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EFFECT OF BOUNDARY LAYER DYNAMICS ON ATMOSPHERIC NEW PARTICLE FORMATION

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Academic dissertation

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Janne Lampilahti
Helsinki, March 2022

Janne Lari Petteri Lampilahti
University of Helsinki, 2022

Abstract

In the atmosphere new aerosol particles can be formed from low-volatility gases in a process called new particle formation. These particles can impact air quality and also climate through the interaction with clouds. The precursor gases are usually emitted from the surface to the boundary layer. Various mixing and transport processes take place in the boundary layer affecting whether new particle formation occurs or not and at what intensity. However, these effects are not well-understood and observations from the atmosphere are scarce.

In this work we studied the relationship between boundary layer dynamics and new particle formation by conducting and analyzing airborne measurements of aerosol particles and meteorology. Our measurements were done in a boreal forest environment in Hyytiälä, Finland using an instrumented Cessna 172 aircraft. A Zeppelin airship also measured in Hyytiälä and in Po Valley, Italy.

In Hyytiälä we found that sub-3 nm particles and clusters decrease in number concentration the higher up one goes inside the mixed layer. This indicates that precursor gases are emitted by the forest, while turbulent convection is transporting the emissions to higher altitudes. This could mean that new particle formation events tend to start close to the forest canopy. From the Zeppelin we observed that a new particle formation event started within the mixed layer.

We found that roll vortices (a common type of organized convection) can induce long and narrow zones of new particle formation within the mixed layer. This is likely because roll vortices can effectively transport precursors from the surface to the favorable low temperature conditions at higher altitudes. We also found that new particle formation frequently takes place at an elevated altitude decoupled from the surface in the topmost part of the residual layer. The mixing of residual layer and free troposphere air appears to be a key trigger for new particle formation in this layer.

In Po Valley we observed that new particle formation started close to the surface after the mixed layer began to increase in height. The particles did not form in the residual layer and then mix down, rather different precursor gases were likely present in the residual layer (sulfuric acid) and in the surface layer (e.g. ammonia) and the mixing of these two layers started nucleation within the shallow mixed layer.

Our results show that boundary layer dynamics plays an important role in new particle formation and these effects should be considered in new particle formation studies. Further work is needed to quantify and parameterize these effects.

Keywords: boundary layer, new particle formation, airborne measurements

Contents

List of publications

This thesis consists of an introductory review, followed by four research articles. In the introductory part, these papers are cited according to their roman numerals.

Paper I. Leino, K., Lampilahti, J., Poutanen, P., Väänänen, R., Manninen, A., Buenrostro Mazon, S., Dada, L., Franck, A., Wimmer, D., Aalto, P. P., Ahonen, L. R., Enroth, J., Kangasluoma, J., Keronen, P., Korhonen, F., Laakso, H., Matilainen, T., Siivola, E., Manninen, H. E., Lehtipalo, K., Kerminen, V.-M., Petäjä, T., & Kulmala, M. (2019). Vertical profiles of sub-3 nm particles over the boreal forest. *Atmospheric Chemistry and Physics*, *19*(6), 4127–4138. https://doi.org/10.5194/acp-19-4127-2019

Paper II. Lampilahti, J., Manninen, H. E., Nieminen, T., Mirme, S., Ehn, M., Pullinen, I., Leino, K., Schobesberger, S., Kangasluoma, J., Kontkanen, J., Järvinen, E., Väänänen, R., Yli-Juuti, T., Krejci, R., Lehtipalo, K., Levula, J., Mirme, A., Decesari, S., Tillmann, R., Worsnop, D. R., Rohrer, F., Kiendler-Scharr, A., Petäjä, T., Kerminen, V.-M., Mentel, T. F., & Kulmala, M. (2021). Zeppelin-led study on the onset of new particle formation in the planetary boundary layer. *Atmospheric Chemistry and Physics*, *21*(16), 12649–12663. https://doi.org/10.5194/acp-21-12649-2021

Paper III. Lampilahti, J., Manninen, H. E., Leino, K., Väänänen, R., Manninen, A., Buenrostro Mazon, S., Nieminen, T., Leskinen, M., Enroth, J., Bister, M., Zilitinkevich, S., Kangasluoma, J., Järvinen, H., Kerminen, V.-M., Petäjä, T., & Kulmala, M. (2020). Roll vortices induce new particle formation bursts in the planetary boundary layer. *Atmospheric Chemistry and Physics*, *20*(20), 11841–11854. https://doi.org/10.5194/acp-20-11841-2020

Paper IV. Lampilahti, J., Leino, K., Manninen, A., Poutanen, P., Franck, A., Peltola, M., Hietala, P., Beck, L., Dada, L., Quélever, L., Öhrnberg, R., Zhou, Y., Ekblom, M., Vakkari, V., Zilitinkevich, S., Kerminen, V.-M., Petäjä, T., & Kulmala, M. (2021). Aerosol particle formation in the upper residual layer. *Atmospheric Chemistry and Physics*, *21*(10), 7901–7915. https://doi.org/10.5194/acp-21-7901-2021

1 Introduction

Fine particles suspended in gas are called aerosol particles. In the atmosphere aerosol particles differ greatly in terms of their properties such as size, composition and concentration (e.g. Seinfeld & Pandis, 2006). Various processes change these properties of the particles. One such process is new particle formation (NPF) in which nanometer-sized clusters form by nucleation from low-volatility trace gases and subsequently, over days, grow to diameters of $\sim$100 nm (e.g. Kulmala et al., 2013). NPF is a large source of aerosol particles in the atmosphere (e.g. Dunne et al., 2016; Gordon et al., 2017). This is important because aerosol particles can influence earth's radiative balance through direct scattering and as cloud condensation nuclei by affecting the formation and properties of clouds. However quantifying the effect of aerosol particles on climate is difficult due to the complexity of the processes involved (IPCC, 2013).

NPF involves the formation of clusters through collisions of gaseous precursor molecules. Once the clusters reach a critical size or stable enough composition they can spontaneously grow larger. A competing process with the growth of the clusters is their coagulation to the larger pre-existing aerosol particles (e.g. Kulmala et al., 2014). In the atmosphere NPF involves multiple components. The precursor gases and formation mechanisms have been extensively studied but are still not fully understood (e.g. Lee et al., 2019; Zhang et al., 2012).

Sulfuric acid is the most important low-volatile compound involved in atmospheric nucleation (e.g. Petäjä et al., 2009; Sipilä et al., 2010). However additional compounds that stabilize the clusters are needed to account for the observations. Such compounds include bases like ammonia (e.g. Benson et al., 2009; Korhonen et al., 1999) and amines (e.g. Almeida et al., 2013; Jen et al., 2014; Petäjä et al., 2011), as well as oxidized organics (e.g. Schobesberger, Junninen, et al., 2013) and ions (e.g. Kirkby et al., 2011). In addition to stabilizing clusters, oxidized organics also contribute to the growth of the particles (e.g. Ehn et al., 2014; Tröstl et al., 2016). In coastal environments iodine oxides are important precursors for NPF (e.g. He et al., 2021; O'Dowd et al., 2002; Sipilä et al., 2016). Sunlight drives the oxidation and production of many precursor vapors which means that NPF mostly takes place during daytime. However, NPF with limited particle growth is observed during nighttime as well (Buenrostro Mazon et al., 2016; Rose et al., 2018).

Many of the processes related to NPF take place in the planetary boundary layer (BL).

The BL occupies the lowest ∼1-2 km of the atmosphere and it is characterized by its strong coupling to the surface. This means that changes in surface properties are seen in the BL in less than an hour or so (e.g. Stull, 1988). During daytime energy from the sun heats the surface, which drives convection and turbulence creating a mixed layer (ML). The ML is capped by a stable layer also known as the entrainment zone because this is where the air from above is mixed down or entrained into the ML. After sunset the ML settles into a less turbulent residual layer (RL). The so-called free troposphere (FT) lays above the BL and is much more decoupled from the surface. Figure 1 illustrates the typical daily evolution of the BL.

The occurrence of NPF depends on the emissions and concentrations of precursor gases but it is also closely linked to the large-scale meteorological situation (e.g. Nilsson, Paatero, et al., 2001) as well as BL evolution and dynamics (e.g. Nilsson, Rannik, et al., 2001; Wu et al., 2021). Generally NPF occurs in air masses that are clean (reduced sink for precursor gases and clusters) and have low cloudiness (allows photo-chemistry to drive the production of precursor gases) (e.g. Dada et al., 2017). For example in a Finnish boreal forest these air masses usually originate from the north west (Sogacheva et al., 2008; Tunved et al., 2006). Such air masses are also characterized by deeper MLs, which can favor NPF (Nilsson, Rannik, et al., 2001). This is because convective mixing starting at the surface after sunrise causes dilution via entrainment of cleaner air from aloft, reducing the sink from pre-existing aerosol particles.

The formation rate of aerosol particles increases when the temperature decreases due to reduced volatility of the precursor gases (Simon et al., 2020; Stolzenburg et al., 2018). Vertical transport in the BL can expose the precursor gases to lower temperatures and initiate NPF. The temperature dependence of the particle formation rate is highly nonlinear, and calculating the formation rate based on mean conditions is likely underestimating the actual particle production in the BL since in reality the temperature fluctuates due to mixing processes (Anttila et al., 2004; Easter & Peters, 1994). The different layers in the lower atmosphere can have different chemical compositions and the higher in altitude one goes the lower the temperature gets. NPF might not be observed at the surface but for example still be taking place in the RL (e.g. Stratmann et al., 2003; Wehner et al., 2010). Also mixing of adjacent atmospheric layers could trigger NPF (e.g. Khosrawi & Konopka, 2003; Nilsson & Kulmala, 1998).

Atmospheric NPF is generally a regional phenomenon spanning tens to hundreds of kilometers horizontally. This is evidenced by the fact that the particle growth can be observed for hours from a stationary point while the airmass is being advected (e.g. Hussein et al., 2009). Such growing particle modes in particle number size distributions are colloquially called "bananas". The number concentration of the growing particle mode usually fluctuates considerably. One reason is because the sources of the precursor gases can be localized. For example in coastal areas iodine compounds released from exposed algae during low tide can induce NPF plumes (e.g. O'Dowd et al., 2002). Also in an urban environment with emissions from traffic and industry the sources are highly local. However even in a homogeneous "background" environment like a boreal forest the growing particle mode usually has discontinuities and fluctuations in the number concentration, which points to the important role of BL meteorology.

At many measurement sites the onset of NPF coincides better with the onset of convective mixing than with the increase in solar radiation. The particles are likely formed and then mixed down from right above the shallow ML (e.g. Größ et al., 2018; Kontkanen et al., 2016; Meskhidze et al., 2019; Nilsson, Rannik, et al., 2001). This is supported by airborne measurements that have found sub-10 nm particles at the top of a shallow ML (e.g. Chen et al., 2018; Platis et al., 2015; Siebert et al., 2004). Such small particles are usually the result of recent NPF when no pollution sources are nearby. Although, some argue that the particles are released from elevated pollution sources and remain trapped in the RL (Junkermann & Hacker, 2018). In any case these particle layers appear to be a feature of more urban environments. In a clean boreal forest environment where the RL is less polluted similar particle layers have not been found at the top of the shallow morning ML, instead the NPF events appear to start within the ML or maybe even close to the surface (Boy et al., 2004; Laakso et al., 2007). In the boreal forest a common observation are sub-10 nm particle layers above the ML at a couple thousand meters above ground (Väänänen et al., 2016). Horizontal variation in the intensity of NPF appears to be common in any environment. In the boreal forest proposed explanations include surface variability due to lakes, variable cloudiness and BL dynamics (O'Dowd et al., 2009; Schobesberger, Väänänen, et al., 2013).

In this thesis we carried out and analyzed airborne measurements of aerosol particles and meteorology in order to study how NPF is related to BL dynamics. The specific topics and research questions are summarized below and in Figure 1.

1. Where does the onset of NPF take place in the BL and what is its relationship to precursor sources and BL dynamics? Also what are the differences between a

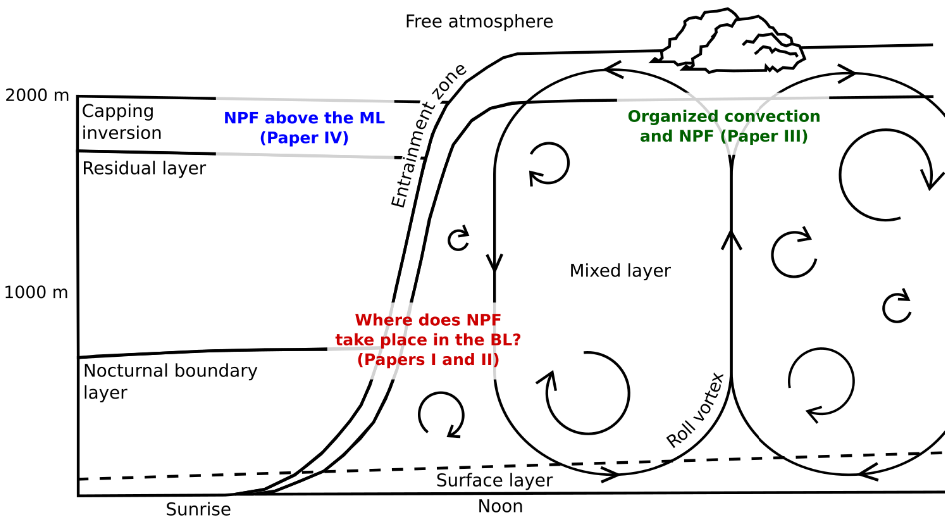

Figure 1: A schematic figure illustrating a typical diurnal cycle of the planetary boundary layer. The different research topics in this thesis are also presented in the figure.

clean boreal forest environment and a more polluted urban background environment? (Papers I and II)

2. What is the role of organized convection at creating spatial variability in NPF? (Paper III)

3. Is there NPF taking place above the ML and what is its connection to BL dynamics? (Paper IV)

2 Measurements

2.1 Airborne measurements

2.1.1 Cessna 172

Most of the airborne measurements in this thesis were done on board a Cessna 172 airplane equipped with meteorological and aerosol instruments. The Cessna campaigns

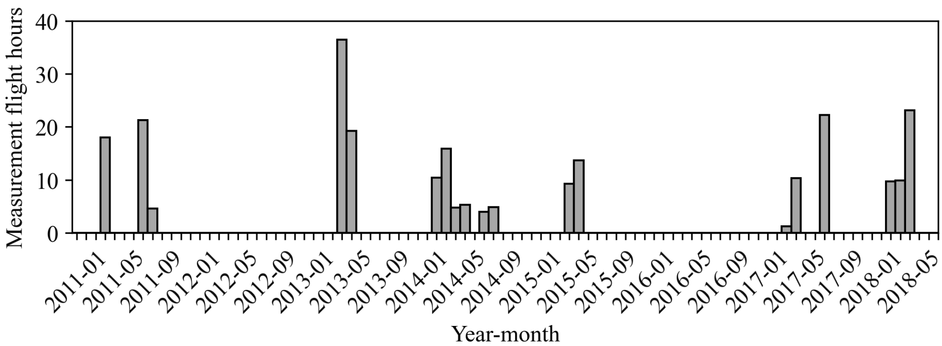

Figure 2: Monthly flight hours in the Cessna campaigns between 2011-2018.

have been organized almost every year since 2009 (Schobesberger, Väänänen, et al., 2013; Väänänen et al., 2016, Papers I-IV). The campaigns used in this thesis are summarized in Figure 2.

In the measurement setup the sample air was collected through a forward facing inlet nozzle (McNaughton et al., 2007) into a main sampling line that was routed to the inside of the airplane's cabin. A flow rate of 45-50 lpm was maintained by the movement of the aircraft and a manual valve operated by a researcher on board. The instruments located in the cabin sampled the air from the center of the main sampling line. Until 2014 the inlet was installed under the starboard wing but in 2015, in order to minimize losses, the main sampling line was shortened and routed through the port side window (Figure 3).

The instruments inside the Cessna's cabin were mounted into a rack and they were powered using 12 V lead-acid batteries. The rack and the batteries were situated behind the two front seats. One of the instruments in the rack was a scanning mobility particle sizer (SMPS; Wang and Flagan, 1990). The SMPS used a bipolar charger to bring the sample aerosol into charge equilibrium, a short Hauke type differential mobility analyzer selected different sized particles based on their electrical mobility and a TSI 3010 condensation particle counter (CPC) then counted the particles. A CPC works by condensing vapor (usually butanol) onto the surface of the particles growing them to $\sim$1 μm size, and then counting them using optics. One measurement cycle lasted 4 minutes and two flow rate modes were used to get a particle number size distribution between 10-400 nm. A TSI 3776 CPC and a particle size magnifier (PSM; Vanhanen

Figure 3: Inlet during the 2018 Cessna campaign.

et al., 2011, Lehtipalo et al., 2022) measured the number concentration of >3 nm and >1.5 nm particles respectively. In the PSM diethylene glycol is first used to grow the particles before detection with a conventional butanol-based CPC (in our case TSI 3010). This two-stage method allows smaller particles to be detected. Meteorological sensors measuring temperature and relative humidity were mounted under the wing. Pressure sensor was inside the unpressurized cabin. A turbulence probe (AIMMS-20, Aventech Research Inc.) was mounted outside the starboard wheel in 2015 to measure the wind vector at 20 Hz.

The measurement profiles extended from approximately 100 m to 3000 m above ground, which is enough to measure the ML, RL and the lowest parts of the FT. The profiles were flown perpendicular to the mean wind direction in order to avoid exhaust fumes. The flying speed relative to the surrounding air was 36 m s^{-1}. The ascent and descent rates during a vertical profile were usually around 0.8 m s^{-1}. The measurement flights lasted 2-3 h and were flown mostly during the morning and afternoon local time.

2.1.2 Zeppelin NT

In Paper II we studied the measurements done during the PEGASOS project on board a Zeppelin NT airship (Figure 4). In June 2012 the Zeppelin was measuring in Po Valley, Italy and in May-June 2013 in Hyytiälä, Finland.

The data was analyzed only from flights that had on board a specific set of instruments that was designed to measure atmospheric NPF. An atmospheric pressure interface

Figure 4: The instrumented Zeppelin measuring in Finland (image credit: Riikka Väänänen).

time-of-flight mass spectrometer (APi-TOF; Junninen et al., 2010) measured the elemental composition of ambient air ions and their clusters. From the APi-TOF data one can deduce the compounds that form the clusters in the initial steps of NPF. A neutral cluster and air ion spectrometer (NAIS; Manninen et al., 2016; Mirme and Mirme, 2013) measured the particle (2-40 nm) and ion (0.8-40 nm) number size distributions. Also an SMPS, a PSM and meteorological sensors were on board the Zeppelin.

During a typical measurement flight the Zeppelin did multiple vertical profiles over a small area (~10 km^2) downwind from the ground-based measurement. The altitude range during measurement profiles was 100-1000 m above ground and the air speed was ~20 ms^{-1}.

2.2 Ground-based measurements

The airborne measurements in Finland (Papers I-IV) were flown around the SMEAR II station in Hyytiälä (23.5E, 61.85N, 181 m; Hari and Kulmala, 2005). Corresponding aerosol data was measured at the SMEAR II station and we used it to complement the airborne observations. The environment around Hyytiälä is mainly Scots pine forests with some small lakes, agricultural fields and dispersed settlements. Closest large city is Tampere 60 km south west from Hyytiälä.

In 2014 high spectral resolution lidar measurements and 4-hourly balloon soundings were done in Hyytiälä in the context of the BAECC campaign (Petäjä et al., 2016). In paper IV this dataset allowed us to study the BL structure and evolution in combination with the aerosol data. We also extensively used the vertical flux of particles larger than 10 nm, measured at 23 m above the ground (Buzorius et al., 2000). In paper III we utilized the Finnish meteorological institute's weather radar in Ikaalinen (60 km west from Hyytiälä) to detect organized convection over the Cessna's flight track based on signals from airborne insects.

In Paper II the ground-based measurements in Po Valley, Italy, were done in San Pietro Capofiume. Po Valley is considered a pollution hotspot. The measurement site is situated in a large agricultural area and therefore can be considered to measure the background conditions in Po Valley (Hamed et al., 2007).

3 Results and Discussion

Atmospheric NPF is influenced by many processes including emissions of precursor gases, atmospheric chemistry, aerosol dynamics and boundary layer dynamics (e.g. Lee et al., 2019). Some of these processes are illustrated in Figure 5. The main source of sulfuric acid in the atmosphere is the oxidation of sulfur dioxide. Sulfur dioxide is released to the atmosphere during for example coal burning and volcanic eruptions. In the marine boundary layer the oxidation of dimethyl sulfide produced by plankton is a source of sulfuric acid (e.g. Rosati et al., 2021). Ammonia and amines are emitted from various sources including agriculture, vegetation, oceans, biomass burning and combustion (e.g. You et al., 2014). The organic vapors participating in NPF are the oxidation products of anthropogenic or biogenic volatile organic compounds (VOCs). For example the boreal forest in Hyytiälä is a major source of biogenic VOCs (e.g. Hakola et al., 2003). Iodine compounds are important precursors in coastal areas and they are released for example from exposed algae during low tide (e.g. Sipilä et al., 2016). The iodine compounds from marine and coastal areas can be transported large distances over land and possibly participate in NPF far from the coast (Beck et al., 2021).

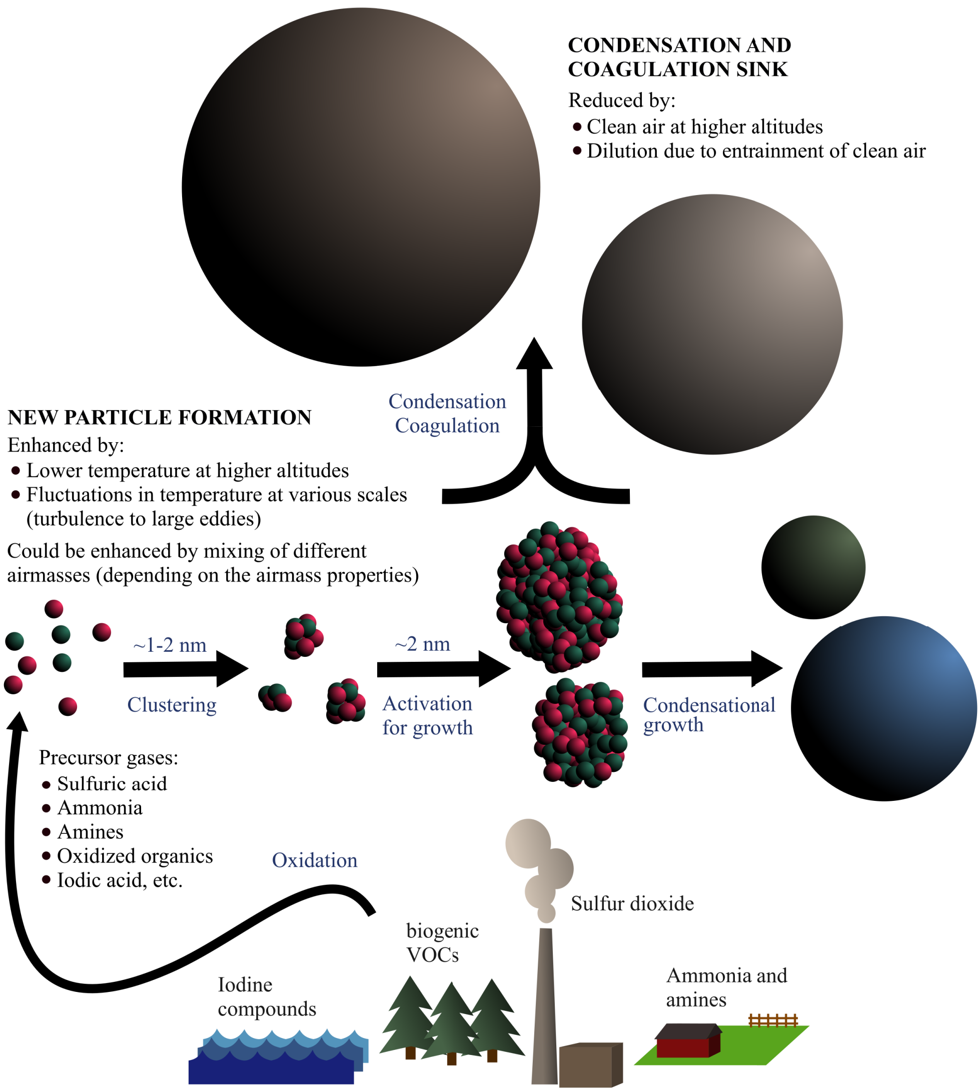

Figure 5: Schematic diagram of different processes involved in atmospheric NPF.

3.1 The onset of NPF and its relationship to precursor sources and BL dynamics

The first steps of NPF take place below 3 nm. In order to directly study NPF one needs to measure the particles in this size range (Kulmala et al., 2012). In Paper I we used the PSM and the TSI 3776 CPC on board the Cessna to measure the number concentration of particles larger than 1.5 nm and 3 nm respectively. Subtracting the latter number concentration from the former gave the number concentration in the size range 1.5-3 nm. In addition the increase in 2-4 nm ion number concentration is a sensitive indicator of NPF (Leino et al., 2016) and we used it in Paper II to capture the onset of NPF on board the Zeppelin. The 2-4 nm ion concentration was measured by the NAIS.

The main finding from Paper I was that on NPF event days (17 profiles) the 1.5-3 nm particles showed on average a steadily decreasing number concentration from 100 m ($\sim$3000 cm^{-3}) to 2700 m ($\sim$300 cm^{-3}) above ground. The median number concentration of sub-3 nm particles at 100 m was comparable to the median of 2900 cm^{-3} reported by Kontkanen et al., 2017 at the station during spring. A case study (13 Aug 2015) showed that the number concentration reached its minimum at the top of the ML and did not show notable increase at higher altitudes in the RL and the FT. The results make sense since the boreal forest is a major source of organic compounds that contribute to NPF (e.g. Ehn et al., 2014). Turbulent mixing coupled with coagulation losses would result in a vertical profile where the number concentration decreases from the surface to the top of the ML. Particles in the 3-10 nm size range had a more constant average vertical profile, probably because they had more time to get mixed. In the Hyytiälä case study of Paper II we also found that the NPF event started within the ML and there was no NPF in the RL.

These results are in agreement with previous findings from Hyytiälä. O'Dowd et al., 2009 measured 3-6 nm and 6-10 nm particles over the forest regions of southern Finland during three NPF events. The authors concluded that particles in the smallest size range were first detected close to the canopy and as the particles grew in size they mixed more uniformly into the ML. Laakso et al., 2007 used a hot-air balloon and an air ion spectrometer that measured charged particles down to 0.8 nm to study NPF over Hyytiälä. The authors concluded that NPF started within the ML, possibly close to the surface.

In Paper II we also studied the onset of NPF in Po Valley, which is a more polluted environment compared to Hyytiälä. We observed that the NPF event started inside the shallow ML roughly at the moment when the ML began to grow in height. In a polluted environment high levels of sulfur dioxide may end up in the RL and oxidize into sulfuric acid. In Po Valley we observed higher levels of sulfuric acid in the RL on board the Zeppelin. In Hyytiälä the sulfuric acid levels remained low in the RL. Also there was a sudden increase in sulfur dioxide concentration at the surface as the ML began to grow, probably due to entrainment from the RL. During the night sulfur dioxide concentration becomes low close to the surface due to deposition. We found 2-4 nm ions in the RL indicating NPF, but these particles did not grow to larger sizes and they were not responsible for the NPF event in the ML. It appeared that the conditions in the RL were not suitable for particle growth beyond ~3 nm. In Po Valley ammonia concentration increases in the morning at the surface likely due to agricultural activities (Sullivan et al., 2016). When the convective mixing begins sulfuric acid rich RL air is mixed down into the ammonia rich ML air. Ammonia can stabilize the sub-3 nm clusters allowing the particles to grow larger, which results in an NPF event (e.g. Kürten et al., 2016).

Our results from Po Valley are similar with previous results from urban environments. For example Kontkanen et al., 2016 and Meskhidze et al., 2019 found that the onset of convective mixing and the entrainment of RL air closely coincided with the onset of NPF. Airborne measurements have found NPF on top of the shallow morning ML. The newly formed particles were entrained down when vertical mixing started (e.g. Chen et al., 2018; Platis et al., 2015; Siebert et al., 2004; Stratmann et al., 2003). However in our case the NPF event did not start above the ML, rather it seems the mixing of precursors from the RL with the precursors in the ML was the crucial step that initiated NPF within the ML.

3.2 The role of organized convection at generating horizontal variability in NPF

While on average the sub-3 nm particles had an increasing number concentration towards the surface in Hyytiälä, there was still substantial horizontal variation in the intensity of NPF within the ML (Väänänen et al., 2016). Particularly interesting features of horizontal variability were long (>10 km), narrow (1-5 km) and ML deep zones

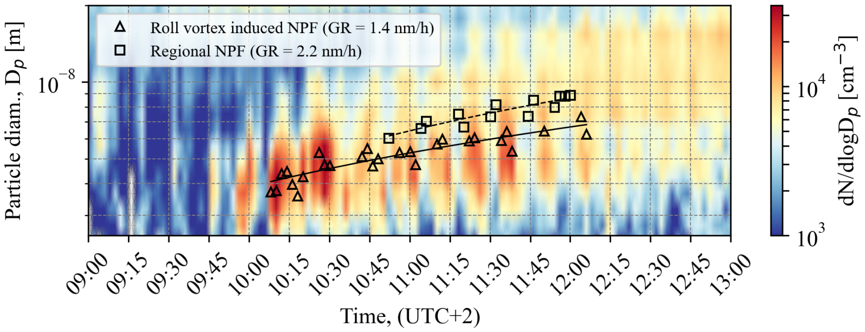

Figure 6: Particle number size distribution measured on board the Zeppelin by the NAIS on May 8, 2013. For roughly two hours between 10:00-12:00 the Zeppelin periodically flew through a zone where roll vortices were enhancing a regional NPF event creating a striped "tiger banana". This allowed us to calculate the growth rate (GR) of the particles in the enhanced NPF zone. The triangles and the squares mark the fitted (using log-normal distribution) average mode diameters.

of sub-10 nm particles. These zones were approximately aligned with the mean wind direction. The particle number concentration in such zones could increase by an order of magnitude compared to the surrounding air. Air mass back trajectories showed that the particle zones did not come from any specific source area and their locations on the measurement flight area were more or less random. The Zeppelin allowed us to measure the particle growth rate in such a zone (Figure 6). Combining the particle growth rate with back trajectories suggested that the particles were formed by a line source. This description of the particle zones fits surprisingly well with the description of roll vortices, which are a type of organized convection. More specifically roll vortices are narrow and long helical circulations that extend the depth of the ML (e.g. Etling & Brown, 1993, Figure 7). In Paper III we investigated if roll vortices were inducing the elongated particle zones.

We identified the particle zones from the airborne data and found that there was a significant association with roll vortices ($p < 0.03$). The analysis included the Cessna campaigns between 2013-2015. A nearby weather radar allowed us to check if roll vortices were present over the measurement area during a flight. We also found that the particle zones observed on board the Cessna were associated with concentrated "stripes" in the ground-based particle number size distribution. These stripes would

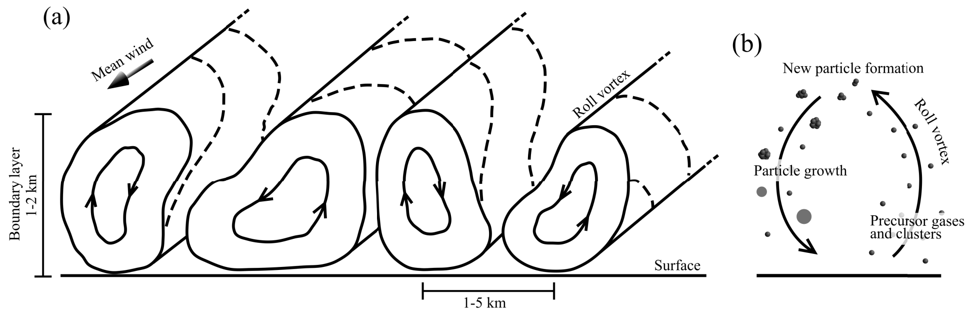

Figure 7: (a) Shows a schematic drawing of roll vortices and (b) shows a schematic drawing of roll vortex induced NPF.

last ∼1 hour, occur in the sub-20 nm sizes and show a simultaneous down-up fluctuation in vertical particle flux (given that there were enough particles above 10 nm in diameter, detected by the flux system's CPC). The particle stripes would often occur at the same time with regional NPF events and have similar particle size. The stripes were caused by the particle zones drifting over the measurement station. We used this ground-based signal to estimate statistics on roll induced NPF outside the flight campaigns and to calculate median growth rates, formation rates and spatial coverage. The frequency of roll induced NPF was estimated from the airborne campaigns. Combining the estimates we calculated that the roll induced NPF increased the production of 3 nm particles by 61±39 % and 10 nm particles by 24±7 % compared to only homogeneous regional NPF.

During one of the flights using the wind vector measurements on board the Cessna we were able to directly link a long and narrow particle zone to adjacent roll vortices. The highest number concentration appeared to be in the downdraft, suggesting that the particle formation rate was highest in the top parts of the ML.

Past studies suggest that organized convection and roll vortices could have a role in atmospheric NPF. Easter and Peters, 1994 modeled nucleation in a sulfuric acid-water system in the context of different atmospheric mixing processes such as large eddies. One of the conclusions was that nucleation could be enhanced in updrafts where adiabatic cooling lowers the temperature. Buzorius et al., 2001 noticed that in Hyytiälä NPF events often took place when roll vortices were present in the ML. The authors speculated that the vertical transport of precursors from the surface to the top of the ML could be driving NPF.

The boreal forest is a source of NPF precursors and Paper I shows how on average the number concentration of sub-3 nm particles and clusters decreases towards the top of the ML. Paper III provides evidence that organized convection, and especially roll vortices, can transport these precursors and clusters aloft and initiate or enhance NPF at higher altitudes. This results in horizontal variability in the intensity of NPF, either as isolated zones of NPF or as enhancement of regional NPF along the roll vortices.

3.3 Elevated layers of NPF

It was discussed in Section 3.1 how in Hyytiälä NPF events seem to start within the ML. However sub-10 nm particle layers above the ML, at some thousands of meters above the ground, are often observed on board the Cessna suggesting that NPF is also taking place above the ML (Schobesberger, Väänänen, et al., 2013; Väänänen et al., 2016). Previous observations of elevated NPF layers are from more polluted environments where the particles are usually forming on top of a shallow morning ML at some hundreds of meters above the ground (e.g. Chen et al., 2018; Platis et al., 2015; Siebert et al., 2004) or from a marine BL where NPF seems to be taking place in a layer on top of cloud topped ML (e.g. Dadashazar et al., 2018). Also NPF layers have been observed in the top parts of the free troposphere in the outflow from deep convective clouds (e.g. Clarke & Kapustin, 2002; Krejci et al., 2003; Williamson et al., 2019). The particle layers observed on board the Cessna do not seem to fit any of these observations. In Paper IV we investigated the particle layers further.

Using the Cessna's SMPS data we studied the average vertical profile of particle number size distribution during NPF event days over Hyytiälä in 2011-2018. We found that during the morning between 9:00-12:00 local time a distinct sub-20 nm particle layer was present at roughly 2700 m above sea level while the ML height was approximately 1000 m. The ML was capped by a temperature inversion, but another temperature inversion, most likely capping the RL, was exactly where the particle layer was found at 2700 m. The air masses to the elevated particle layers arrived mostly from north west of Hyytiälä, which is a very clean sector and is associated with NPF (Tunved et al., 2006).

We also looked at data collected during the BAECC measurement campaign in 2014, which included balloon soundings, lidar profiling and airborne as well as ground-based aerosol measurements in Hyytiälä (Petäjä et al., 2016). We looked for cases where a new growing sub-25 nm particle mode that lacked the growth from smallest detectable sizes appeared in the number size distribution at the surface. Here we call such an event ”banana tail” (Buenrostro Mazon et al., 2009). We also required that the beginning of the particle mode was associated with a downward peak in particle flux, which would suggest that the particles were mixing downwards. The banana tails are different from the particle stripes caused by the roll vortex induced NPF since in a banana tail the particle size would grow for several hours and show no abrupt ending.

Then we checked if we could determine the top of the RL from the balloon soundings and Cessna flights by looking for a temperature inversion that was left from the previous day's ML. Then from the lidar's backscatter signal we determined the approximate time when the ML reached the top of the RL. We compared this time with the time the new sub-25 nm particle mode appeared. We found that the times correlated well ($R = 0.93$, Figure 8). The aerosol particles that mixed down grew for several hours suggesting that they covered a large horizontal area, which is a characteristic of NPF events.

The observations suggest that the aerosol particles were formed in the upper parts of the RL. The interface between the RL and the FT is a mixing place for air masses with different properties. Precursors can be transported to the upper RL either from the surface by convection during the previous day or as a result of long-range transport in the FT, or both. The mixing in the interface layer likely initiates NPF either by creating favorable thermodynamic conditions in the temperature inversion for NPF to occur (Khosrawi & Konopka, 2003; Nilsson & Kulmala, 1998) or by mixing different precursors from the RL and the FT to initiate NPF. The aerosol particles can be entrained to the surface where they are observed as a growing sub-25 nm particle mode that is missing the initial growth because the particles already grew past the smallest sizes above the ML.

To find out how common NPF is at the top of the RL in Hyytiälä we collected statistics on the banana tail events with downward peak in particle flux from 2013-2017. Based on this we estimated that ~42 % of the NPF events observed in Hyytiälä appear to start on top of the RL.

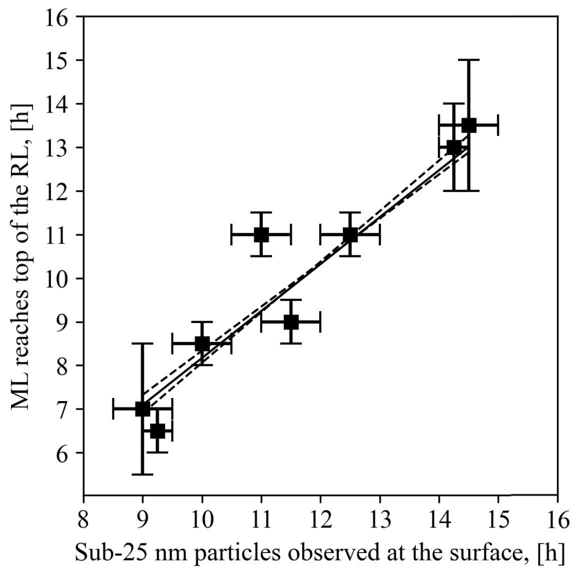

Figure 8: Correlation between the ML reaching the top of the RL and the appearance of a growing sub-25 nm particle mode that lacks the initial particle growth at the surface.

4 Review of papers and the author's contribution

Paper I. We studied the spatial distribution of sub-3 nm particles and clusters in the lower troposphere over a boreal forest using the instrumented Cessna. I participated in the Cessna measurements and the data analysis.

Paper II. We studied the onset of NPF and its relationship to BL dynamics using an instrumented Zeppelin airship. Measurements were done in Po Valley, Italy and Hyytiälä, Finland. I analyzed most of the data and wrote the paper.

Paper III. We investigated the reasons for a particular pattern of horizontal variability in NPF that was observed on board the Cessna and the Zeppelin. We found that a type of organized convection called roll vortices was responsible for generating the variability. I participated in most of the Cessna measurements, analyzed the data, and wrote the paper.

Paper IV. We analyzed the elevated sub-10 nm aerosol particle layers observed on board the Cessna. We found that the layers were likely due to NPF in the interface between the RL and the FT. I participated in most of the Cessna measurements, analyzed the data, and wrote the paper.

5 Conclusions

In this thesis we conducted airborne measurements of aerosol particles and meteorology in order to study how the dynamics of the BL affects the formation of new aerosol particles. The airborne measurements were done using an instrumented Cessna 172 airplane and a Zeppelin NT airship. The measurements were performed in Hyytiälä, Finland and in Po Valley, Italy. In both locations ground-based measurements were utilized to complement the airborne data. Below is a summary of our findings.

1. We measured that in Hyytiälä sub-3 nm particles and clusters increased in number concentration towards the surface, indicating that the NPF events start close to the forest (Paper I). The NPF event observed in Hyytiälä on board the Zeppelin started inside the ML and there were no signs of NPF in the early morning RL or on top of the shallow ML (Paper II). In Po Valley the NPF event started inside

the ML when convective mixing began. This is likely because sulfuric acid from the RL mixed with ammonia (amines, organics) in the ML creating conditions for NPF to occur (Paper II).

2. Measurements on board the Cessna and the Zeppelin in Hyytiälä showed that roll vortices (a type of organized convection) could induce or enhance NPF along the rolls (Paper III). This created horizontal variation in the intensity of NPF over the boreal forest. The likely cause is that roll vortices effectively lift precursors and clusters from the canopy to the top of the ML where temperature is lower and particle formation rate is increased.

3. Data from multiple Cessna campaigns as well as ground-based data suggest that the interface between the RL and the FT is a hot spot for NPF above Hyytiälä (Paper IV). Aerosol particles formed in this elevated layer can be entrained into the ML and observed at the surface as growing particles that lack the initial growth from smallest detectable size.

This thesis shows how BL dynamics can have important effects on the occurrence and intensity of NPF. The importance of these effects for example on the overall production of cloud condensation nuclei in the atmosphere remains unclear though. Therefore more work needs to be done in order to accurately quantify and parameterize these effects. This requires more observations and statistics for example on roll vortex induced NPF and on NPF in the RL-FT interface.

A valuable future improvement would be to develop a better method to detect roll vortex induced NPF. One approach could be based on particle fluxes (Buzorius et al., 2001) but also one could perform drone observations with lightweight CPCs coupled with a wind measurement (e.g. Brus et al., 2021).

Next step in analyzing the elevated NPF layers could be a more comprehensive back trajectory analysis on the so-called banana tail events in Hyytiälä. Such analysis might be able to separate banana tails that occur due to a change in advected airmass from banana tails that are mixed down from aloft. This could also provide information on the origin of the RL and FT air masses, which could elucidate why aerosol particles are forming in the interface between the two layers.

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