-2800 m above ground and the explanation for these layers is not clear (Leino et al., 2019; Schobesberger et al.,

2013; Väänänen et al., 2016). For example Väänänen et al. (2016) found that for the 2013-2014 airborne measurement campaigns 16/36 (∼44%) profiles showed a sub-25 nm particle layer above the ML at altitudes greater than 1800 m asl.

In this study we used co-located airborne and ground-based measurements to study nanoparticles over a boreal forest in Hyytiälä, Finland. We aimed to characterize the elevated nucleation mode particle layers that were a frequent observation in the previous studies. Specifically we were looking at the following questions: (1) where in terms of atmospheric layers, how often and why do these aerosol particle layers occur, and (2) how they are related to ground-based observations, and what implications this has for data interpretation."

Comment: The airplane flight profiles seem to be different between Fig. 3 and Fig. 4. Are these two different profiles? If so, please explain.

Answer: There was a mistake in the time range given in the Fig. 3 caption. The correct time range is 12:00-13:12. Furthermore we combined the May 2, 2017 case study figures into a single figure (Fig. 1).

Comment: Fig. 4 shows that the negative flux was measured at the surface starting at 9:30 am. However, according to Fig. 3, there was no significant vertical gradient between the surface and the 1000 m. Please explain the presence of strongly negative fluxes between 9:30 am and 12:30 pm. According to Fig. 4, a new 10 nm particle mode only appeared at the ground-level at âĹij12:35 pm. So, what causes negative fluxes in the morning?

Answer: The previous correction to the time range should remove the confusion here. In addition we added some text about the particle mode and the negative flux in the morning:

"At the ground level a new particle mode with lower number concentration coupled with negative particle flux also appeared at around 10:00. It may be that these particles were also mixed down from higher altitudes, but in the absence of airplane measurements during that time, we cannot be sure."

Comment: Please include several more case studies so the reader can compare the similarities and contrast the differences. For each case study please show the normalized spectral density plots so the reader can ascertain that there was indeed a growth event following the appearance of >10 nm-sized particles at the near-surface layer.

Answer: While a particle layer was observed on multiple flights, it is rare to find cases where one can directly observe a particle layer mixing down from the airplane and link the ground-based observations to the airborne observations. Ideally the BL development should also be clear in the lidar and the soundings so that comparison can be made to the aerosol observations. We added one more case study (May 19, 2018) to the paper. The case is analyzed in the below text and Fig. 2:

"3.2 Case study: May 19, 2018

On May 19, 2018 another case of nucleation mode particles mixing down into the ML was observed. Figure 4A shows that during the airplane's ascend the lower edge of the particle layer was observed at ∼1200 m asl and the top of the layer was at 2000 m asl. The N3-10 increased in the layer from ∼1000 cm-3 up to ∼10000 cm-3. When the airplane descended back into the ML the N3-10 was increased to around 6000 cm-3 throughout the ML, suggesting that the particle layer was mixed into the ML. The air masses arrived from a similar sector as in the May 2, 2017 case. SO2 and CO concentrations in Hyytiälä remained low when the particles were mixed down (0.05 ppb and 127 ppb for SO2 and CO, respectively).

Figure 4B shows particle number size distribution measurements from the measurement airplane and from the field station. The particle layer was observed as increased number concentration in the smallest size channels of the SMPS at 9:00 before the airplane flew above the ML. Roughly 20 minutes later a similar-sized particle mode appeared in the ground-based data. For this day there were no particle flux data. The

new particle mode continued to grow larger inside the ML for several hours.

Figure 4C shows the TKE dissipation rate on May 18-19, 2018 from Hyytiälä and temperature soundings from Jokioinen. On May 18, 2018 the ML went up to 2500 m asl in Hyytiälä. The Jokioinen soundings show that at 6:00 the top of the RL was at about 1800 m asl, marked by the subsiding inversion left from the previous day's ML. The particle layer mixed down from approximately 2000 m asl."

Comment: Please include the flux values for each of the 8 cases shown in Fig. 8. Since the DMPS was running at the ground site, it would be interesting to know the detected start and the end time of the events, as well as the growth rate for different size particles.

Answer: We added a table that summarizes the cases and includes the negative particle flux peak values (picture of the table in Fig. 3). Regarding the growth rates we added the following sentence to the text:

"The mean growth rate of the appearing particle modes was 2.2 nm h-1 which is similar to 2.5 nm h-1 reported by Nieminen et al. (2014) for 3-25 nm particles during NPF events in Hyytiälä."

Comment: Fig. 8 shows 6-hour differences between the times when the mixed layer reaches the top of the residual layer. Please provide an explanation based on the full analysis of the meteorological data.

Answer: We added the following paragraph to the end of section 3.5 in order to explain these differences:

"The time that the ML reaches the upper RL depends on the height of the RL, which in turn depends on the height of the ML on the previous day and the rate at which the top of the RL subsides. The mixing time also depends on the rate at which the ML on the day of interest grows. For example on March 28, 2014 the ML height on the previous day and the RL height during the night were 1300 m and 1100 m, respectively. On

April 4, 2014 the corresponding numbers were 2800 m and 2200 m. Because of this on March 28, 2014 the ML reached the upper RL much earlier at ∼7:00 compared to April 4, 2014 when the ML reached the upper RL at ∼11:00. For example on April 15, 2014 the ML grew slowly in the morning due to presence of low clouds that limited thermal convection. Because of this the ML reached the top of the RL relatively late at 13:00"

Comment: Please compare the monthly fractions of new particle formation events (Fig. 9) in Hyytiälä with the data reported in other studies discussed above.

Answer: We added the following paragraphs comparing the studies:

"The monthly distribution of upper RL NPF events follows the distribution of ML NPF events, with a peak during spring (Mar-May). This is in line with previous studies that classified NPF events in Hyytiälä (Dal Maso et al., 2005; Nieminen et al., 2014). This makes sense since the conditions favoring ML NPF would also favor upper RL NPF. However, Buenrostro Mazon et al. (2009) and Dada et al (2018) found that the tail events and transported events had a peak during the summer months (Jun-Aug).

On 16% of the NPF event days NPF only took place in the upper RL but not in the ML. This number is smaller than the 36% found by Dada et al. (2018) for transported events and the 26% found by Buenrostro Mazon et al. (2009) for tail events. This might be because we restricted to cases where a negative peak in particle flux was associated with the appearance of nucleation mode particles. For example, a case where the particles were horizontally advected to the measurement site would not be expected to cause a negative peak in the particle flux and therefore would not be classified as upper RL NPF."

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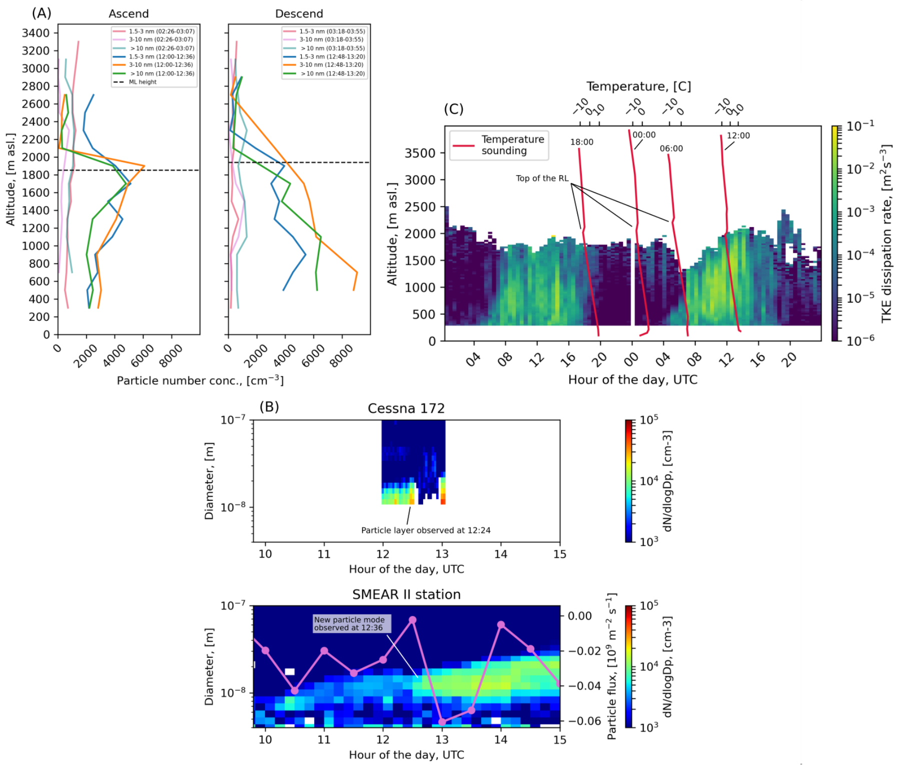
(A)
Ascend
Descend
Altitude, [m asl.]
Particle number conc., [cm^{-3}]
(C)
Temperature, [C]
Temperature sounding
Top of the RL
18:00
00:00
06:00
12:00
Altitude, [m asl.]
Hour of the day, UTC
TKE dissipation rate, [m^{2}s^{-3}]
(B)
Cessna 172
Diameter, [m]
Particle layer observed at 12:24
Hour of the day, UTC
dN/dlogDp, [cm-3]
SMEAR II station
New particle mode observed at 12:36
Particle flux, [10^9 m^{-2} s^{-1}]

Fig. 1.

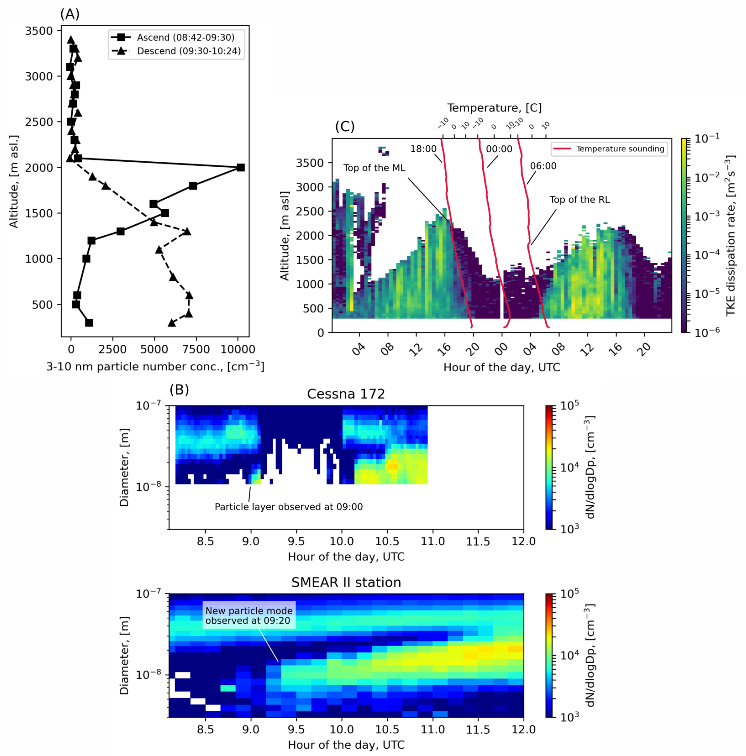
(A)
Ascend (08:42-09:30)
Descend (09:30-10:24)
Altitude, [m asl.]
3-10 nm particle number conc., [cm^{-3}]
(C)
Temperature, [C]
Temperature sounding
18:00
00:00
06:00
Top of the ML
Top of the RL
Altitude, [m asl]
Hour of the day, UTC
TKE dissipation rate, [m^{2}s^{-3}]
(B)
Cessna 172
Diameter, [m]
Particle layer observed at 09:00
Hour of the day, UTC
dN/dlogDp, [cm^{-3}]
SMEAR II station
New particle mode observed at 09:20

Fig. 2.

Table 1: rl_h = residual layer height during night or early morning (m asl), rl_ht = time when the rl_h was observed (time when the sounding was released, hour of the day, UTC), mode_t = nucleation mode particle mode first appears (hour of the day, UTC), mode_t1/mode_t2 = nucleation mode particle mode appearance confidence interval (hour of the day, UTC), rl_t = new mixed layer reaches the top of the residual layer (hour of the day, UTC), rl_t1/rl_t2 = new mixed layer reaches the top of the residual layer confidence interval (hour of the day, UTC), bl_h = observed maximum height of the previous day's boundary layer (m asl.), dp = mean mode diameter for the newly appeared particle mode, when they first appear (nm), gr = growth rate calculated for the newly appeared partice mode (nm h^{-1}), pf = the value of the negative particle flux peak (10^9 m^{-2} s^{-1}).

date	rl_ht	rl_h	mode_t1	mode_t	mode_t2	rl_t1	rl_t	rl_t2	dp	bl_h	pf	gr
20140328	5.3	1100	8.5	9	9.5	5.5	7	8	20	1300	-0.25	2.28
20140331	7.6	2400	14	14.5	15	12	13.5	14	10	2200	-0.06	2.1
20140404	8.5	2200	10.5	11	11.5	10.5	11	11.5	8	2800	-0.04	1.39
20140409	5.5	1500	9	9.25	9.5	6	6.5	7	8	1800	-0.13	1.18
20140415	5.3	1600	14.5	14.25	15	12	13	14	11	1700	-0.18	1.94
20140422	0.0	1800	12	12.5	13	10.5	11	11.5	17	1900	-0.17	1.0
20140518	0.0	1500	9.5	10	10.5	8	8.5	9	13	1900	-0.11	2.91
20140705	5.3	1500	11	11.5	12	8.5	9	10	12	1700	-0.1	4.83

Fig. 3.