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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Roll vortices induce new particle formation bursts in the planetary boundary layer" *by* Janne Lampilahti et al.

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We thank the referee for the constructive comments, please find our responses below.

General Comments

This study presents evidence from field data of the formation of aerosol particles from volatile organic compounds (New Particle Formation, NPF) due to the transport of boreal forest air to the upper regions of the atmospheric boundary layer by the convective boundary layer rolls. This is a relevant topic that deserves to be studied and understood, since it can have direct impact on the estimation and modeling of aerosols in the atmosphere, which are relevant for air quality, weather and climate. This study presents a dataset that shows clear evidence of the relationship between convective rolls and

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Discussion paper

NPF. However, the manuscript needs some improvement in terms of the scientific writing. Due to its relevance, I suggest (1) improvements to the scientific presentation of the study, and (2) some additional analysis and discussion that can help future studies on the development of better measurements and models for this phenomenon.

Introduction: it is too short and some important information is lacking. For ex- ample, it needs more details on what is NPF (how it is defined, range of particle sizes of interest, where it comes from), why it is important (where it is used, where it is not used but should be used) and what are the mechanisms in which ABL dynamics might influence NPF. It would be important to describe in details what is already known about the relationship between NPF and convective rolls, what is not known (or never observed in field data), and what will be investigated here exactly. Why convective rolls, but not convective conditions in general? With this information the reader should be convinced about the relevance of this study. Right now this description (and consequently the motivation) of the study is su- perficial, only someone in the field will recognize its importance. It is important to convince the general audience as well. Some interesting information that should be in the intro is mentioned in the Conclusion section and in the caption of Figure 10.

Answer: we extended the first paragraph in the introduction to give more details on NPF. We moved the explanation on how roll vortices could induce NPF above the boreal forest from the conclusions to the introduction along with Figure 10. We made the the rest of the introduction mode detailed by following to the comments below.

Methods: the section already starts with “Zeppelin measurements”, without introducing the reader with the big picture of the methods of the study. It would be useful to start with a overall description (type of data, location, overall goal with each type of data, etc). After situating the reader, then go to the details. All the details needed to reproduce the analysis should be given. Some information is described in the results section (or in the caption of figures), some is missing (see details below). I’m very confused about the different particle size ranges mentioned in different moments of the manuscript. It

there a range of interest?

Answer: we added in the beginning a paragraph giving an overview of the measurements. In the detailed sections we took the below comments into account and rephrased or added text. In the conditions for roll-induced NPF we were looking at sub-20 nm particles, since this data was readily available from all measurement platforms.

Results: these results are very interesting, but they are too focused on the measurements of particles, but not on the atmospheric conditions. Maybe the gas and meteorological data at the surface could be used to provide quantitative information about the roll-induced NPF? It would be interesting to characterize the roll days with their micrometeorological variables, and to try to better identify the differences between the days with and without NPF. If this is not possible, it should be addressed in the manuscript, with a discussion of what should be done in future field studies in order to provide better quantitative data that can be used to model this phenomenon.

Answer: we agree that the analysis could be expanded. For example developing more comprehensive methods to measure the phenomenon and studying the cluster composition during roll-induced NPF. However we find this further analysis is beyond the scope of this study.

We added to the conclusions: “In order to fully understand roll-induced NPF, better measurement and analysis methods need to be developed. For example measuring the fluxes of sub-10 nm particles and doing airborne flux measurements. More measurements with a turbulence probe on board need to be performed. It would also be interesting to study the cluster composition during roll-induced NPF.”

Specific Comments

l. 25: “the small clusters and particles originating from these bursts grow in size similar to particles typically ascribed to regional scale atmospheric NPF”. The difference

between regional scale NPF and rolls induced NPF should be made clearer.

Answer: we rephrased the text to show the difference between roll-induced and regional scale NPF more clearly:

"the small clusters and particles originating from these localized bursts grow in size similar to particles typically ascribed to atmospheric NPF that occurs almost homogeneously at a regional scale."

l. 40: "In observational studies enhanced nucleation mode particle concentrations have been observed in turbulent layers in the lower atmosphere. For example inside the residual layer (Wehner et al., 2010) and in the inversion capping a shallow mixed layer (Platis et al., 2015; Siebert et al., 2004)." It is not clear how these two layers would favor the development of NPF, compared to other ABL conditions.

Answer: we added text explaining why BL dynamics can be important for NPF:

"Numerical studies have shown that fluctuations in ambient temperature and relative humidity, caused by for example small-scale turbulence, large eddies such as roll vortices (Easter and Peters, 1994), or mixing over a temperature inversion (Nilsson and Kulmala, 1998) can lead to significant enhancements in new particle formation rate compared to only mean conditions. This is because the formation rate has a nonlinear dependence on temperature and the gas-phase concentrations of the precursor vapors. Therefore, fluctuations in these variables, as opposed to mean conditions where the fluctuations are averaged out, can have a net enhancing effect on the source strength of aerosol particles by NPF."

Now it should be more clear why turbulence would favor NPF in these layers. We also edited the text a bit:

"In observational studies, increased nucleation mode particle concentrations have been measured in atmospheric layers where turbulent fluctuations were enhanced. For example in turbulent layers inside the residual layer (Wehner et al., 2010) and in

the inversion capping a shallow mixed layer (Platis et al., 2015; Siebert et al., 2004)."

l. 40: what is "nucleation mode particle"?

Answer: we added "sub-25 nm" to the text where we introduce NPF.

l. 43: "Other airborne measurements have found significant horizontal and vertical variability in the number concentration of nucleation mode particles within the BL." Can you expand on that? What level of variability? Anything measured within the ABL has variability, what makes this one worth pursuing?

Answer: we added text about the degree of variation that can be found

"Other airborne measurements have found that during NPF the number concentration of nucleation mode particles shows considerable, up to an order of magnitude, variation within the BL"

l. 49: "Convection in the planetary BL often organizes into counter-rotating horizontal roll vortices or rolls that extend to the top of the boundary layer". What is the horizontal and time scales of these rolls? How can they be identified by micrometeorological variables? This is relevant to evaluate if the measurements are appropriate. Why this specific type of convection is more relevant for NPF than others?

Answer: we added Figure 1 that shows 3d view of roll circulation with labels that explain the scale. The methods to identify roll vortices in the BL are outlined in the methods section. We also moved the explanation and the associated figure of the concept behind roll-induced NPF from the conclusions to the introduction.

l. 55: "and the overall effect of rolls on aerosol particle formation is unknown". Is it completely unknown? Can you be more specific on what is known, what is unknown? You have cited papers that discuss this.

Answer: we rephrased the text to be more specific

"However direct observations of the effects of roll vortices on NPF are lacking."

l. 68: “We used the positively charged particles and the data was averaged to 4 min time resolution”. Why? Is that equivalent to the total concentration of particles?

Answer: we modified the text to read

“We used the total particle data from the positive polarity of the instrument.”

In our case both polarities looked roughly the same in terms of data quality so the choice was more or less arbitrary, but in any case one should perform the analysis on one polarity only so that the data is most comparable.

l. 72: “The data was corrected for diffusional losses in the one meter long, 37 mm inner diameter, inlet tube and converted to standard conditions (293.15 K and 1 atm).” How? Can you provide at least a reference, so that someone could reproduce what was done exactly?

Answer: we added a reference to the diffusion loss calculation

“Gormley, P. G. and Kennedy, M.: Diffusion from a Stream Flowing through a Cylindrical Tube, Proc. R. Ir. Acad. Sect. Math. Phys. Sci., 52, 163–169, 1948.” We also added the equation for the conversion to standard conditions.

Sec 2.1: it is not clear after this section if the zeppelin data is only profiles or if there are measurements fixed at a given height.

Answer: we made the text more specific:

“The zeppelin measurements consisted of consecutive profiles. Each profile was a slow and even ascend (∼25 min) from ∼100 m up to ∼1 km above ground followed by a fast descend (∼5 min) while the speed relative to the surrounding air (airspeed) was kept at ∼20 m/s.”

l. 89: “Particle number concentration in the 3-20 nm range was calculated by subtracting the total particle number concentration measured by the Scanning Mobility Particle Sizer (SMPS) from the number concentration measured by the Ultrafine Condensation

Particle Counter (UCPC)." Not clear what that means. Why are you interested in this range only? The SMPS is mentioned in Table 1 as measuring between 10-400 nm. No information about UCPC is given. This description is not clear.

Answer: we modified the text

"We used the particle number concentration in the 3-20 nm size range as an indication of particles that likely originated from NPF. The 3-20 nm particle number concentration was calculated by subtracting the total particle number concentration measured by the Scanning Mobility Particle Sizer (SMPS) in the size range 20-400 nm from the number concentration measured by the Ultrafine Condensation Particle Counter (UCPC). We skipped the smallest size bins of the SMPS because they were in some cases noisy."

We also added UCPC in the Table 1.

l. 91: "The SMPS starts to lose accuracy in terms of spatial distribution of the aerosol particles due to its 2 min averaging period when the horizontal scale becomes less than 4 km." How does that apply to your study? Is this scale comparable to the phenomenon that you are investigating? Is this relevant? What about the other instruments used?

Answer: we decided to leave this part out because of the following reasons:

During the roll-induced NPF observations the number concentration from the UCPC was elevated during a large part of at least one SMPS scan. In these cases the SMPS total number concentration did not increase at all (the particles were below the detection limit) or the number concentration was momentarily (one or more SMPS scans) increased in the smallest size bins (10-20 nm) of the SMPS. An example is presented in Figure 2 where purple arrows show the times when the airplane flew through a roll-induced NPF.

In light of this we would say that the calculated 3-20 nm number concentration was in our cases a reliable indication that the number concentration was increased in the 3-20 size range. Therefore mentioning this limitation here is not relevant and can lead to

confusion.

l. 93: “A turbulence probe, capable of measuring the 3d wind vector, was only installed at the end of the 2015 campaign.” This sentence is completely lost here. What is this going to be used for? And how? Any details on this instrument? Measurement frequency, probe model, post processing?

Answer: we added the following paragraph:

“In order to detect roll vortices on board the airplane we installed a turbulence probe (Aventech Research, AIMMS-20) at the end of the 2015 campaign. The AIMMS-20 was capable of measuring the the 3d wind vector at 20 Hz, but for the analysis we averaged the data to 1 s.”

Sec 2.2: it is also not clear after this section if the airplane data used is only profiles or if there are measurements fixed at a given height.

Answer: we wrote the following:

“Typical measurement tracks consisted of $\sim$30 km long flight segments flown roughly perpendicular to the mean wind direction over the same area while doing a single vertical profile from 100 m to 3000 m above ground. The ascend and descend speeds were on average $\sim$1 m/s.”

l. 122: “The CPC had a 10 nm cutoff size” what is the measurement range? what is CPC?

Answer: the CPC measured all particles above 10 nm. We rephrased the text:

“The system measuring the vertical particle flux used an ultrasonic 3d anemometer combined with a condensation particle counter (CPC) at 23 m above ground. The CPC had a 10-nm cutoff size.”

l. 132: what is Aitken mode?

Answer: we added “Aitken mode (25-100 nm)”

Sec 2.4: Is there an exact quantitative criteria for NPF days, or was it selected by inspection only?

Answer: this is done by inspection, we added this to the text.

l. 147: What is the time interval used? What size ranges are used? How is the coagulation sink obtained? It is important to provide all information from the data to the results presented.

Answer: we added the following:

“We calculated the CoagSd from the DMPS data and for the number concentrations we used the NAIS data, so that the final time resolution of the formation rate was 4 min. The size ranges used from the NAIS data were 3-6 nm for J3 and 10-20 nm for J10.”

Sec. 2.8: I did not see the use of the ABL height in the results section.

Answer: in roll-induced NPF condition (i) we specify that the concentrated longitudinal sub-20 nm particle zone should be inside the BL and this is where we checked the BL height.

l. 193: “Figure 2 shows a frequent observation in the measurement data:” which data?

Answer: rephrased to “Figure 2 shows a frequent observation in the ground-based aerosol particle measurements”

Results section: why is the particle range size different in different analysis (for example figs 4 and 5, or between conditions (i) and (ii))

Answer: in the roll-induced NPF conditions (i) and (ii) we are looking at sub-20 nm particles.

We changed the size range in Figure 4 to be 3-20 nm instead of >1.5 nm in order to be consistent. In Fig. 5 the size range is also 3-20 nm.

In Figure 3 the SMPS stopped working in the middle of the flight, which is why we are only showing data from the UCPC (>3 nm). However, we know from the simultaneous ground-based observations that the observed particles were sub-20 nm.

l. 208-216: this paragraph should be in the Methods section.

Answer: we think it is necessary to explain the case study before defining the two roll-induced NPF conditions. Otherwise it is very difficult for the reader to understand why we define the conditions the way we do.

l. 228-229: which statistic test was performed? All information necessary to reproduce your results should be given.

Answer: the statistical test was Fisher's exact test. It is mentioned in the text.

l. 229-232: can you verify in the data what micrometeorology conditions characterize NPF and non-NPF days?

Answer: NPF events generally occur on sunny days with a lot of atmospheric mixing. Our data does agree with this (see Figure 3). However this figure adds little to understanding roll-induced NPF so we chose to leave it out of the manuscript.

l. 235: "This timescale is associated with mixing throughout the convective BL" did you calculate it? Compared with references?

Answer: we changed this sentence to be more specific to rolls: "This timescale is similar to the period of a typical roll vortex". A reference (Easter and Peters, 1994) was given in the introduction. This time scale is also similar to the mixing throughout the BL since the rolls circulate the air throughout the depth of the BL.

l. 238-244: instead of Table 3, it would be useful to show plots related to the estimation of GR. Also, what is the particle range size of your GR estimate?

Answer: we see that a good example of growing roll-induced NPF particles is already shown in Figure 8 where mean mode diameters are fitted to multiple subsequent roll-

induced NPF observations and over time they show a growth trend. We added fit lines and the resulting GRs to the figure also. For the GR estimate the median lower size was 7.5 nm and the median upper size was 15 nm. We added this information to the text.

Figure 8: “and power spectra of the wind components from the station’s mast showed that the rolls were moving over the site” this would be interesting to see, maybe it could be added to this figure as a third panel?

Answer: the power spectra, along with two other supporting figures showing the movement of the roll-induced NPF and the rolls in the weather radar image, can be seen in our reply to Referee #1 (Fig. 4, explanation of the figure is in the answers). We find that the figure is quite technical and would not add significant extra value.

We made the description in the caption more precise:

“The roll-induced NPF event was moving over the measurement area from southwest to northeast. Weather radar observations showed that rolls were present over the measurement site and power spectra of the wind components from the station’s mast showed that the rolls were moving over the site at the same rate (one roll in $\sim$20 min), and in the same direction as the roll-induced NPF.”

The analysis in Figure 9 is not clear to me. It needs to be better explained in the methods and results section, not only in a figure caption. All the details needed to reproduce your results should be presented.

Answer: we added a detailed explanation to the text, see our answer to the first referee.

Figure 10 is more appropriate for the introduction than conclusion. A good de- scription of the physical process that motivates this study is in the caption of the figure, and it would be important for the reader to know about these things since the beginning.

Answer: we moved the figure to the introduction.

Technical Corrections

l. 40: “In observational studies, enhanced” (add the comma)

Answer: fixed

l. 52: “(Buzorius et al., 2001; Nilsson et al., 2001)” you don’t have to cite the same thing twice on the same sentence.

Answer: fixed

l. 55: “However direct observations (...)”, rephrase.

Answer: we rephrased this to “However direct observations of the effects of roll vortices on NPF are lacking.”

l. 67: what is “mobility diameter”?

Answer: the SMPS, DMPS and NAIS diameters are electrical mobility equivalent diameters. We decided to refer to simply diameters throughout the text to avoid confusion.

l. 79: “while the airspeed was kept at 20 m/s” not clear what that means

Answer: we modified the text to read “the speed relative to the surrounding air (airspeed) was kept at $\sim$20 m/s.”

Table captions: remove the word “Explanations:”

Answer: fixed

l. 85: Table 1 also mentions the Zepelin data, why is it mentioned only in the Airplane section?

Answer: we moved the sentence to the overview in the beginning of the methods section.

l. 96: “such that the aircraft was either descending, ascending or staying level”, maybe rephrase as “measurements performed during descending, ascending...”

Answer: rephrased

“Typical measurement tracks consisted of ∼30 km long flight segments flown roughly perpendicular to the mean wind direction over the same area while doing a single vertical profile from 100 m to 3000 m above ground (Figure 3). The ascend and descend speeds were ∼1 m/s.”

l. 98: “The measurement airspeed was 36 m/s”, again, not clear.

Answer: the airspeed is now explained in the Zeppelin section, so this should be clear.

It goes from section 2.4 to section 2.8

Answer: fixed

l. 143: why no equation number?

Answer: equation number added

l. 221: “station. Whereas” change to comma

Answer: changed

I don’t think Table 2 is necessary, the statistics are sufficient.

Answer: we agree that the detailed information presented is not necessary, so we removed the table and the references to it from the text.

l. 225: “roll-inducued”

Answer: fixed

Table 3: as Table 2, I don’t think this is necessary. It should be presented the statistics, but the information for each individual day is not necessary for the understanding of the study. If you decide to keep these tables, maybe put them in an appendix or supplemental material.

Answer: we agree and decided to remove Table 3 from the text. The important statistics

(average growth and formation rates) can be found in the text and the important figure derived from Table 3 is Figure 9.

l. 265: equation number

Answer: equation number added

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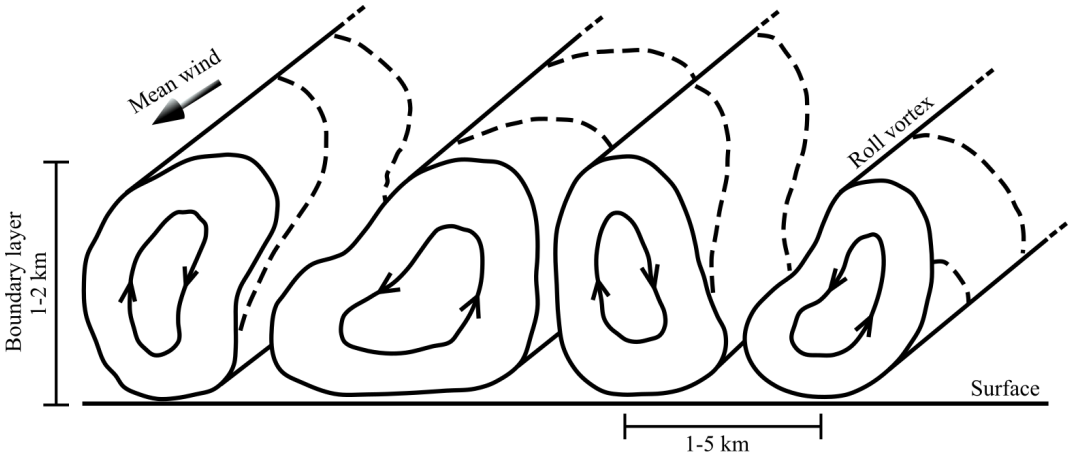

Fig. 1.

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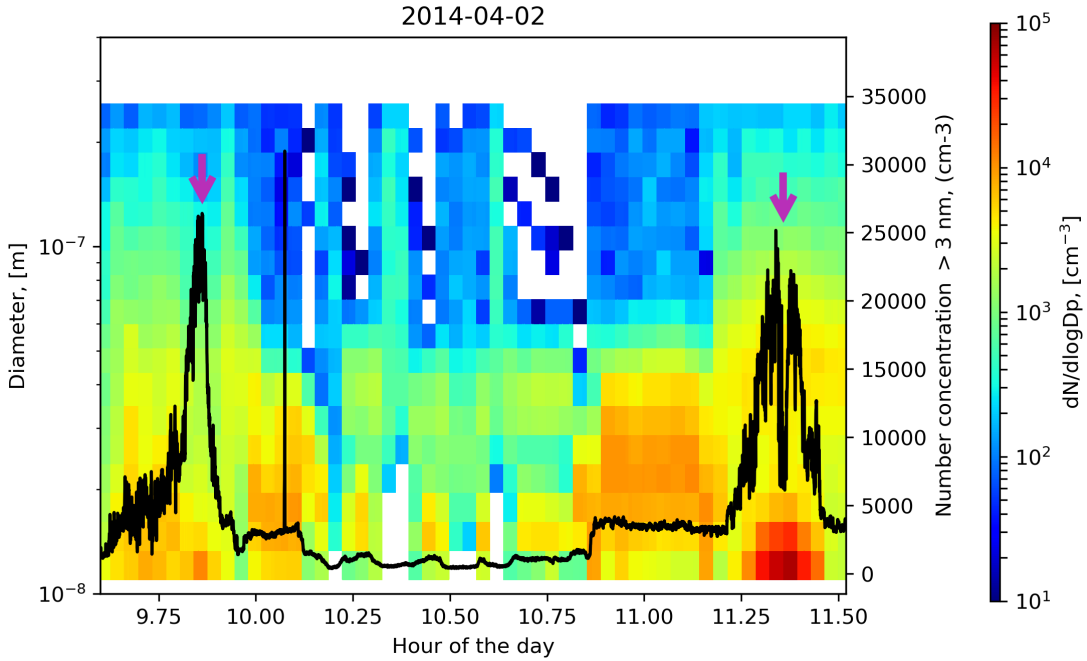

Fig. 2.

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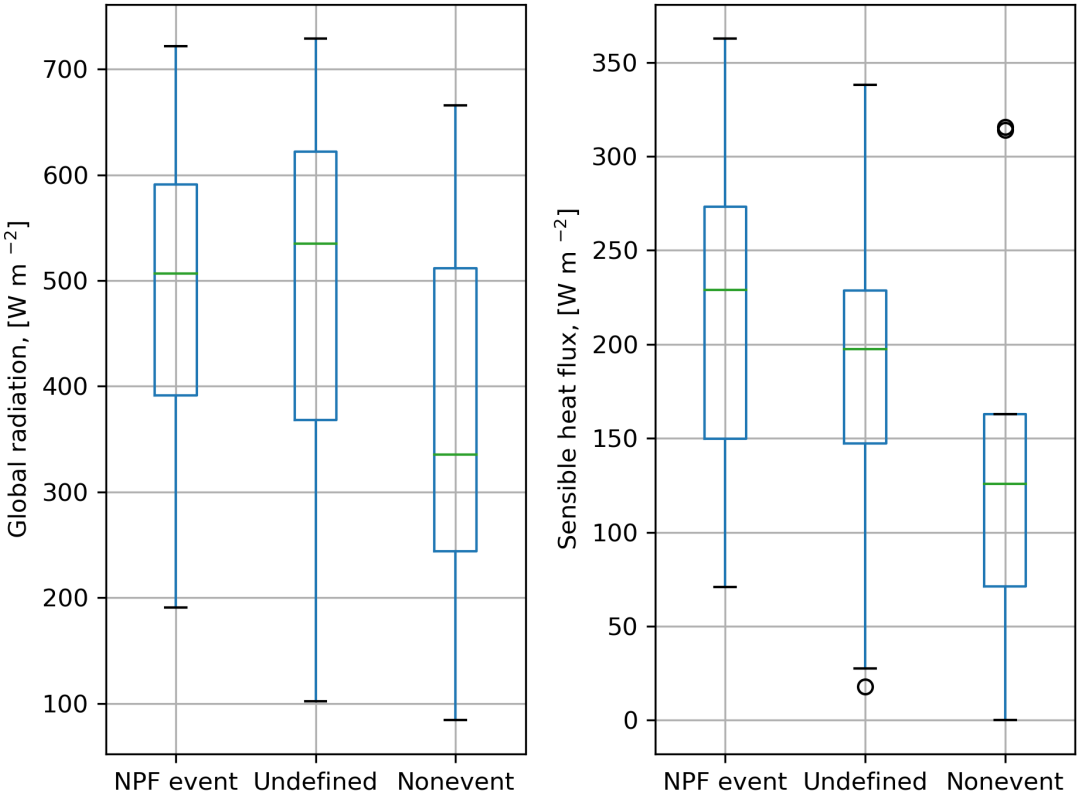

Fig. 3.

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