

Authors' response and marked-up manuscript

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Airborne observations of newly formed boundary layer aerosol particles under cloudy conditions by Altstädter et al.

First of all, the authors would like to thank both referees and Wolfgang Junkermann for their valuable support and constructive criticism that will improve the present form of the manuscript. Just to make clear the choice for the selected days on April 4, 2014 and June 21, 2015: The two days are of special interest in terms of similar weather conditions (strato cumulus clouds) influenced by a passage of low pressure system and low precursor gases (of SO₂), but affected by different wind directions in order to investigate the impact of anthropogenic emissions on new particle formation, as assumed by e.g. Junkermann et al. (2016). During our observations, however, no clear new particle formation event with high and continuous growth rate was observed at ground. Only sporadic clusters were formed during short intervals of cloud gaps. NPF formation between clouds has been recently studied by Wehner et al. (2015), but the study differs from our observations, due to (now) high turbulence on local scale. In our opinion, the study of the goal is now clarified, supported by your contributions. Thank you!

On April 4, the site was influenced by east wind direction, so that anthropogenic emissions due to polluted air masses from Eastern Germany might have been observed that coincide with the increase of NO_x and will confirm the study of Junkermann et al. (2016). At this point, we do not exclude the impact of anthropogenic emissions on NPF, but in our opinion, the sources are local and not from long distances. With the help of the UAS profiles, an increase of N₅₋₁₀ was observed in connection to a condensation sink, shown by OPC data, and enhanced moisture above the inversion layer at around 9 UTC, which spread out even further until noon around 14 UTC.

The overall observation was that the new particle formation event was not captured by the TSMPS at ground, but was proven by the UAS (even in an altered state). Similar sporadic appearances of ultrafine particles were investigated on June 21. During the second measurement day, the profiles were performed as part of the MelCol2015 campaign. Therefore, additional instrumentation was available at the same time. We used data from an eddy-covariance station that confirmed the rapid upward and downward transport of aerosol particles, so that mixing processes, initiated by large turbulent fluxes within the ABL, led to insufficient particle growth rate. Turbulent fluxes were estimated by an LES model that was used for the campaign. Unfortunately, there was no meteorological data available, therefore the ABL was characterised by the LES. In our opinion, the second measurement day is still interesting and unique by the use of various instrumentations that further link to UAS profiles and ground observations.

As the whole set up was presented in previous studies already (Altstädter et al., 2015; Platis et al., 2016 and Bärffuss et al., 2018), the authors decided "just" to briefly introduce the used instrumentation. In our opinion, the aerosol and meteorological sensors are sufficiently described in Section 2.1 (information on manufacturer, accuracy and response time). Nevertheless, we added the following lines on p.4, l. 6-9:

The aircraft design and instrumentation's set up were introduced in Altstädter et al. (2015) and supported by the results of Platis et al. (2016). During review process, the aircraft was re-engineered and we refer to the newest study of Bärffuss et al. (2018) who presented a profound overview of ALADINA's flight operation, data procedure and the current payload that" is equipped with aerosol instrumentation and meteorological sensors with high temporal resolution.

(...) p4., 16-18:

The laboratory results were confirmed by Bärffuss et al. (2018) during field studies in Melpitz. The UAS was placed at the same aerosol inlet as ground monitoring and N7-12 coincide with ground data in the same particle size within the deviations of +/-20% during sampling period of 1.5 h.

Please, find attached the authors' responses and the marked-up manuscript including the changes and remarks corresponding to the referees in different colours (RC1 in green, RC2 in blue, SC1 in yellow), respectively.

In addition, during review process, one of the co-authors has changed the affiliation and some references were revised. Please, take into account the remarks given by the authors at the end of these comments (see 4th point, marked-up in magenta).

Comments of referee #1 (RC1)

"The paper by Altstädter et al. aims at showing evidence for the occurrence of NPF aloft under cloudy conditions and vertical transport of the newly formed particles close to the ground. Observations were performed from the Melpitz ground based station as well as from the unmanned aerial system ALADINA. The combination of these two datasets is of great interest, however the choice of the two case studies included in this work is not fully justified (6 cases available in total), and is even more questionable that instrument failures occurred during both of them. With the exception of some sentences which are unclear, the paper is well written. However, measurements/observations are often under-used, or used to support conclusions which sometimes do not seem to be correct, or inconsistent between the different sections. Also, references are lacking, which makes it difficult to evaluate the significance of this work compared to earlier studies; additional references would also help better understanding some of the tools/methods which are only very briefly introduced. All in all, I would not recommend the publication of the manuscript in its current form. However, because I think the dataset is highly valuable, and even if major revisions are needed, I really encourage the authors to consider submitting a new version of their manuscript. Here are some comments/suggestions to help in this process."

Author's response to referee #1

The authors acknowledge the anonymous referee for the detailed proof reading and for the specific suggestions that will highlight the outcome of the current manuscript. The authors agree that some parts might irritate the reader, as the additional instrumentation was not the same during the two measurement days. Please, take into account the remarks on p. 1 of the authors' response.

In the following, the authors will go through the comments that were given by the first referee.

Please, find attached the point-to-point response and the marked-up manuscript version including the changes (removed is marked in red and new is coloured in green).

Point-to-point response:

RC1: "Comment 1, P1, L12: The use of « closure » is misleading as LES -model was used in the absence of measurements, and not to confirm observations."

Authors' response: The authors agree that the sentence contains a false statement, as the LES model was used in order to derive meteorological data due to missing data by the UAS and not for comparison.

We have reformulated the last sentences of the abstract in order to state clear the outcome of the study on p. 1, l. 16-20:

During the first measurement day, a maximum of $N_{5-10} = 3.5 \times 10^3 \text{ cm}^{-3}$ was detected above an inversion layer in connection with a decline of larger particles and layer of high moisture that existed until noon. However, altered clusters (particle size between 30 and 50 nm) were measured at ground during the process in relation to an increase of NO_x so that anthropogenic emissions might have been favoured the NPF occurrence. Whereas by south-west wind conditions, pronounced turbulent fluxes of sensible and latent heat, taken from an LES model output" (..)

RC1: "Comment 2, P2, L9-12: I assume that the authors refer to the Class III events identified by Gröss et al. (2015) to support the occurrence of NPF under cloudy conditions. I think this reference should be used with caution in this context, as NPF event classification is based on a new approach in the above mentioned study. Thus, I would recommend to at least explicitly mention the use of this alternative method, especially because according to the authors those Class III events are "very weak events with very small-scale particle bursts that do not evolve into a fully developed and spatially distributed nucleation event. In any case, this class of observations includes what most researchers would call "nonevents". Also, in its current form the sentence on L9-12 sounds weird to me; I would either remove the last part, starting from "and these", or replace "is possible" by "might be possible"."

Authors' response: Thank you very much for that hint. The authors agree that the comparison with Class III events presented in Größ et al. (2015) (now: Größ et al (2018)) is irritating. According to the revised version of the manuscript, the authors have adjusted their descriptions. Class III events were classified as "low NPF intensity, including non-events". Similar observations during clouds were measured with NAIS called case 4, Fig. 3d. The literature was removed to Section 3.2, because the authors think that this reference is similar and needs to be addressed (p. 10, l. 12-15):

A similar observation of clustered NPF during cloudy conditions and west wind in Melpitz was shown in the recent study of Größ et al. (2018). Insufficient particle growth rate was significantly connected to fluctuations of hydroxyl radicals and H₂SO₄ (shown in the so called Case 4) that were steadily suppressed by clouds in contrast to high increase of H₂SO₄ during clear sky. The authors suggested local processes and steadily influenced sources as cause for this formation behaviour.

As the authors wanted to stress the hypothesis that nucleation is expected more often than assumed so far in current literature, the authors changed the lines “is possible” into “*might be possible*” (see p.2, l. 16).

RC1: “Comment 3, P2, L19-21: The concept of “breaking waves” should be briefly recalled/clarified. Also, I assume there is a word missing in the last part of the sentence.”

Authors' response: Thank you a lot for the correction. Yes, the sentence is not complete in the present form. Further we believe that the mentioning of breaking waves is in fact not relevant for the scope of the paper and we decided to skip that in the modified version of our manuscript (p. 2, 26-28).

Further, Bigg (1997) suggested that high humidity and temperature fluctuations may enhance new particle formation. Besides, Easter and Peters (1994) (...)

RC1: “Comment 4, P3, L12: The expression “the appearance of nucleation and particle growth” is confusing, and should be replaced by something like “the appearance of nucleation mode particles and their subsequent growth”, as the nucleation process itself is not observed at ground level.”

Authors' response: The authors acknowledge the first referee for the comment. The suggested lines “the appearance of nucleation mode particles and their subsequent growth” were changed into the marked up manuscript on p.3, l.19-20.

RC1: “Comment 5, P3, L16: Regarding the expression “In contrast to typical NPF events at ground by high incoming solar irradiance”. It is in my view too simplistic and a bit confusing, as it suggests that only radiation is driving the occurrence of NPF events observed close to the ground. Some other factors such as the presence of primary precursors (e.g. SO₂) as well as the strength of their sink are also determining the process.”

Authors' response: The authors intend to highlight the observations during cloudy conditions in contrast to the previous results by clear sky conditions, shown in Platis et al. (2016). Indeed, further factors cause new particle formation, as mentioned in previous paragraph (see p. 2, l. 8-10 of the discussion paper). But in this case, the authors stated “typical NPF events at ground by high incoming solar irradiance” as the main difference from previous studies to the current manuscript. In order to avoid any lacking description, the authors reformulated the sentences (p. 3, l. 15-21):

Previous investigations of Platis et al. (2016) showed NPF in correlation with temperature and humidity fluctuations of several orders of magnitudes higher than in the remaining part of the ABL. In addition, downwards transport of freshly formed particles to ground level was observed and supported by the appearance of nucleation mode particles and their subsequent growth relating to an increase of SO₂ concentrations by clear sky.

RC1: “Comment 6, P4, L1-6: Was the effect of pressure on the CPCs detection efficiency accounted for in the data analysis or did you assume it could be neglected over the range of altitudes discussed in the present study?”

Authors' response: The CPC data was not corrected for pressure effect, because for the pressure range of interest, 900 -1025 hPa, the counting efficiency of the CPC changes only minimal, i.e., less than 4% (Heintzenberg et al., 1999), which is still within the range of the overall CPC uncertainty.

RC1: “Comment 7, P4, L27: I would suggest to move the reference to Manninen et al. (2010) to the next paragraph, where other NPF related results are reported, or find a way to “group” all the NPF related observations in section 2.2 in order to clarify the message.”

Authors' response: The authors decided to split the section into four paragraphs in order to obtain a clear structure: 1. Short description of measurement site, 2. air mass characterisation, 3. previous studies of NPF, 4. used instrumentation in this study.

The transition was done by the change on p. 5, l. 9-10; in order to group the following reference of Manninen et al., 2010: The research site Melpitz of TROPOS is well known in NPF studies and benefits from comparison of airborne data with monitoring at ground.

RC1: “Comment 8, P5, L10-12: More explanations should be reported about LIDAR and Ceilometer measurements/data analysis since they provide information on the occurrence of clouds, which are to some extent in the scope of the present work. At least, earlier studies involving these instruments should be mentioned. In particular, the meaning of « range corrected signal » (captions of Fig. 3 and 5) should be explained.”

Authors' response: The authors agree that lidar and ceilometer are not sufficiently introduced or referenced. However, the authors' intention is to **show briefly** the existence of clouds and not to go any deeper into the description of the instrumentation.

The range-corrected signal is the uncalibrated attenuated backscatter coefficient. Attenuated by molecules and particles. As at 1064 nm attenuation is weak, it is a good proxy for the particle backscatter coefficient.

Following lines and references were added (p. 5, l. 25-29):

Clouds and aerosol layers are indicated in the lowermost 3–4 km with backscatter signals of ceilometer (in the wavelength of 1064 nm; e.g. Heese et al., 2010; Wiegner and Geiß, 2012) and lidar (in the wavelength of 532 nm; e.g. Pal et al., 1992; Engelmann et al., 2016). During the first measurement day, ceilometer data is taken into account, whereas during the MelCol 2015 experiment, the PollyXT lidar (e.g. Althausen et al., 2009) was available. The given range corrected signal is the uncalibrated attenuated backscatter coefficient that is damped by molecules and particles, respectively.

- Engelmann, R., Kanitz, T., Baars, H., Heese, B., Althausen, D., Skupin, A., Wandinger, U., Komppula, M., Stachlewska, I. S., Amiridis, V., Marinou, E., Mattis, I., Linn{e}, H., and Ansmann, A.: The automated multiwavelength Raman polarization and water-vapor lidar PollyXT: the neXT generation, *Atmos. Meas. Tech.*, 9, 1767-1784, <https://doi.org/10.5194/amt-9-1767-2016>, 2016.

-Heese, B., Flentje, H., Althausen, D., Ansmann, A., and Frey, S.: Ceilometer lidar comparison: backscatter coefficient retrieval and signal-to-noise ratio determination, *Atmos. Meas. Tech.*, 3, 1763-1770, <https://doi.org/10.5194/amt-3-1763-2010>, 2010.

-Pal, S. R., W. Steinbach, and A. I. Carswell, 1992: Automated method for lidar determination of cloud-base height and vertical extent. *Applied Optics*, 31, 1488–1494

-Wiegner, M. and Geiß, A.: Aerosol profiling with the Jenoptik ceilometer CHM15kx, *Atmos. Meas. Tech.*, 5, 1953-1964, <https://doi.org/10.5194/amt-5-1953-2012>, 2012.

RC1: “Comment 9, P6, about the “Results and discussion section”: It is surprising to have results about the frequency of occurrence of the phenomenon of interest in this study (i.e. occurrence of NPF in so-called non-favourable conditions) reported in the abstract (P1, L6-8) and in the conclusion (P10, L32 – P11 L2) which are not further discussed in the abovementioned section, or not even recalled. I would strongly recommend to introduce and discuss those at the beginning of section 3, before the case studies. In particular, the frequency of this process occurring close to the inversion layer should be for instance compared to that of “clear” banana shape NPF events detected at the ground level. This is needed to assess the importance of the observed phenomenon. Also, the choice of the two cases investigated in the present work is not clear, and should be supported by additional explanations, especially because data were missing from ALADINA during the selected days (CPC data during first case, meteorological parameters during second case). In specific, are those two cases representative of all 6 days when similar phenomenon was observed, or do they all display contrasting conditions?”

Authors' response: Just one comment: At the moment, another study is in preparation that focuses on statistics of ultrafine particles obtained by profiling with ALADINA. However, this is not the scope of the current paper. During our observations, only one “clear” banana shape occurred on July 1, 2015 in relation to high SO₂ concentrations and updraft of ultrafine particles during clear sky. The results were presented at the European Aerosol Conference 2017 in Zurich, already. However, the whole study is still being processed, as it belongs to the MelCol 2015 campaign. The authors decided not to introduce the campaign, as an overview paper is currently in progress and will be published in the near future. Regarding the other points, we would like to refer to the very beginning of the authors' response on p. 1. In our opinion, the two case studies are representative, as the main wind directions were considered. We agree that the high occurrence of NPF phenomena during cloudy conditions should be revised in the results. Therefore, the sentences were added on p. 6, l. 25-28:

To almost one third of the performed flight days with ALADINA, bursts of ultrafine particles were observed under cloudy conditions close to the inversion layer. Two of the six measurement days were selected explicitly due to similar weather conditions, low SO₂ concentrations, but different wind (NE, SW) directions that however mainly influenced the site (e.g. Spindler et al., 2012; Engler et al., 2007).

RC1: “Comment 10, P6, about “Section 3.1, Case I April 2014”: In my view, the data shown in this section do not fully support the conclusions provided on P8, L10-19. In particular, I believe that some of the observations meant to evidence the transport of small particles downwards do not support the occurrence of such phenomenon. More detailed comments regarding this section are provided below.

Starting with ALADINA vertical profiles:

- Detection of small particles close to the inversion layer and/or in the vicinity of clouds has already been observed. Therefore, the authors should refer to earlier studies in order to highlight how their results are similar or differ from observations reported in the literature;

- As mentioned previously, it is complex to really “follow”, and thus validate, the transport of particles downwards because of missing CPC data during 2 of the vertical profiles performed with ALADINA. Looking at the profile from 11:47 on Fig.4, high N5 values are observed in a layer between 100 and 250 m a.g.l., which seems to be disconnected from the layer above 600 m a.g.l. where high N5 values are also detected. The authors suggest that the small particles (N5-10) observed close to the ground at 13:50 were transported from the upper layer (>600 m): how can they exclude the formation of small particles in the lower layer (100-250 m), and further transport of these specific particles close to the ground?

Authors' response: Thank you for the comment. Indeed, the description is misleading. We cannot exclude a second layer with ultrafine particles in the heights between 100 and 250 m. However, we doubt that the layer is mixed from the upper part, as the capping inversion is still existent.

- Also, particles in the range 5-10 nm are observed at ground level with the CPCs onboard ALADINA at 13:50, while particle number size distributions measured at the same time with the SMPS do not show any clear change in this size range compared to the distributions measured earlier during the day. How do the authors explain this? It might be useful to show the values of N5-10 calculated from SMPS measurements, in a similar way as done in Fig. 9 and 10. The analysis of fluxes such as those discussed in the second case study might also give further insight into the processes observed from ALADINA;

Authors' response: As it was discussed in previous parts, during the two measurement days, different instrumentation were available at the TROPOS research site. We inserted one comment for the reader in order to avoid any confusion of the different instrumentation in Section 3.2. (p. 9, l. 17-18)

ALADINA flights were performed as part of the MelCol 2015 experiment so that additional instrumentation was available at site.

- Last, have the authors evaluated the threshold of N5-10 above which the concentrations can be considered significant? Looking at the last profile (13:50) N10 > N5 are observed above 800 m, and the magnitude of the resulting negative concentrations appears to be similar to the magnitude of some of the positive concentrations which are discussed in this work.

Now focussing more on ground based measurements; they do not seem to support at all the conclusions on P8, L10-12 (“the lifted layer of freshly formed particles was transported downwards, which can be further seen by the temporal appearance of the small particles of a few nm in diameter in the aerosol data at ground level”):

Authors' response: The authors agree that the statement is wrong. A brief summary is given later on (p.9, l. 12-25). There is no proof of downwards transport in the first case and in the following, the sentences (p.9, l. 6-9) were deleted and explanations are given now in the concluding remarks.

- Two of the small particles concentration peaks in the SMPS were observed before the occurrence of the cloud. This suggest that those were not formed under cloudy conditions (as suggested by the title), and since they were seen well before the first vertical profile was obtained from ALADINA, they cannot really support/validate the observations from these profiles;

- Assuming the last two peaks observed during daytime might result from transport of particles downwards, why are they so sporadic? Which process (P8, L12 “other processes”) could explain such a behaviour? The authors should discuss more the disappearance of these particles in the spectrum;

Authors' response: In our opinion, the “other processes” and most of the questions in Comment 10 are well explained by the response to the short comment and added to the conclusion (p. 13, l. 21-31).

However, no clear typical formation event like a “banana shape” was identified by ground data. Bursts of ultrafine particles occurred in connection with increase of NO_x concentrations affected by east wind conditions under dense strato cumulus clouds. The authors' hypothesis is that the events observed here are locally restricted and not homogeneous over larger regions. This is because in particular the clouds create inhomogeneities in the intensity of solar radiation reaching the ground. On the other hand, clouds and their surrounding regions, as well as inversion layer, are influenced by large gradients in various parameters. These gradients in combination with local turbulence may create strong nonlinearities being a favourable environment for new particle formation which has been discussed theoretically by Easter and Peters (1994) and shown in-situ by Wehner et al. (2015). The dynamics of such an event being locally restricted to small regions are completely different and cannot be explained by characteristics of a large-scale event and relation to air mass.

The small NPF events may occur suddenly, followed by fast growth but also fast dilution with surrounding air, seen in sporadic clusters of TSMPS data.

- It is also surprising that the authors did not discuss at all the fact that these peaks coincided almost perfectly with the peaks of NO_x. This observation suggests a local source of anthropogenic particles; this is further supported by the occurrence of two of the peaks during the night, when NPF is usually not favoured."

Authors' response: Further, we have considered the fact, that NO_x coincide with the appearance of small particles measured with the TSMPS in Figs. 2a,b (see p. 1, 17-19; p. 7, l. 17-19).

During the first measurement day, a maximum of $3.5 \times 10^3 \text{ cm}^{-3}$ for N₅₁₀ was detected above an inversion layer in connection with a condensation sink and layer of high moisture that existed until noon. However, altered clusters (particle size between 30 and 50 nm) were measured at ground during the process in relation to an increase of NO_x so that anthropogenic emissions might have been favoured the NPF occurrence.

The values of NO_x varied between 10.2 and 34.6 $\mu\text{g m}^{-3}$ during the day, with temporary higher concentrations in the early morning at 03:50 UTC, from 07:55 to 09:40 UTC and in the afternoon around 18:00 UTC that further coincide with the appearance of ultrafine particles estimated with the TSMPS data.

RC1: "Comment 11, P8, L15-19: The authors suggest that small particles observed close to the ground are originally formed close to the inversion layer, where, based on Fig. 4, the concentrations of large particles are significantly decreased compared to lower altitudes (and this idea is further mentioned in the conclusions, on P11, L34). This behaviour is thus not similar to what was reported by Rose et al.(2015), who found that in contrast NPF was occurring under "high" CS values, at least when compared to non-event days. Same comment applies to P11, L5-8."

Authors' response: Yes, indeed. We have deleted the phrases and compared our studies with more suitable literature of Easter and Peters (1994) and Wehner et al. (2015) instead of Rose et al. (2015). For detailed explanations, please take into account the answers to the short comment.

RC1: "Comment 12, P8, regarding instrumental setup and data analysis: Why are the instruments and methods applied different between the two case studies (eg: ceilometer/LIDAR, NO_x measurement/no NO_x measurement, no fluxes investigation/fluxes investigation)? I understand that LES-model was used in the second case in the absence of measurements from ALADINA. However, it would have been interesting to have similar simulations for the first case in order to evaluate the ability of the model to reproduce measurements, and give further insight into the reliability of the model outputs used in the analysis of the second case. Also, more information is needed regarding measurement of CO₂ fluxes and LWP, as well as additional description/explanation for Fig. 7a. Last, the authors should provide more information about the use of this fluxes analysis in the frame of studies dedicated to NPF: to which extent is this approach novel, was it used previously, was it modified compared to earlier work?"

Authors' response: As it was stated already, the LES model and EC station were only available during the second measurement day, as part of the MelCol 2015 campaign. At this point, we point to the detailed explanations on p.1 of the authors' response.

RC1: "Comment 13, P9 and following: the authors refer to their observations as nucleation; this is not correct since only >7 nm particles are observed. The use of "formation events" (eg P9 L6, L9) is also questionable, and an expression such as "the appearance of small particles" would in my view better describes the observations."

Authors' response: The authors agree that formation events might be misleading with regard to TSMPS data in Fig. 6, as the measured aerosol particles are altered already. We have changed the expressions in the results and abstract, accordingly. P. 10, l. 3, p. 10, l. 8, p. 12, l. 15, p. 13, l. 12, p. 13, l. 17

In case of the expression "NPF" for the size of 5 and 7 nm, respectively, we refer to the authors' response to the short comment by Wolfgang Junkermann (here p. 13ff)

RC1: "Comment 14, P9, L5-7: Based on the surface plot of the particle number size distribution (Fig. 6a), the increase of the particle concentration seems to be seen up to 30 nm (instead of 20 nm), and it is thus not correct to say that these events coincided with a decrease of the particle concentration in the range 20-50 nm."

Authors' response: Thank you for the correction. We have changed the size range between 30 and 50 nm instead of 20-50 nm (p. 9, l. 30, p. 10, l. 4).

RC1: “Comment 15, P9, L22-23: The sentence “The water vapour mixing ratio q increased during the day in the vertical distribution between 0 and 1500 m” is confusing and should be rephrased.”

Authors' response: The authors agree that the formulation causes confusion and the sentence was changed to: The water vapour mixing ratio q reached maxima between the heights of 950 and 1210 m near the inversion layer. The total maximum of $q=18.5$ g/kg was estimated (...) p. 11, l. 2-3

RC1: “Comment 16, P9, Figure 8: “The vertical profile of turbulent kinetic energy... showed a strong connection with the structure of the ABL” (L24-25). What does this sentence mean? Also, regarding the analysis provided on P10, L6-8: it is quite complex to discuss/assess the effect of the parameters shown on Fig. 8 a and b since the vertical distribution of N7-12 is shown to vary faster than the time resolution of the model outputs.

Authors' response: The authors agree that the description of the TKE is not sufficient. We intend to describe fast mixing processes in the well-mixed boundary layer that could be taken from the highest TKE rates. Yes, the model output is in 1 h interval, but in our opinion, it is still necessary to calculate the rates in order to explain the fast mixing processes seen in the CPC data. As it was described already, no other meteorological data was available in the verticality.

RC1: “Comment 17, P10, L12: The expression “validate” might be too strong, as some of the profiles do not start from ground level, and thus do not allow for a direct comparison. This analysis is anyway very interesting, and, as mentioned previously, I would recommend to do it also for the first case study.”

Authors' response: Thank you for the comment. Indeed, the expression “validate” is too strong in this case. We have changed the corresponding lines (p. 11, l. 25).

For comparison, TSMPS data was estimated for the same size of N_{7-12} (...)

In our mind, the integration of TSMPS data into vertical profiles would not benefit for the first case, as only larger particles were captured by the ground observation. The focus of Fig. 4 is on the impact of ABL processes and the progress in time.

RC1: “Comment 18, P11, L3-20: It seems that some of the conclusions regarding the first case study reported in this section are different from those provided in section 3.1. In particular, the sentence on P11 L13-14 “however ground observations did not catch the newly formed boundary layer aerosol” does not suggest anymore the transport of particles downwards, and contrasts with the sentence from P8 L10-12 “the lifted layer of freshly formed particles was transported downwards, which can be further seen by the temporal appearance of the small particles of a few nm in diameter in the aerosol data at ground level”. Surprisingly, a possible connection with anthropogenic sources of particles is mentioned but NO_x measurements shown in Fig 2b are not used to support this hypothesis. Also, the reference to Bianchi et al. (2016) should be reconsidered; the end of the sentence should be rephrased and the message clarified, as Bianchi et al. (2016) suggest that organic compounds from anthropogenic origin (and not CO) are involved in the NPF events they observe. CO is only used as a tracer for the anthropogenic origin/signature of the sampled air masses.”

Authors' response: Thank you for the detailed explanations in Comment 18 and Comment 19. The conclusion was revised in the discussion paper. Please, take into account the changes. Further, the correlation of NO_x and sporadic events, as role for anthropogenic emission is considered in the revised form.

Thank you for the comment regarding the literature of Bianchi et al. (2016). Yes, indeed the outcome of the reference was stated wrong in our manuscript. The authors wanted to express that NPF occurs under more conditions as expected so far (p. 13, l. 1-4).

The study of Bianchi et al. (2016) at the Jungfraujoch site in the free troposphere might support the present observations that NPF occurs under more conditions as expected so far; NPF was not related to sulphuric acid formation, which is a common identifier for nucleation, instead NPF depended on high concentrations of organic compounds from anthropogenic origin.

RC1: “Comment 19, P11, L21-33: While the authors consider the possibility for the particles formed aloft to have disappeared on L24, they assert on L25-27 that these particles are those observed close to the ground after vertical mixing. These sentences seem to be a bit contradictory, and the authors should be more careful in providing their conclusions, and use some formulations such as “might”, “could”, “suggest” ... In particular, the analysis of the particles and CO₂ fluxes used to support the conclusions seems to be uncomplete. Indeed, while CO₂ fluxes show similar behaviour during the events observed at 10:10 and 16:10, particles fluxes show contrasting behaviour, being slightly positive during the first event and negative during the second one. This cannot be simply summarized as “During sporadic formation events, significant deposition occurred, taken from the negative values of particle fluxes. In

contrast, in between formation events, emission was observed, indicated by positive particle fluxes” (P9, L13-15). Also, as already suggested for the first case study, the authors should discuss more the disappearance of the particles.”

Authors' response: Thank you! We have completed the explanations of the particle fluxes (p. 10, l. 15-25).

In order to understand possible local processes in the current work, additional estimations of particle fluxes will be considered for the day. Fig. 7 displays particle fluxes taken from the EC (eddy-covariance) station that was deployed several metres away from the UAS operation and ground monitoring. Short lifetime and quick fluctuations of emission (shown in blue, positive particle flux) and deposition (shown in red, negative particle flux) are prevailing during the day. Significant deposition occurred with a maximal particle flux of $-26 \times 10^6 \text{ m}^2 \text{ s}^{-1}$ at 12:30 UTC, taken from the 30 s averaging, that is in relation to the occurrence of ultrafine particles and the brake of ABL clouds. Deposition with a maximum of $-75 \times 10^6 \text{ m}^2 \text{ s}^{-1}$ was shown by Buzorius et al. (2001) during a clear nucleation event with particle growth rate, measured with cut-off sizes of 7 and 14 nm, respectively. Moreover, the study calculated the high frequency 87% of downwards fluxes during nucleation events that might lead to the assumption of local sources from upper layers so that not only sources near ground level should be considered. Possible causes for NPF in the ABL due to turbulence were as well previously suggested by Nilsson et al. (2001).

RC1: “Comment 20, P11, L23: “The maximum ... and increased rapidly while descending”. This sentence is confusing, please try to rephrase.”

Authors' response: Thank you! Yes, the sentence is not correct and it was reformulated: (...) decreased while descending (p. 13, l. 8)

Technical corrections

RC1: “P3, L14: “high” should be replaced by “large”.”

Authors' response: Thank you for the comment. The authors have done the change, although the reference was not considered in the section (p. 12, l. 21).

RC1: “P5, L10: “and” instead of “und”.”

Authors' response: Thank you for the hint of the typo! The authors have changed it accordingly, although the sentence was removed, anyway (p. 5, l. 30).

RC1: “P6, and following sections: For clarity I would suggest to give all particle concentrations as N, and not dN/dlog(dp).”

Authors' response: The authors agree that the expression might cause confusion. We have done the changes accordingly (p. 7, l. 9).

RC1: “P6, L8: “of” instead of “on”.”

Authors' response: Thank you. Change was done (p. 7, l. 4).

RC1: “P7, L11-12: I would suggest to change the expression “in the particle diameter”. The authors can simply refer to N5, N10, N5-10, N390, N500 and N700 as those were defined before.”

Authors' response: The authors agree that it is easier to follow the results by the simple expression of N5, N10, N5-10, N390, N500 and N700, respectively, as the cut off sizes (CPCs) and size bins (OPC) were mentioned already in Section 2.1 p. 4, l. 1-12. The sentence was reformulated to “total aerosol particle number concentration N5 (red line) and N10 (blue line), given by the two CPCs, and the total aerosol particle number concentration with the three OPC channels of N390 (pink line), N500 (green line) and N700 (turquoise line) ...” (p. 8, l. 8-9). However, we decided to maintain the description in Fig 4.

RC1: “P7, L31: Please try to reformulate the expression “particles belonging to the diameter”.”

Authors' response: According to the previous comment, the authors used the simple term of N390. Therefore, the sentence was changed to: “At the same time, N390 was evenly distributed below the inversion layer...” (p. 8, l. 28)

RC1: “P8, L32: I would suggest to change the expression “in the particle diameter”.”

Authors' response: *The authors have done the following change in order to avoid the expression "in the particle diameter": "in the size between 30 and 50 nm" (p. 9, l. 30).*

RC1: "P9, L1: "in the particle diameter range between 3 and 10 nm" instead of "of 3 and 10 nm"."

Authors' response: *Thank you! We have reformulated that sentence (p. 9, l. 32-33).*

RC1: "P10, L6: "790 m" instead of "790 nm"."

Authors' response: *Thank you for the comment. The typo was changed (p. 11, l. 19).*

RC1: "P10, L7: "N7-12 decreased to the total aerosol number concentration...". This is confusing, please try to reformulate."

Authors' response: *The authors agree that the sentence causes confusion. Therefore, we have deleted the expression "to the total aerosol number concentration" (p. 11, l. 20).*

RC1: "P10, L16: "integration of a size scan"."

Authors' response: *The authors have done the change and the sentence is now complete (p. 11, l. 30).*

RC1: "P11, L7: "high concentrations of sulphuric acid"."

Authors' response: *The change was made on (p. 12, l. 21).*

RC1: "P17, Fig. 1: I would suggest to show the location of the station on the map, even if it said that the station is located in the domain centre."

Authors' response: *We will mark the domain centre in the new version.*

RC1: "P18, Fig. 2: the ticks on the x axis do not ease the reading of the times (10 intervals for 6 hours). Same applies to Fig. 6."

Authors' response: *Thank you for the hint! We will revise the ticks of the x axes accordingly.*

RC1: "P20, Fig 4: Logarithmic scales should be used for particle concentrations as it is very difficult to seen the variations of N5-10 and N700."

Authors' response: *Thank you for the suggestion. If the variations will be more significant to capture, we will change the scales in the new version.*

Comments of referee #2 (RC2)

"In summary this paper presents observations of non-surface layer new particle formation under cloudy conditions and near surface vertical transport of newly formed particles. Observations in the vertical were made using the UAS system ALADINA in the region of the TROPOS research site near Melpitz: TROPOS facilities providing surface observations. 53 hours of flight data was available spanning a period 23 days between October 2013 and July 2015. Of the available data two periods were selected as case studies.

Although the paper is well structured and presented the quality of the English is poor making for a difficult read and confusion as to what point the author is trying to convey – there are far too many corrections to list here. There is also a tendency for the author to use emotive/subjective phrases for example: "All in all, the research site Melpitz of TROPOS offers a great potential for observing NPF within the ABL and inter-comparison of airborne data with monitoring at ground." These are major failings and ones that need to be addressed: if the author is not a native English speaker it is recommended that they find somebody who is to assist in editing.

The work presented is unique and will be of great interest to the community but there needs to be a substantial "tightening up". The paper gives the impression of slackness and carelessness in experimental techniques, discussion of relevant processes, and how conclusions have been arrived at. At this stage and in this form I would not recommend publication but I would encourage the authors to re-submit after revision."

Author's response to RC2

The authors acknowledge the anonymous referee #2 for the support in review process. The given comments will help to "tightening up" the manuscript in its present form. As it was assumed already, the authors are not native speakers, but will take into account the editorial support, if still necessary after revision. In most cases, the authors will refer to previous comments to referee #1 and to the detailed answers to Wolfgang Junkermann. So we therefore ask you for understanding that most of the responses are rather short.

Please, find attached the point-to-point response and the marked-up manuscript version including the changes (removed is marked in red and blue is coloured in blue).

Point-to-point response:

RC2: "1. The authors cite a number of papers concerning the experimental set up at both the field site on for the UAS. It would be expected for a basic discussion of both these setups to be included in the paper and a discussion of the various factors that will effect measurement uncertainty and what has been done to mitigate them. This is very important when it comes the particulate measurement as inlet losses and transmission losses can seriously bias a measurement: for example is the fact that you are not seeing particle above a certain size because they are not there in the first place or because they have all been lost before reaching the sensors."

Authors' response: The authors agree that this is a major point. However, we decided not to repeat the technical part of the measurements, as it was clarified in the previous paper by Altstädter et al., 2015, already. In addition, the results in Platis et al., 2016 showed a good agreement between UAS and surface data (seen in NAIS). The focus of the current publication was only on the results. Nevertheless, we have extended the section (see comments on p. 1).

RC2: "Basically have you characterised the both the ground and UAS sampling and measuring systems. If you have then discuss it and the implications. If this has not been done you would be advised to do so."

Authors' response: Yes, we did several comparisons with the UAS and ground measurement, sampled at the same aerosol inlet at ground. One event was shown (for instance), in Bärffuss et al., 2018. The system was redesigned in the meteorological set up, but the aerosol instrumentation is still valid. The uncertainty of +/-20 % of the CPCs was verified by comparison of N7-12 with TSMPS data in the same diameter size. Please, take into account our remarks at the very beginning of the authors' response on p. 1. (...) The following lines were added to the new manuscript (p4., 16-18):

The laboratory results were confirmed by Bärffuss et al. (2018) during field studies in Melpitz. The UAS was placed at the same aerosol inlet as ground monitoring and N7-12 coincide with ground data in the same particle size within the deviations of +/-20% during sampling period of 1.5 h.

RC2: "2. There is also the point regarding humidity and this applies to both the ground and UAS measurements. You are measuring the humidity of the ambient air sample upstream of your sensors. If that air mass is warmed (resulting in its RH reducing) or purposefully dried then this will impact on particle size. There is a need to be exact as to what the conditions are at the point of measurement and to indicate what effect this might have."

Authors' response: This is correct, the relative humidity needs to be considered for size-resolved measurements without drying the sample flow. Ground based aerosol measurements at Melpitz are always performed under dry conditions according to Wiedensohler et al. (2012), that means the measurements are performed below 40% rH in the

sample flow. Thus, further heating does not influence the particle size anymore. But for measurements on ALADINA rH may have an influence. In general, a change in rH does not alter the particle number concentration but it may change the particle diameter. This could influence the lower cut of the CPCs in comparison with dry measurements and the upper and lower diameters of OPC size channels. In general the hygroscopic growth factors are smaller for smaller diameters. For a typical aerosol composition in Melpitz a 10 nm-particle has a growth factor of approximately 1.3 at 90% rH and a 500 nm-particle a growth factor of 1.8. This is the maximum difference in diameters for our measurements if we compare dry and ambient measurements. Since the aerosol flow is also heated while entering the inlet system of ALADINA, the real deviation will be smaller.

As mentioned above, the hygroscopic growth does not change the number concentration measured using ALADINA.

RC2: “3. The need to be exact also extends to terms you are using. You refer to size but what do you actually mean: radius, diameter, and are these optical, mobility or aerodynamic equivalent values. This needed to be exact also extends to the gas phase measurements: are the concentration in ambient air or dry air? What are the uncertainties in these measurements?”

Authors' response: The gase phase measurements are in ambient air. In case of the OPC we talk about optical diameters. For the CPC cut-off detection a DMA is used, thus mobility diameter.

The uncertainties were given in Section 2.1 on p. 4: CPCs (TSI) +/- 20% and OPC (Met One Instruments) +/- 15%

RC2: “4. With regard to your CPCs. In the first instance you are using two units: one with a lower cut of 5 nm (diameter?) and one with a lower cut off of 10nm (diameter?) while in the second you are using two units: one with a lower cut off of 7nm (diameter?) and one with a lower cut off of 12nm (diameter?) and on page 4 lines 1 – 6 this is clear. Lines 10 – 14 on page 7 this is not the impression the text gives. This is what you say: "total aerosol particle number concentration measured with two CPCs in the particle diameter of 5 nm (red line) and 10 nm (blue line)". What I think you are talking about is the total aerosol concentration measured by the two cpc: red line cpc with 5nm lower cut off, blue line the cpc with a 10nm lower cut off.

On line 12 of the same page you say: "OPC in the particle diameter of 390 nm (pink line), 500 nm (green line) and 700 nm (turquoise line)". An OPC has specific measurement bins that will have a centre value of particle diameter and a specified width – you need to add the width of the bin

to this kind of statement and state explicitly that you are using the bin centre value.

Authors' response: Thank you for the comment, however, the size bins were described in the previous section. Please see the description of the OPC on p. 4, l. 19-24. For simplification, the bins were named to N390 for size range between 390 and 500 nm, N500 for 500 to 700 nm and N700 stand for 500 to 700 nm, respectively.

RC2: “5. Your figures show no error bars nor is there any reference in the captions: this needs to be addressed. On the subject of figures, although I am not colour blind many of your readers may be. It is good practice to choose colour schemes that those who are colour blind can interpret.”

Authors' response: Thank you for the hint! We will change the colour scale in a different way. Please take into account the given references of the instrumentation (marked up in the discussion paper).

RC2: “6. In your discussion of the use of the LES you use the term “closure”: however what you are actually doing is using the model output to substitute for missing observations.

Authors' response: Yes, indeed. Please, see the comments to the first referee. We have changed the misleading expression accordingly (p. 1, l. 20).

RC2: “Again the need for exactness but there is also the question of the validity of this approach.

What evidence can you present that would indicate that the LES has accurately representing the conditions at the site. Presenting a comparison the model data with flights where you have full observations would be advised.”

Authors' response: Again, we refer to the first comments of the authors' response, as the data set was not the same during the both measurement days. The comparison of LES model with UAS data is not the scope of the paper, but we will consider another measurement day for the overview paper of the MelcCol 2015 campaign. Further, the output considers data from the German weather station in Torgau (3 km away from the site) and radiation measurements from the Melpitz site. Additional information was provided in the text in Section 2.3 (p. 6, l. 19-23).

“The utilised model set-up is similar to the one described in Heinze et al. (2017), and it includes large-scale forcing tendencies due to advection of heat and water vapour, subsidence from COSMO (Consortium for Smallscale

Modeling, Baldauf et al., 2011) reanalysis (Heinze et al., 2017); direct, diffuse and terrestrial radiation (1 min averages) directly measured at the field site and soil data (temperature and moisture) from the German weather service (Deutscher Wetterdienst, DWD) station "Klitzschen bei Torgau", which is located 3 km away from the Melpitz field site."

RC2: "7. Lidar and ceilometer. You put a deal of emphasis on the importance of these observations without the necessary explanation and discussion of the observation themselves. What are you actually looking at what is the implication to the interpretation arising from the change of laser wavelength?"

Authors' response: The authors intended to obtain a full daily analysis of the cloud coverage and aerosol layers, as measurement flights were only operated below clouds and during sunlight. Besides, the data and lidar was used to affirm the existence of clouds during the UAS observations. For detailed description of the backscatter signal, we refer to p. 5, l. 25-29.

RC2: "8. In your paper introduction you introduce many concepts and make many statements backed up by a paper reference – you could really do with expanding this by adding brief textual explanations."

Authors' response: Thank you very much for the hint. The authors agree that the introduction is short in its present form. However, we wanted to summarise the so-called favoured conditions for NPF that are stated in the different literature from "(e.g. Wiedensohler et al., 1996; Keil and Wendisch, 2001; Birmili et al., 2003; Kulmala et al., 2004; Hamed et al., 2010; Hamburger et al., 2011)".

RC2: "9. You introduce "gravity waves" but do not provide an explanation as to why this is relevant and the process that are at play."

Authors' response: Thank you for the comment. Please, note the answer to referee #1 (p. 3, Comment 3 of authors' response).

RC2: "10. Use of the term inter-comparison. This is a tautology and a very common mistake – simply use the term comparison."

Authors' response: Thank you very much for that hint! We have changed it accordingly (p. 5, l. 9).

Short comments by Wolfgang Junkermann (SC1)

"The paper shows some interesting data on elevated ultrafine particle layers under total overcast conditions obtained, using an unmanned aerial vehicle, the UAV ALADINA, flying vertical profiles through the planetary boundary layer and lower free troposphere.

The occurrence of ultrafine particles under such meteorological conditions would not be expected following the general description of new particle formation observed elsewhere.

The authors nevertheless, claim a clear identification of a new particle formation event. Typically for the production of ultrafine particles via new particle formation or, with the old expression, gas-to-particle conversion, shortwave radiation is suggested to be required as an initial step to convert first sulphur dioxide into sulphuric acid, which then reacts in the atmosphere often with ammonia or similar compounds producing initial aerosol clusters. This is well established in a large number of publications, see for example for a review Kulmala et al, 2011, and Kulmala et al, 2013 for an analysis of the expected particle generation process. Growth occurs then via (EL)VOC, further sulphuric acid water etc. . The data shown here were measured under conditions without sufficiently high UV radiation levels to produce sulphuric acid in the atmosphere, thus requiring a different production process. Such an occurrence of ultrafine particles under totally overcast conditions has been observed several times and was reported for example over Germany (Junkermann et al, 2016), a paper that is cited in the manuscript. Here these particles were apportioned to primary industrial sources

burning fossil fuel. Hence, as several of these elevated sources are located around Melpitz the observations are not really surprising. Anyway, the data analysis leaves several questions"

Authors' response: First of all, the authors acknowledge Wolfgang Junkermann for his detailed comment and discussion. However, we disagree in some points. Please, find attached the corresponding answers.

SC1: "1) The manuscript states that NPF occurrence with subsequent downward transport was clearly identified. The question: What does 'new' mean within this context? Was the particle formation (cluster production and subsequent growth) really been observed?"

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

A gas to particle (NPF) event however, should have as a minimum requirement initially the majority of particles in the lowest size bins of the distribution. The number of particles in the size between 7 and 12 nm (or N7-12) has then to be higher than the number of particles > 12 nm before and at the time the growing nucleation mode is passing this size range. Here, within the data presented the difference between the size bins is marginal, compared to the overall number concentrations indicating rather an aged air mass. Occasionally the N10 is even higher (measurement uncertainty?) than N5., in Fig. 4b and 4c N10 is missing at all."

Authors' response: This paragraph might be misleading. The UAS delivers a snapshot of the current state and not the formation process itself, due to missing data of chemical gas phase and the use of CPCs models with a lower cut-off size of at least 5 nm. The authors did not state at any time, that sources for NPF were investigated by the set up. Besides, this is not the goal of the publication, as it is only to show the evidence that NPF occurs more often as expected so far and would not be taken into account by pure surface data. We use the expression "new particle formation" or NPF for particles in the size range of 5 to 10 nm (or 7 to 12 nm), in contrast to larger, aged aerosol particles.

Now, the answer to this comment is more detailed, because all the following comments are linked to this one: Yes, that is correct, NPF consists of two steps, but the result is the occurrence of small particle. That means, if we observe the result, we can conclude that particles were formed by gas-to-particle conversion, because this is the only process to produce such small particles in the atmosphere. We observed an increased number concentration in the size range below 12 nm. The only way to produce such small particles is gas-to-particle conversion, thus we know that NPF took place without measuring the clusters itself. This is a well-established method to investigate and identify NPF events.

We think the events observed here are locally restricted and not homogeneous over larger regions. This is because in particular the clouds create inhomogeneities in the intensity of solar radiation reaching the ground. Thus, we cannot expect a homogeneous development of the boundary layer. On the other hand, clouds and their surrounding regions as well as inversion layer are regions with large gradients in various parameters. These gradients in combination with local turbulence may create strong nonlinearities being a favourable environment for new particle formation which has been discussed theoretically by Easter and Peters (1994) and shown in-situ by Wehner et al. (2015). The dynamics of such an event being locally restricted to small regions are completely different and cannot be explained by characteristics of a large-scale event and relation to air mass.

The small NPF events may occur suddenly, followed by fast growth but also fast dilution with surrounding air. If the stadium of dilution starts, the number concentrations decrease immediately and if an event is detected in this stadium the number concentration below 12 nm is less significant.

SC1: “2) Following Kulmala et al, 2013, it takes several, ~about five or more, hours for a new particle to grow into the lowest size range of the particles that can be observed with the instruments on ALADINA. Observation of particles at 08:00 UTC thus requires a production at ~03:00 UTC or even earlier during the night. These particles have to be generated in a different area, a minimum of five hours upwind, which is according to HYSPLIT at least 70 km. Growth rates are normally smaller or even zero at night. Thus the source could be even further away.”

Authors' response: Growth rates can be much higher in combination with turbulence. Small fluctuation in combination with large gradients may create local supersaturations of potential precursor gases for nucleation and growth. The lifetime of such events could be extremely short as shown in Wehner et al. (2015), where Markku Kulmala has been co-author and did not see any conflict to his earlier studies. The formation and growth of these small and short events is different to those happening over larger areas and longer time scales. The latter ones are often combined with the typical banana-shaped measurement on ground and occur over larger areas.

SC1: “3) Where is the production area of the particles, which are 'clearly' identified as originating from NPF, according to HYSPLIT ~70-100 km for 5 h?”

Authors' response: At this point, we refer to the comment above. The NPF process here is different and occurs on a much shorter time scale.

SC1: “4) Why are the meteorological and atmospheric chemical conditions favorable for new particle formation > 70-100 km upwind, during the night and in the elevated layer?

Where do the precursor chemical compounds come from? What could be the initial step for nucleation cluster generation at night?”

Authors' response: Again we would like to refer to the previous points: NPF did not start hours ago, because the growth rates are much higher in this case. These events are very local, thus we assume the particles have been formed right in the measurement region and less than one hour ago.

SC1: “5) Is there probably a source for ultrafine particles upwind that could explain the results?

HYSPLIT shows that the air mass has been close to the German-Polish border on the evening of April 3. Here we have at least three power stations that are sources for primary particles in the respective size ranges as well as additional large amounts of sulphur dioxide, ammonia (from the SCR cleaning process) and also internally produced sulphuric acid (see also Junkermann et al, 2016). Size distributions, independence on time of the day (Junkermann and Hacker, 2015) and laboratory results confirm primary emission of 'New' particles (Brachert et al, 2013). Such particle production does not require UV radiation or OH radicals. A quick HYSPLIT analysis: For June 21 the winds in the altitude of 600 m above ground come from the northwest passing the power station Buschhaus as the next possible candidate. Further upwind (up to 24 h) along the backtrajectory the industries of Groningen (NL) are located. For westerly to southwesterly winds in the PBL the power stations Schkopau and Lippendorf as well as the refinery Leuna would be possible sources for primary ultrafine particles and further precursors over Melpitz (see also Platis et al, 2015). For size distributions and source strength of such sources see Junkermann et al, 2011; 2016.”

Authors' response: If there would be a source of ultrafine particles upwind of the site, the particles would grow fast during the transport. The authors doubt that ultrafine particles are directly emitted by a combustion source. Modern burning facilities using oil or natural gas emit gases which may form ultrafine particles directly in the exhaust due to cooling and condensation. This happens also in the exhaust of cars and these ultrafine particles can be measured already close to the tailpipe. However, they were not emitted as ultrafine particles, they were formed afterwards. Combustion processes produce larger particles with diameters above 50 nm and no ultrafine particles. Furthermore, ultrafine particles in Melpitz and elsewhere have been characterized to be volatile. This means they consist of condensable material and were not formed during e.g. a combustion process.

Furthermore, if the ultrafine particles observed in Melpitz would have been formed far away, they would not appear as single bursts. Such small structures with sharp gradients to the environment would dilute very quickly within the order of minutes in maximum. Thus, it is simply not possible that these particles were produced elsewhere and transported to the site.

SC1: “6) Page 9, line 20, atmospheric boundary layer conditions were unfortunately not available from the UAS. Instead a model is used to characterize the ABL. However, this model is not able to reproduce the measured vertical structure (Fig. 9). Why are no model data shown for 07:00 to 08:00, the time window before the

aircraft measured the pronounced vertical profile? Later on the PBL rapidly mixed. The vertical structure was visible only within the first 15 minutes of the model 1 h time window.”

Authors' response: The authors agree that the presence of different data set of the two measurement sites was not clear in the old version of the manuscript. Please, take into account the comments on the first page of the authors' response. But, we do not fully understand the questions (in point 6). Fig. 8 shows modelling profiles of potential temperature, mixing ratio and TKE for each hour from 6 to 16 UTC. In Fig. 9, aerosol profiles obtained with ALADINA and ground-based instrumentation is shown. However, aerosol profiles are not derived from the model.

SC1: “7) Fine particles: particle number concentrations > 390 nm on April 4 are fairly high.

We can compare to particle numbers measured with a GRIMM 1.108 (fraction > 400 nm, second size bin) over Kathmandu (missed approach to Kathmandu international airport) in January 2014, to data gathered during flights in the extremely dusty Mexico City basin during Milagro 2006 or to data from the heavily polluted Po-Valley (QUEST, 2004). The number concentrations there were only about half or less of what has been seen over Melpitz. Under these conditions the condensational sink (CS) would be extremely high and according to most of the published literature a NPF event would be very unlikely. How is this in agreement with the summary / abstract statement that New Particle Formation has been clearly observed? The summary claims: Further, the NPF event was linked to a condensation sink of larger particles belonging to the accumulation mode at the same altitude. That's not in agreement with the fig. 4, the layers are clearly separated.”

Authors' response: The OPC number concentration in the size class 390 – 500 nm (N_{390}) was between 200 and 250 cm^{-3} , which is not extremely high. To validate the vertical measurements we can take the number size distributions measured on ground and integrate over the corresponding size range. Additionally we have to consider that on ALADINA the sample flow was not dried, while on ground, the number size distribution was measured below 40% rH behind a dryer. Thus, if we want to compare both measurements, we have to consider the hygroscopic growth of the particles. On April 4 between 9 and 14 UTC rH varied between 70 and 90% corresponding to a growth factor between 1.17 and 1.25. Using this, the diameters of N_{390} move to smaller sizes and the size range for dry particles would be 320 ± 10 nm to 410 ± 10 nm. Integration over this size range results in values between 230 and 320 cm^{-3} during the observed period. This fits well within the uncertainties to the results from vertical measurements. The layers are not separated in Figs. 4a and 4b. We mentioned the occurrence of a second layer in Fig 4c (p. 8, l. 32-33) that did not coincide with the first layer above/near the inversion layer.

SC1: “8) Fig. 9 and page 11, line 22/23: 'The maximum of total aerosol particle number concentration was 1.6_105 cm^{-3} at the altitude of 420ma.g.l. and increased rapidly while descending'. Not in agreement with the figure. The number concentration below 420 m actually is always lower than the maximum at 420 m and decreased between 08:13 and 08:26 UTC. Probably also a result of changing wind directions (~15 deg within 2 hours)”

Authors' response: Thank you for the comment. The authors agree that the sentence is wrong and have changed it accordingly (p. 13, l. 8).

SC1: “Summarizing: The paper does not really show evidence or proof for gas to particle conversion respective new particle formation. Though interesting and good to identify ultrafine particle layers the instrumentation used is most probably not appropriate to detect such particle formation events. An SMPS or NAIS would be necessary. The results are contrary to the general literature about new particle formation and unfortunately, there is no attempt to analyze where the particles observed are produced or originating from. However, the measurements would be in agreement with emissions from a few well known 'continuous generators for ultrafine particles' e.g. power stations /refineries and the final statement that a thorough meteorological analysis (but, not only in the vertical) is necessary to understand ultrafine particle behavior in the PBL is strongly supported.”

Authors' response: The authors did not intend to state new particle formation production, as it is well known that the CPCs are not valid for the smallest diameters of a few nm. However, as it is stated in the title, the authors wanted to show, that new particle formation was observed under conditions that are in general not favourable for NPF. Besides, you can take from the title that only observations are stressed and not the initiation of NPF.

Authors' changes

Please not additional changes that were done by the authors. For instance, some literature was added/revised during the review process.

- p.1, affiliations: *Now at: Laboratory for Air Pollution/Environmental Technology, Empa, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland
- p.1, l. 3-4 Inserted affirmed by ceilometer and lidar data
- p.1, l. 9 Inserted “mixed and mainly”
- p.1, l. 12 changed two cases into “two measurement days”
- p.5, l. 19” was expected” instead of were expected
- p.15, l. 10-12 Inserted new literature for COSMO (Baldauf et al., 2011)
- p.15, l. 13-15 During review process, the publication of Bärffuss et al. (2018) was accepted.
- p.15, l. 30-31 New literature of Buzorius et al. (2001)
- p.16, l. 1-3 Added new literature of Engelmann et al. (2016) for polly
- p.16, l. 7-9 During review process, the publication of Größ et al. (2018) was revised.
- p.16, l. 19-20 Inserted new literature of Heese et al. (2010).
- p.17, l. 20-21 New reference of Pal et al. (1992)
- p.18, l. 20-21 New literature of Wiegner and Geiß (2012)
- p.19, l. 4 changed typo of resolution

Airborne observations of newly formed boundary layer aerosol particles under cloudy conditions

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Abstract. This study describes the ~~formation of freshly formed~~ [appearance of ultrafine](#) boundary layer aerosol particles under classical called "non favourable" conditions by the existence of low-level strato cumulus clouds, [affirmed by ceilometer and lidar data](#). Airborne measurements for vertical profiling atmospheric boundary layer properties and aerosol particles in the diameter range between 5 nm and 10 μm were repeatedly performed with the unmanned aerial system ALADINA (Application of Light-weight Aircraft for Detecting IN-situ Aerosol) at the research site of TROPOS (Leibniz Institute for Tropospheric Research) in Melpitz during three seasons between October 2013 and July 2015. More than 100 measurement flights were performed during this period on 23 different days with a total flight duration of 53 h. On 26 % of the measurement days, new particle formation was measured close to the inversion layer and was observed to be [mixed and mainly](#) transported downwards during short time intervals of cloud gaps. However, the typical banana shape of new particle formation and growth was not seen at ground, only sporadic events appeared with low particle growth rate and thus might not have been classified as NPF by pure surface studies. This presentation focuses on two ~~cases~~ [measurement days](#) influenced by the passage of a low pressure system and minimal concentrations of SO_2 as indicator for precursor gases at ground; I) on April 4, 2014 by east wind and II) on June 21, 2015 under south-west wind. ~~For a closure, an LES-model output was used for the second study in order to derive a full analysis of atmospheric boundary layer growth over the measurement site at small-scale with high spatial resolution.~~ [During the first measurement day, a maximum of \$N_{5-10}=3.5 \times 10^3 \text{ cm}^{-3}\$ was detected above an inversion layer in connection with a decline of larger particles and a layer of high moisture that existed until noon. However, altered clusters \(particle size between 30 and 50 nm\) were measured at ground during the process in relation to an increase of \$\text{NO}_x\$ so that anthropogenic emissions might have been favoured the NPF occurrence. Whereas by south-west wind conditions, pronounced turbulent fluxes of sensible and latent heat, taken from an LES model output](#) ~~turbulent fluxes of sensible and latent heat~~ in the vertical distribution initiated fast mixing processes of freshly formed boundary layer aerosol. Furthermore, CO_2 and particle fluxes indicated downward transport and high deposition during sporadic ~~nucleation events~~ [appearances of ultrafine particles](#), so that ~~NPF-occurrence~~ [the existence of ultrafine particles](#) above with subsequent downward transport was clearly identified.

1 Introduction

The knowledge of atmospheric aerosols is still incomplete and thus contributes to the most significant uncertainties in climate model predictions, especially the aerosol sources, sinks and transformation processes (IPCC, 2007). Depending on their optical properties, aerosols interact significantly with solar radiation and hence influence the climate directly. Especially the formation of new particles due to nucleation by gas to particle conversion is subject of investigations. In this context, the formation and growth rates of nucleation mode particles (ca. 1-15 nm) need a more profound understanding, as by subsequent and sufficient growth, the particles can act as cloud condensation nuclei and therefore influence the Earth's climate indirectly via affecting the hydrological cycle (e.g., Kerminen et al., 2005). Particle burst events were measured worldwide on different platforms like research stations for long-term monitoring, ships and airborne systems. Studies were performed at various altitudes from the boundary layer up into the lower stratosphere and suggested different sources and proposed several mechanisms for new particle formation (e.g., Wiedensohler et al., 1996; Keil and Wendisch, 2001; Birmili et al., 2003; Kulmala et al., 2004; Hamed et al., 2010; Hamburger et al., 2011). According to these studies, new particle formation (NPF) is likely during preconditions of low temperature, high water vapour content, low surface of pre-existing particles that are often in relation to a low condensation sink, presence of precursor gases and high incoming solar radiation. However, further studies presented events of NPF under non favourable conditions, likewise under the presence of clouds (e.g., Wiedensohler et al., 1997) ~~Größ et al. (2015)~~, and these investigations stress the hypothesis that nucleation ~~is~~ might be possible under a wider range of conditions than it has been expected so far.

In particular, the small-scale vertical distribution of aerosols in the atmospheric boundary layer (ABL) needs a more profound understanding and has to be implemented in models (Boy et al., 2006). A strong connection between the vertical variability of aerosols and thermodynamic structures, caused by turbulence in the continental boundary layer, has been identified by Boy et al. (2003) at SMEARII (Station for Measuring Ecosystem-Atmosphere Relations) in Hyytiälä in Finland. The study suggested a connection between NPF and higher values of turbulent kinetic energy, in particular for so called "A-events" with high formation and growth rate of particles with a particle diameter of 3 nm. Nilsson et al. (2001) assumed that formation mechanisms are caused by dynamic processes in the mixed layer and entrainment zone. ~~Further, Bigg (1997) suggested the existence of breaking waves in the mixed layer as another possible mechanism for NPF, as breaking waves occurring in regions with high humidity and temperature fluctuations.~~ Bigg (1997) suggested that high humidity and temperature fluctuations may enhance new particle formation, that was supported by in-situ measurements shown in the previous study of ALADINA in Platis et al. (2016). Besides, Easter and Peters (1994) assumed favourable conditions near the inversion due to mixing processes that was afterwards demonstrated by Siebert et al. (2004) with the help of vertical profiling the continental ABL with a balloon-borne system. However, the measurements mentioned so far, except for the balloon, were conducted only in one fixed location so that the situation on a larger scale and in particular, a temporal development in different altitudes is missing.

In this context, the use of airborne systems for atmospheric research is essential to deliver a detailed three-dimensional picture of the aerosol spatio-temporal distribution from the surface up to the free troposphere. The results of O'Dowd et al. (2009) showed the large-scale variability of the particle concentrations along air mass trajectories, and NPF and growth was

observed over distances of several 100 km. For small-scale differences in atmospheric conditions (e.g. cloud occurrence), NPF is distributed heterogeneously. The continental cloud top is favourable for NPF, furthermore the cloud-free regions in between of two cloud parcels (Keil and Wendisch, 2001).

The study of Wehner et al. (2015) presented the variety of nucleation mode particles around clouds over Barbados. The measurements were performed with the helicopter-borne measurement payload ACTOS (Airborne Cloud Turbulence Observation System) in the boundary layer. A high frequency of occurrence of nucleation events (around 83 % of all measurement days) was observed, whereby 50 % of the events existed on a small-scale within 100 m in horizontal extent. In addition, the important role of the entrainment zone for NPF was confirmed by the large data set of 91 flights and the influence of clouds for favourable NPF conditions was verified. Hamburger et al. (2012) studied atmospheric aerosols with the research aircraft FAAM BAe-146 and DLR Falcon 20 and suggested nucleation events in the ABL caused by the presence of high pressure systems and one event in the free troposphere around 8 km altitude due to the updraft during frontal passages. All in all, a high vertical variability of atmospheric aerosols at different locations but in particular in the ABL was observed, however no detailed evolution of the vertical distribution of ABL aerosol in the same area.

This publication presents results of the unmanned aerial system (UAS) ALADINA that has already been extensively used for boundary layer aerosol field studies in Melpitz (Altstädter et al., 2015; Platis et al., 2016). ~~Those previous~~ Previous investigations of Platis et al. (2016) showed NPF in correlation with temperature and humidity fluctuations of several orders of magnitudes higher than in the remaining part of the ABL. In addition, downwards transport of freshly formed particles to ground level was observed ~~on a day of high incoming solar irradiance, therefore along a classical day for new particle formation. Furthermore, the downward transport of freshly formed particles was~~ and supported by the appearance of nucleation ~~and particle growth~~ mode particles and their subsequent growth and relating to an increase of SO₂ concentrations by clear sky. increasing concentrations of sulphur dioxide measured at ground.

Due to the ~~high~~ large data set of vertical profiles that were performed during three different seasons with ALADINA at the research site of TROPOS since 2013, a more complex role of new particle formation influenced by ABL processes is identified. In contrast to typical NPF events at ground by high incoming solar irradiance, other events were observed that might have not been considered by surface observations due to sporadic appearance of ultrafine particles. The publication benefits from the comparison of ALADINA with instrumentation at ground and one model output in order to get a better understanding of NPF conditions between surface and the free troposphere.

This article is structured as followed: Section 2 provides an overview of the unmanned research aircraft ALADINA and operation at the research site of TROPOS. An overview of the expanded version of the LES model output, that was used for a closure of ABL processes during the second case study, is presented in Subsect. 2.3. Section 3 shows the results of newly formed boundary layer aerosol under cloudy conditions during the two case studies, respectively. Finally, Sect. 4 concludes the presented study with the main goal to show the complexity and more frequent occurrence of NPF in the ABL, as expected so far.

2 Instrumentation, model and research site for profiling boundary layer aerosol

2.1 Aerosol and meteorological payload on the unmanned research aircraft ALADINA

The aircraft Carolo P360 "ALADINA" (Application of Light-weight Aircraft for Detecting IN-situ Aerosol) was designed and developed for atmospheric research in order to investigate the vertical and the horizontal aerosol distribution depending on atmospheric boundary layer properties. ~~A detailed description of the airplane is given in Altstädter et al. (2015) and information on the new set-up can be found in Bärffuss et al. (2018).~~ The aircraft design and instrumentation's set up were introduced in Altstädter et al. (2015) and supported by the results of Platis et al. (2016). During review process, the aircraft was re-engineered and we refer to the newest study of Bärffuss et al. (2018) who presented a profound overview of ALADINA's flight operation, data procedure and the current payload that ~~The payload on ALADINA~~ is equipped with aerosol instrumentation and meteorological sensors with high temporal resolution.

The total aerosol particle number concentration is derived by two Condensation Particle Counters, CPCs (model 3007, TSI Inc., St Paul, USA), with different lower threshold diameters. In the first case study here, the cut-off sizes were 5 and 10 nm, respectively. The difference in the particle number concentrations of both CPCs (N_5 and N_{10}), in the following referred to as N_{5-10} , is used for the number concentration of freshly formed particles. During the second case study, the lower threshold diameters of both CPCs were 7 and 12 nm (N_7 , N_{12} ; N_{7-12}), respectively. The CPCs were characterised to measure within an uncertainty of $\pm 20\%$ with a fast response time of 1.3 s. The laboratory results were confirmed by Bärffuss et al. (2018) during field studies in Melpitz. The UAS was placed at the same aerosol inlet as ground monitoring and N_{7-12} coincide with ground data in the same particle size within the deviations of $\pm 20\%$ during the sampling period of 1.5 h.

In addition, an Optical Particle Counter, OPC (model GT-526, Met One Instruments Inc., Washington, USA), is installed and measures the size distribution of aerosol particles with six channels from 0.39 to 10 μm (ambient) in particle diameter with an uncertainty of $\pm 15\%$ and a temporal resolution of 1 s. Here, the aerosol particle size distributions are analysed in the size windows between 390 to 700 nm, as larger particles were not relevant in the study due to minimal appearance. In the following, the particle size distributions of the three channels refer to the total aerosol particle number distribution in the size range between 390 and 500 nm (N_{390}), between 500 and 700 nm (N_{500}) and 500 to 700 nm (N_{700}).

The meteorological instruments are mounted at the tip of the aircraft nose next to the aerosol inlet. The sensor package consists of one five hole probe for measuring the three-dimensional wind vector with a temporal resolution of up to 40 Hz and wind speed with an accuracy of $\pm 0.5 \text{ m s}^{-1}$ (Wildmann et al., 2014a). The fast temperature sensors have a resolution of 10–20 Hz with an accuracy of $\pm 0.1 \text{ K}$ (Wildmann et al., 2013). Additionally, one humidity sensor is integrated that probes the water vapour content with a response time of 1.5 s with $\pm 3\%$ RH accuracy (Wildmann et al., 2014b).

2.2 Research site Melpitz and available instrumentation during experiments

The research site Melpitz of TROPOS (51° 32' N, 12° 56' E, 87 m a.s.l.) is located in the lowlands of Saxony, 41 km NE of Leipzig, Germany, and surrounded by flat grass, agricultural areas and forests (e.g. Spindler et al., 2001; Spindler et al., 2004).

The flat surface and the fact that no obstacle is in the direct vicinity of the station, enables the use as airfield for a safe take-off and landing of the UAS.

Air masses arriving at Melpitz consist up to 60 % of originally maritime air, due to long distance transport by westerly winds and with predominantly enhanced concentrations of organic matter, sulphate and nitrate (Spindler et al., 2012). In the other frequent case, air masses are transported from industrial regions of the continental site of Eastern Europe and therefore polluted with distinctly higher aerosol particle number concentrations in the boundary layer (e.g. Engler et al., 2007; Junkermann et al., 2016). Aerosol loads are in this case primarily characterised by anthropogenic emissions and classified as "near city background" by Putaud et al. (2004).

The research site Melpitz of TROPOS is well known in NPF studies and benefits from inter-comparison comparison of airborne data with monitoring at ground. Manninen et al. (2010) investigated NPF events at 12 different European sites including Melpitz within the framework of the EUCAARI (European Integrated project on Aerosol, Cloud, Climate, and Air Quality Interactions) project. During the intensive measurement period from March 2008 till April 2009, 57 % of the available analyses were classified as NPF event days with the major occurrence in warm seasons and a significant maximum in May. The highest occurrence of NPF during summer was measured by Hamed et al. (2010), who focused on NPF events in connection with sulphur dioxide (SO₂) concentrations in a ten year period for the Melpitz site from 1996–2006. The fraction of NPF occurrence was 30 to 50 % and related to an increase of SO₂ concentrations. During the earlier 1.5 y period between 1996 and 1997, 50 % of the available data was classified as nucleation with a maximum in June. However, only 30 % corresponded to NPF in the period from 2003 and 2006 with the highest frequency from June–September.

To conclude, high occurrence of NPF in spring and summer ~~were~~ was expected during airborne observations of the presented study. In order to demonstrate a consistent analysis, the concentrations of precursor gases are studied that were measured at ground station in 1 min intervals with the major role of anthropogenic SO₂ for new particle formation. A detailed description of meteorological sensors and gas analysers on site can be taken from Hamed et al. (2010). Sulphur dioxide concentrations were studied as main precursor gas at ground in 1 min intervals. The temporal evolution of the aerosol at ground level was measured by a Twin Scanning Mobility Particle Sizer, TSMPS, with a 20 min scan between 3 nm and 800 nm in particle diameter (Wiedensohler et al., 2012). Clouds and aerosol layers are indicated in the lowermost 3–4 km with backscatter signals of ceilometer (in the wavelength of 1064 nm; e.g. Heese et al., 2010; Wiegner and Geiß, 2012) and lidar (in the wavelength of 532 nm; e.g. Pal et al., 1992; Engelmann et al., 2016). During the first measurement day, ceilometer data is taken into account, whereas during the MelCol 2015 experiment, the Polly^{XT} lidar (e.g. Althausen et al., 2009) was available. The given range corrected signal is the uncalibrated attenuated backscatter coefficient that is damped by molecules and particles, respectively. ~~In order to classify the diurnal cycle of clouds in the vertical structure and top of the ABL, ceilometer and lidar data were used.~~ During the second measurement day In addition, EC data of particle fluxes were estimated via an available eddy-covariance (EC) station was used for the second case study in order to observe vertical transport processes of aerosol particles and to link between surface and UAS data. The effect of local mechanical effects due clouds, is further investigated by fluxes of CO₂.

2.3 Large-eddy simulations with forced mesoscale model output for MelCol 2015

ALADINA was operated during the field study Melpitz Column (MelCol) from June 16 until July 1, 2015. For the second case study on June 21, 2015, an LES model output was available and is used for ABL description in order to derive a continuous data set of the vertical distribution of latent heat fluxes ($w'q'$), sensible heat fluxes ($w'\theta'$) and turbulent kinetic energy (TKE).

5 The model itself is described in the following.

Simulations associated with the MelCol 2015 measurement campaign are performed with the All Scale Atmospheric Model (ASAM, Jähn et al., 2015). It has recently been used to investigate heat island effects on atmospheric boundary layer modification, cloud initiation and vertical tracer mixing in the trade wind regime (Jähn et al., 2016). For the present study, large-eddy simulations (LES) for selected days during the campaign were performed with respect to the present synoptic situation, including changes due to large-scale advective tendencies and incoming radiation.

The computational domain is $25.2 \times 25.2 \text{ km}^2$ wide with Melpitz field site located in the domain centre. Since the focus lies on atmospheric boundary layer, the model top is set to a height of 4 km. The land use around Melpitz is characterised by mainly different forest types (farm land, shrubland, urban areas), see Fig. 1. Due to the relatively flat environment, orographic structures are not taken into account for the simulations, i.e., the model domain could be chosen as flat surface.

15 The model physics is described by the prognostic TKE equation (Deardorff, 1972; Moeng and Wyngaard, 1989), by the two-moment microphysics scheme (Seifert and Beheng, 2006) with excluded ice phase in order to save computation time and by the multi-layer soil and land-use model (TERRA_ML, Doms et al., 2011) with a revised scheme for the surface layer (Jiménez et al., 2012).

The utilised model set-up is similar to the one described in Heinze et al. (2017), and it includes large-scale forcing tendencies due to advection of heat and water vapour, subsidence from COSMO ([Consortium for Smallscale Modeling, Baldauf et al., 2011](#)) reanalysis (Heinze et al., 2017); direct, diffuse and terrestrial radiation (1 min averages) [directly measured at the field site](#) and soil data (temperature and moisture) from the German weather service (Deutscher Wetterdienst, DWD) station “Klitzschen bei Torgau”, which is located 3 km away from the Melpitz field site.

3 Results and discussion

25 [To almost one third of the performed flight days with ALADINA, bursts of ultrafine particles were observed under cloudy conditions close to the inversion layer. Two of the six measurement days were selected explicitly due to similar weather conditions, low SO₂ concentrations, but different wind \(NE, SW\) directions that however mainly influenced the site \(e.g. Spindler et al., 2012; Engler et al., 2007\).](#)

3.1 Case I April 2014

30 In this section, NPF observations are shown on April 4, 2014, as this day is in contrast to previous investigations from Platis et al. (2016) under clear sky conditions. Large scale analysis revealed that air masses were transported initially from Saharan

regions over Eastern Europe within the last days via north-easterly winds. According to data of surface pressure systems that are publicly available by DWD, the research station was influenced by a low pressure system and especially by the passage of a cold front that finally occluded around 18:00 UTC (-02:00 MEZ).

A typical "banana shape" (Heintzenberg et al., 2007), as a consequence of adequate condensation ~~on~~ of precursor gases, was not observed this day. The development of the size distribution at ground level was calculated from TSMPS (see Subsect. 2.2) data from the research site. The total aerosol particle number concentration in the diameter range of 10 to 800 nm was rather low with a mean concentration of $8 \times 10^3 \text{ cm}^{-3}$ in Melpitz on April 4, compared to other days in spring time (e.g., Hamed et al., 2010; Platis et al., 2016). Particles belonging to a diameter range between 100 and 200 nm were predominant with a mean total aerosol particle number concentration of $1.0 \times 10^4 \text{ cm}^{-3}$ ~~in $-dN/d\log D_p^{-1}$~~ and evenly distributed during the whole day. Larger particles with a particle diameter from 200 to 800 nm played a minor role with a few hundred particles per cm^3 . Only the ultrafine particles with 10 to 100 nm particle diameter could be distinguished in the temporal distribution and were clustered in two events in the early morning at 02:00 and 04:15 UTC, also one further event in the afternoon at 14:00 UTC (Fig. 2a). A fourth sporadic occurrence of particles in the diameter range between 80 and 150 nm was detected at around 19:00 UTC and reached a total maximum aerosol particle number concentration of $1.5 \times 10^4 \text{ cm}^{-3}$.

In addition, gas concentrations of SO_2 and NO_x are presented in Fig. 2b. The SO_2 concentration varied between 1.4 and $4.4 \mu\text{g m}^{-3}$ from the early morning until 10:00 UTC. Afterwards, the concentration increased slowly to a total maximum of $11 \mu\text{g m}^{-3}$ at 20:40 UTC and decreased to $4.8 \mu\text{g m}^{-3}$ in the night. The values of NO_x varied between 10.2 and $34.6 \mu\text{g m}^{-3}$ during the day, with temporary higher concentrations in the early morning at 03:50 UTC, from 07:55 to 09:40 UTC and in the afternoon around 18:00 UTC that further coincide with the appearance of ultrafine particles estimated with the TSMPS data.

The temperature was low between 5.3 and 12.3°C and the relative humidity reached high values up to 98.1 % in the early morning with a maximum at 08:25 UTC. Due to specific more and only short periods of clear sky, the global radiation, G , varied between 400 and 720 W m^{-2} (Fig. 2c). The wind speed was moderate in the range of 2 and 4 m s^{-1} . Besides one sharp change of the wind direction, dd , from north to south-west between 08:15 and 08:55 UTC, the prevailing wind direction was from north-east (Fig. 2c).

3.1.1 Cloudy conditions and heterogeneously mixed atmosphere

A heterogeneous lower atmospheric structure was identified with the ceilometer data and the results for the lowermost 4 km are presented in Fig. 3. A stable night time ABL with a depth of less than 600 m was observed from midnight until around 07:00 UTC. Dense ABL clouds formed within this humid layer from about 07:00 to 10:00 UTC. After 10:00 UTC, the clouds dissolved and the humid convective boundary layer started growing. The maximum ABL depth was observed at around 14:30 UTC with an ABL-top of 800 m. Convection decayed and a residual layer (RL) remained after 16:00 UTC. Various particle (shown as green and yellow colours) and cloud (shown in white) layers were observed at different altitudes during the whole day. Partly, the lofted aerosol layers (steady existent up to 1.5 km) were mixed into the ABL during the growth process. The higher aerosol layers above 1.5 km height were also present during the whole day but did obviously not affect the boundary layer aerosol conditions. After 20:00 UTC, precipitation was observed which did not reach the ground.

3.1.2 NPF in the vertical distribution

In the following, vertical profiles obtained with the UAS ALADINA are shown and the flight times are summarised and connected with weather conditions and gas concentration of SO₂ in Tab. 1. During Case I, on April 4, six measurement flights were performed between a maximum height of 700 and 1000 m from 06:15 until 13:58 UTC. Figure 4 displays four vertical profiles, from left to right, of potential temperature θ , water vapour mixing ratio q , total aerosol particle number concentration measured with two CPCs in the particle diameter of 5 nm (red line) and 10 nm (blue line), respectively, and the total aerosol particle number concentration observed with the OPC in the particle diameter of 390 nm (pink line), 500 nm (green line) and 700 nm (turquoise line) N_5 (red line) and N_{10} (blue line), given by the two CPCs, and the total aerosol particle number concentration with the three OPC channels of N_{390} (pink line), N_{500} (green line) and N_{700} (turquoise line) in the time interval at (a) 09:06 UTC, (b) 10:45 UTC, (c) 11:47 and (d) 13:50 UTC.

The first profile was taken at 09:06 UTC (Fig. 4a) and showed a strong inversion layer in the height between 420 and 550 m a.g.l. that was influenced by air masses of high moisture, identified by the rapid increase of q from 2 to 18.5 g kg⁻¹ within the inversion layer. N_5 decreased continuously from 6.0×10^3 to 4.0×10^3 cm⁻³ between the height of 100 and 420 m a.g.l. Above, NPF event was observed, shown by $N_{5-10} = 3.5 \times 10^3$ cm⁻³ that was strongly connected to the layer of high moisture. Above the inversion, the total aerosol particle number concentration decreased to 3.0×10^3 cm⁻³ for the particles measured with both CPCs and remained constant to the height of 700 m. In case of the OPC data, larger particles were mixed below inversion layer and significantly decreased above. At ground, less than 250 cm⁻³ were measured with the 390 nm channel, 50 cm⁻³ for the particle diameter of 500 nm and larger particles (700 nm to 5 μ m) were not detectable. Up to the inversion layer at 420 m a.g.l., the vertical distributions of N_{390} and N_{500} were constant and decreased rapidly along with the maximum of N_{5-10} . Between the altitude of 470 and 700 m a.g.l., the total aerosol particle number concentration of N_{390} and N_{500} were constant with low values of $N_{390} = 110$ cm⁻³ and $N_{500} = 20$ cm⁻³.

At 10:45 UTC (Fig. 4b), the inversion layer was still detectable and lifted up to the height between 500 and 700 m a.g.l. and connected to the layer of high moisture. Here, only data from the CPC with the lower cut-off size of 5 nm were available. The CPC detected particles up to 0.8×10^4 cm⁻³ between ground level and the capping inversion at the height of 450 m a.g.l. Hereafter, the amount of N_5 increased steadily to 3.8×10^4 cm⁻³ at the altitude around 700 m a.g.l. with two distinct layers of particles. Below the inversion, ultrafine particles were evenly dispersed within the BL, but above the inversion a heterogeneous distribution was observed up to the height of 800 m a.g.l., where the total aerosol particle number concentration decreased rapidly to a minimum of 0.5×10^4 cm⁻³. At the same time, ~~particles belonging to the diameter of 390 nm were~~ N_{390} was also evenly distributed below the inversion and reached values around 220 cm⁻³. Within the height of 450 and 500 m, the concentration decreased to $N_{390} = 130$ cm⁻³. Between 500 and 950 m in altitude, the total aerosol particle number concentration varied between 110 and 120 cm⁻³. The same distribution occurred for particles in the diameter size of 500 nm, but with minimal concentrations.

The next vertical profile was taken at 11:47 UTC (Fig. 4c). A high increase of N_5 with a maximum total aerosol particle number concentration of 3.8×10^4 cm⁻³ was observed in the height between 150 and 250 m a.g.l. A second layer with enhanced

values of N_5 was detected above the inversion layer in the heights of 630 and 950 m a.g.l. with a maximum of $3.4 \times 10^4 \text{ cm}^{-3}$. The results of the OPC were almost the same as 1 h before and significantly affected by the inversion layer.

Later, at 13:50 UTC (Fig. 4d), the total aerosol particle number concentration was detected with both CPCs again. From ground level up to the inversion at 800 m a.g.l., the maximum of $N_{5-10}=0.5 \times 10^4 \text{ cm}^{-3}$ appeared. Above 800 m a.g.l., the decline of N_{390} and N_{500} was present and linked to an increase of ultrafine particles.

~~During the temporal development of ABL aerosol, the lifted layer of freshly formed particles was transported downwards, which can be further seen by the temporal appearance of the small particles of a few nm in diameter in the aerosol data at ground level (Fig. 2a). Nevertheless, the particle growth was not sufficient or interrupted by other processes, so that a typical formation event like a "banana shape" could not be identified by ground data.~~ Regarding OPC observations, accumulation mode particles were still homogeneously mixed in the ABL up to the inversion. To sum up, vertical profiles showed the strong dependence of airborne measured particles on the structure of the ABL. The overall observation was the occurrence of NPF connected to a layer of high moisture close to the inversion depending on the decline of N_{390} and N_{500} , as an indicator for the condensation sink in that altitude that might have favoured the particle formation process. ~~The same behaviour was observed by Rose et al. (2015), who suggested NPF appearance by highest condensation sink in the transition zone between ABL and free troposphere (FT).~~

3.2 Case II June 2015

The second case presents observations from June 21, 2015. ALADINA flights were performed as part of the MelCol 2015 experiment so that additional instrumentation was available at site. Within the last five days starting on June 16, 2015, retrieved back-trajectories showed air masses originated over the Atlantic, so that a low aerosol load was expected. Further, the research site was influenced by a low pressure system that led to a mixed structure of strato cumulus (StCu) clouds in the height of 500–2500 m. The cloud coverage can be further taken from the Polly^{XT} lidar (e.g., Althausen et al., 2009; Engelmann et al., 2016; Baars et al., 2017) data from 06:00 until 18:00 UTC (Fig. 5).

An overview of the performed measurement flights with ALADINA in connection with weather conditions and gas concentrations of SO_2 between 0.8 and $3.7 \mu\text{g m}^{-3}$ can be found in Tab. 2. In this case, seven flights were performed between 08:00 and 15:32 UTC with a total maximum height of 1200 m. The temperature at 1 m height was in the range of 9.4 and 18.3°C during the day. The wind speed was weak between $0\text{--}3.7 \text{ m s}^{-1}$ coming from SW, so that nucleation was not expected by clean air masses, according to the results of Junkermann et al. (2016).

The size distribution measured by TSMPS at ground are displayed in Fig. 6a. The aerosol load was constantly dispersed until 07:20 UTC with the highest total aerosol particle number concentrations of $5.0 \times 10^4 \text{ cm}^{-3}$ ~~in the particle diameter of around 20–50 nm~~ in the size between 30 and 50 nm. Particles belonging to the accumulation mode were insignificant on this day and reached values of only a few 100 particles cm^{-3} . At 07:50 UTC, a sporadic NPF event was observed with a maximum total aerosol particle number concentration of $1.5 \times 10^5 \text{ cm}^{-3}$ ~~in the particle diameter range of 3 and 10 nm~~ in the size between 3 and 10 nm. At the same time, a significant decline of accumulation mode particles was measured. However, the new particle formation event dissolved within 20 min and the total aerosol particle number concentration of particles larger than 20 nm

increased, although the concentrations were still low during the day. In the following, five events of a significant increase of the total aerosol particle number concentration in the diameter range between 7 and 20 nm were temporarily clustered at 10:10, 11:50, 13:20, 14:50 and 16:10 UTC. The sporadic ~~formation-events~~ appearances of small particles were ongoing with a decrease of total aerosol particle number concentration of particles in the diameter of ~~20–50 nm~~ 30–50 nm. Figure 6b displays the diurnal cycle of CO_2 fluxes, F_{CO_2} , estimated with the EC station that was deployed during the analysed measurement day. During non events, the fluxes were positive and reached a maximum of $0.2 \text{ mg kg}^{-1} \text{ m s}^{-1}$ in the night at 01:30 UTC. At 05:00 UTC, negative values of F_{CO_2} occurred with a maximum of $-0.2 \text{ mg kg}^{-1} \text{ m s}^{-1}$ at 08:40 UTC, that was at the same time with the sporadic ~~formation-event~~ appearance of small particles. In order to show a direct dependence of water vapour on nucleation, Fig. 6d depicts the liquid water path, LWP, calculated from the LES model during the measurement period. Highest values of up to 10 kg m^{-2} indicated the existence of dense ABL clouds and nucleation occurred in short time period by mean LWP of 3.5 kg m^{-2} .

A similar observation of clustered NPF during cloudy conditions and west wind in Melpitz was recently presented in the study of Größ et al. (2018). Insufficient particle growth rate was significantly connected to fluctuations of hydroxyl radicals and H_2SO_4 (shown in the so called Case 4) that were steadily suppressed by clouds in contrast to high increase of H_2SO_4 during clear sky. The authors suggested local processes and steadily influences sources as cause for this formation behaviour. In order to understand possible local processes in the current work, additional estimations of particle fluxes will be considered for the day. Fig. 7 displays particle fluxes taken from the EC (eddy-covariance) station that was deployed several metres away from the UAS operation and ground monitoring. Short lifetime and quick fluctuations of emission (shown in blue, positive particle flux) and deposition (shown in red, negative particle flux) are prevailing during the day. Significant deposition occurred with a maximal particle flux of $-26 \times 10^6 \text{ m}^{-2} \text{ s}^{-1}$ at 12:30 UTC, taken from the 30 s averaging, that is in relation to the occurrence of ultrafine particles and the brake of ABL clouds. Deposition with a maximum of $-75 \times 10^6 \text{ m}^{-2} \text{ s}^{-1}$ was shown by Buzorius et al. (2001) during a clear nucleation event with particle growth rate, measured with cut-off sizes of 7 and 14 nm, respectively. Moreover, the study observed high frequencies (87%) of downwards fluxes during nucleation events that might lead to the assumption of local sources from upper layers so that not only sources near ground level should be considered. Possible causes for NPF in the ABL due to turbulence were as well previously suggested by Nilsson et al. (2001).

~~Further, Fig. 7 displays particle fluxes taken from EC (eddy-covariance) on the same date. During sporadic formation events, significant deposition occurred, taken from the negative values of particle fluxes. In contrast, in between formation events, emission was observed, indicated by positive particle fluxes.~~

Again, no typical shape of new particle formation with high and steadily increasing growth rate was observed during this day with dense cloud coverage of strato cumulus, however nucleation appeared sporadically affected by local processes of clouds. ~~by downwards transport that was indicated by negative fluxes of CO_2 and deposition of particle fluxes.~~

3.2.1 ABL properties during the NPF event

Atmospheric boundary layer conditions were not available from the UAS and estimated with the LES-model output (Sect. 2.3) in a vertical resolution of 50 m, beginning at the altitude of 25 m. The top of the ABL height varied between 900 to 1200 m in the

period of 06:00 until 16:00 UTC (Fig. 8). ~~The water vapour mixing ratio q increased during the day in the vertical distribution between 0 and 1500 m.~~ The water vapour mixing ratio q reached maxima between the heights of 950 and 1210 m near the inversion layer. The total maximum of $q=18.5 \text{ g kg}^{-1}$ was estimated in the afternoon in the 1 h time interval between 14:00 and 15:00 UTC in the height of 1200 m. The vertical profile of turbulent kinetic energy, TKE, from the surface up to 1500 m showed a strong connection with the structure of the ABL. with highest rates between 0.6 and 1.15. In the lowermost 50 m, TKE reached highest value and a total maximum of $1.18 \text{ m}^2 \text{ s}^{-2}$ between 06:00 and 07:00 UTC. At 150 m, TKE decreased compared to the surface inversion and was stable between 1200 and 1500 m. Close to the transition zone into the FT, TKE was negligible. All in all, the LES output showed a capped inversion with high moisture and moderate wind speed in the lowermost 200 m and simultaneously the highest value of TKE.

3.2.2 Vertical mixing of NPF

Two of the six sporadic nucleation events (see Fig. 6) were observed with the UAS ALADINA on June 21, 2015. The first case corresponds to the NPF event at around 08:00 UTC and the results are shown in Fig. 9. The vertical profile of sensible heat flux $w'\theta'$ and latent heat flux $w'q'$ were estimated with the LES model and presented for an 1 h interval between 08:00 and 09:00 UTC from the surface up to 1000 m in altitude. During this period, three vertical profiles of the UAS were performed at 08:13 UTC (solid line), 08:20 UTC (dotted line) and 08:26 UTC (dashed line) in the height between 100 and 950 m. Note, during this study, the lower cut-off sizes of the two CPCs were 7 and 12 nm in the particle diameter, respectively (see Subsect. 2.1). At 08:13 UTC (black line), the vertical profile of N_{7-12} showed a high variance; at 100 m the total aerosol particle number concentration was $8 \times 10^4 \text{ cm}^{-3}$ and decreased to $3 \times 10^4 \text{ cm}^{-3}$ at the height of 390 m. Above, a significant maximum of $16 \times 10^4 \text{ cm}^{-3}$ was measured at the height of 500 m and existed up to ~~790 nm~~ 790 m. Between the height of 800 and 950 m, N_{7-12} decreased to ~~the total aerosol number concentration of~~ $10 \times 10^4 \text{ cm}^{-3}$ in connection to a layer of maximum $w'q'=25 \text{ g kg}^{-1} \text{ m s}^{-1}$ and negative values of $w'\theta'$ from -2 to $-18 \times 10^{-3} \text{ K m s}^{-1}$. Only 17 min later, at 08:20 UTC, the lifted layer of enhanced aerosol concentration above 500 m was not observed anymore. However, below the altitude of 410 m, the vertical distribution of N_{7-12} was as before. At 08:26 UTC, the total aerosol particle number concentration of N_{7-12} was homogeneously mixed in the vertical distribution with a mean total aerosol number concentration of $2.5 \times 10^4 \text{ cm}^{-3}$.

For comparison, TSMPS data was estimated for the same size of N_{7-12} . ~~In order to validate the results of airborne data, integrated total aerosol number concentrations in the same particle diameter between 7 and 12 nm of the TSMPS are given.~~ At 07:40 UTC, $N_{7-12}=2 \times 10^4 \text{ cm}^{-3}$ was measured at surface, before nucleation started. During the first flight of ALADINA, the maximum of N_{7-12} was $9 \times 10^4 \text{ cm}^{-3}$ at ground. Further, N_{7-12} decreased to $3 \times 10^4 \text{ cm}^{-3}$ at 08:30 UTC and $1 \times 10^4 \text{ cm}^{-3}$ at 08:50 UTC. ALADINA observations in the lowermost 100 m were consistent with TSMPS data at ground. Thus, ground observations calculated by integration of a size scan over 20 min can not reproduce the significant spread of N_{7-12} within the ABL, but are still consistent with profiling data.

Figure 10 displays the same parameters as given in Fig. 9 for the time interval between 14:00 and 15:00 UTC, corresponding to the fifth sporadic nucleation event seen in TSMPS observations on June 21, 2015. At 14:16 UTC, the vertical profile of ALADINA (solid line) showed a homogeneous distribution of N_{7-12} with a mean total aerosol particle number

concentration of $3.5 \times 10^4 \text{ cm}^{-3}$. The second profile was measured at 14:57 UTC (dotted line) with a significant increase of $N_{7-12} = 1.8 \times 10^5 \text{ cm}^{-3}$ at ground and decreased rapidly in the vertical pattern between the height of 160 and 1100 m. The region corresponds to a maximum of sensible heat flux of $56 \times 10^{-3} \text{ K m s}^{-1}$. The third vertical profile was taken at 15:06 UTC (dashed line) and N_{7-12} was mixed in the vertical distribution by a mean total aerosol number concentration of $4.2 \times 10^4 \text{ cm}^{-3}$.

- 5 The vertical distribution of latent heat flux showed an continuously increase with a maximum of $38 \text{ g kg}^{-1} \text{ m s}^{-1}$ at 1150 m.

Again, airborne data was consistent with ground observations by TSMPS, however the temporal evolution of vertical profiles led to the assumption, that NPF occurred at ground instead of aloft. However, fast mixing was observed and at the same time an increase of the total aerosol particle number concentration in the vertical pattern.

4 Concluding remarks

- 10 Freshly formed boundary layer aerosol was measured with the unmanned aerial system ALADINA under cloudy conditions at the research site of TROPOS in Melpitz. In total, 105 airborne measurements with total flight duration of 53 h were performed on 23 measurement days since October 2013. During six of these measurement days, new particle formation (NPF) events were observed under non favourable conditions (e.g., low concentrations of precursor gases, dense cloud coverage) near the inversion layer and were mixed vertically induced by atmospheric boundary layer (ABL) dynamic processes. However, only
- 15 sporadic ~~nucleation events~~ appearances of small particles were detected at ground and might not have been taken into account as NPF event.

This study focused on two different cases: On April 4, 2014, one formation event was captured above the inversion layer at 420 m a.g.l. in relation to a significant moist layer with a water vapour mixing ratio of 18.5 g kg^{-1} after a cold front passage. Further, the NPF event was linked to a condensation sink of larger particles belonging to the accumulation mode at the same

- 20 altitude. ~~The results are consistent with the study of Rose et al. (2015), where NPF events were mainly observed during periods with a high condensation sink, not during those with high concentrations of high sulphuric acid between the atmospheric boundary layer (ABL) and free troposphere.~~ Ceilometer backscatter signal showed that the atmosphere was heterogeneously mixed with various layers of enhanced concentrations of atmospheric aerosols and several cloud levels during the whole day. Especially from 08:30 to 10:30 UTC, a dense cloud coverage at the altitude around 400 m existed. The layer of the freshly
- 25 formed aerosol spread out vertically in the boundary layer with $3.5 \times 10^3 \text{ particles cm}^{-3}$ between 5 and 10 nm in particle diameter and reached a maximum total number concentration of $3.8 \times 10^4 \text{ cm}^{-3}$ for particles with a diameter size exceeding 5 nm. Ground aerosol was simultaneously investigated with a TSMPS, however ground observations did not catch the newly formed boundary layer aerosol. The research site was influenced by NE wind, so that high emissions of anthropogenic sources were expected from Eastern Europe like stated by Junkermann et al. (2016), who found new particle formation events in connection with local plumes over Eastern Germany. However, changes in wind direction were not seen in wind measurements with
- 30 ALADINA. Nevertheless, the cold front passage might have led to occurrence of different air masses with anthropogenic emissions transported into the ABL. ~~The study of Bianchi et al. (2016) at the Jungfraujoch site in the free troposphere support the present observations; NPF was not related to sulfuric acid formation depended on high concentrations of organic compounds;~~

~~e.g. CO so that anthropogenic sources may be relevant.~~ The study of Bianchi et al. (2016) at the Jungfraujoch site in the free troposphere might support the present observations that NPF occurs under more conditions as expected so far; NPF was not related to sulphuric acid formation, which is a common identifier for nucleation, instead NPF depended on high concentrations of organic compounds from anthropogenic origin.

5 On June 21, 2015, the site was influenced by a low pressure system with strato cumulus clouds and SW wind. At 08:13 UTC, ALADINA measured a NPF event above 450 m a.g.l. in a vertical distribution of high humidity. The maximum of total aerosol particle number concentration was $1.6 \times 10^5 \text{ cm}^{-3}$ at the altitude of 420 m a.g.l. and ~~increased rapidly while descending~~
10 ~~decreased while descending~~. Within 17 min, the newly formed aerosol load was mixed vertically or disappeared as confirmed by pronounced fluxes of sensible heat with a maximum of $81 \times 10^{-3} \text{ K m s}^{-1}$ in the lowermost 160 m. The temporal evolution showed that the formation observed at ground originated from the ABL and was mixed vertically and transported downwards to the surface, as previously observed in Platis et al. (2016) under clear sky conditions. Another sporadic ~~nucleation event~~
occurrence of ultrafine particles was captured with the UAS in the afternoon between 14:10 and 15:10 UTC and showed the formation process occurred at ground and was rapidly mixed in the vertical distribution initiated by significant increase of sensible heat flux with a maximum of $58 \times 10^{-3} \text{ K m s}^{-1}$ in the lowermost 150 m.

15 Besides, the UAS observations were consistent with TSMPS data in the same diameter range of N_{7-12} , clarifying the reliability of the system. In addition, surface observations of CO_2 fluxes and particle fluxes showed the high variability during ~~nucleation events~~ sporadic bursts of ultrafine particles by downdraft and deposition, in contrast to updraft and emission during short periods of non-events by dense cloud coverage.

To conclude, high water content, ~~low surface of pre-population~~ and pronounced fluxes of latent heat, sensible heat and turbulent kinetic energy were clearly the dominant factor in the atmospheric boundary layer to make occur NPF, even if conditions were generally not favourable in contrast to previous study of Platis et al. (2016) during clear sky conditions. ~~However, no clear typical formation event like a "banana shape" was identified by ground data. Bursts of ultrafine particles occurred in connection with increase of NO_x concentrations affected by east wind conditions under dense strato cumulus clouds. The authors' hypothesis is that the events observed here are locally restricted and not homogeneous over larger regions. This is because in particular the clouds create inhomogeneities in the intensity of solar radiation reaching the ground. On the other hand, clouds and their surrounding regions, as well as inversion layer, are influenced by large gradients in various parameters. These gradients in combination with local turbulence may create strong nonlinearities being a favourable environment for new particle formation which has been discussed theoretically by Easter and Peters (1994) and shown in-situ by Wehner et al. (2015). The dynamics of such an event being locally restricted to small regions are completely different and cannot be explained by characteristics of a large-scale event and relation to air mass. The small NPF events may occur suddenly, followed by fast growth but also fast dilution with surrounding air, seen in sporadic clusters of TSMPS data.~~ To summarise, the observations lead to the assumption, that occurrence of NPF within the boundary layer are underestimated, if only ground observations are available and a more profound understanding of the vertical structure of the ABL is necessary in order to characterise NPF events.

Code and data availability. At the current state, the data sets are not publicly accessible, as analyses and further publications of the campaign MelCol 2015 are still in progress by other participants, but will be delivered upon request. The open access of data in e.g. PANGAEA will be provided in future. To get access to the source code and additional scripts for pre- and postprocessing of LES model, registration at the TROPOS Git hosting website gitorious.tropos.de/ is mandatory. Additional information can be found at the ASAM webpage
5 (<http://asam.tropos.de/>).

Competing interests. The authors declare that they have no conflict of interest.

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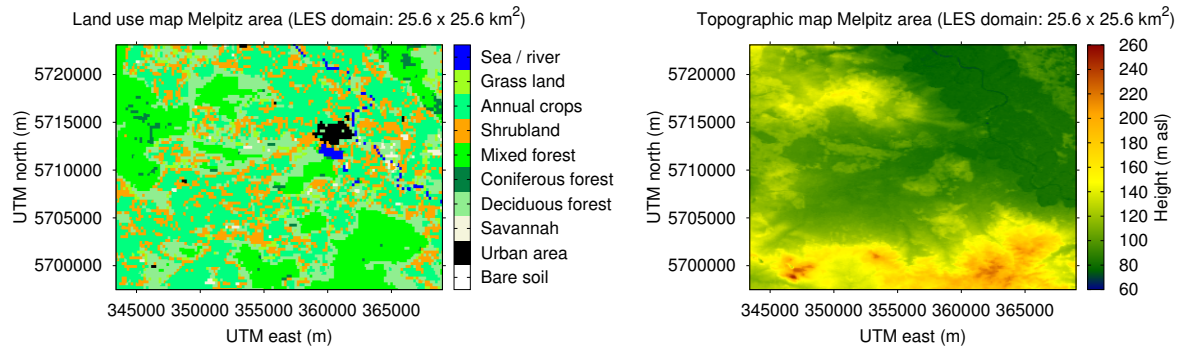


Figure 1. Land use and topography of the simulation domain [considered in the All Scale Atmospheric Model \(ASAM, Jähn et al., 2015\)](#) around the Melpitz field site, [that is located in the domain centre](#). The large variation of vegetations on small-scale are indicated by different colours (left) and the topography ranging from 60 to 260 m above sea level (a.s.l.) is also indicated with the same ~~resolution~~ [resolution](#) (right)

[Information: The domain centre will be marked in the new manuscript.](#)

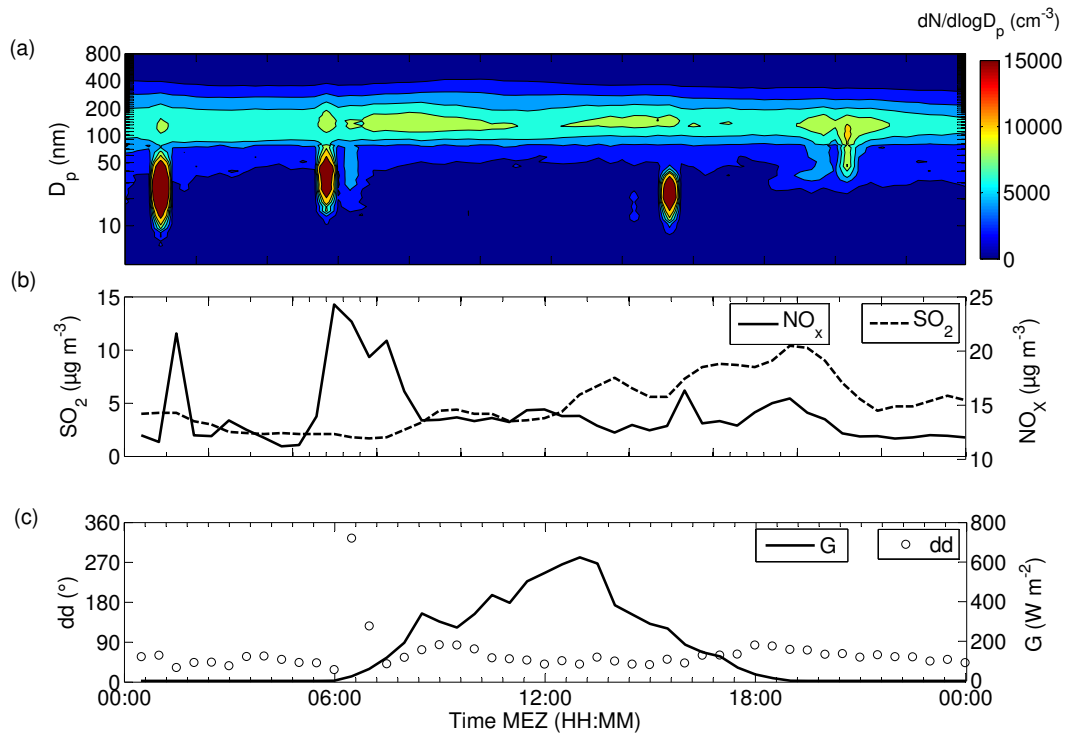


Figure 2. Ground observations in Melpitz on April 4, 2014. (a) Particle size distribution measured with TSMPS ~~in the particle diameter range D_p from between~~ 3 to 800 nm , (b) gas concentrations of SO_2 and NO_x , (c) wind direction dd and global radiation G . [Information on ground instrumentation can be taken from Wiedensohler et al. \(2012\) and Hamed et al. \(2010\).](#) ~~Information: x axes will be unitised in the new manuscript.~~

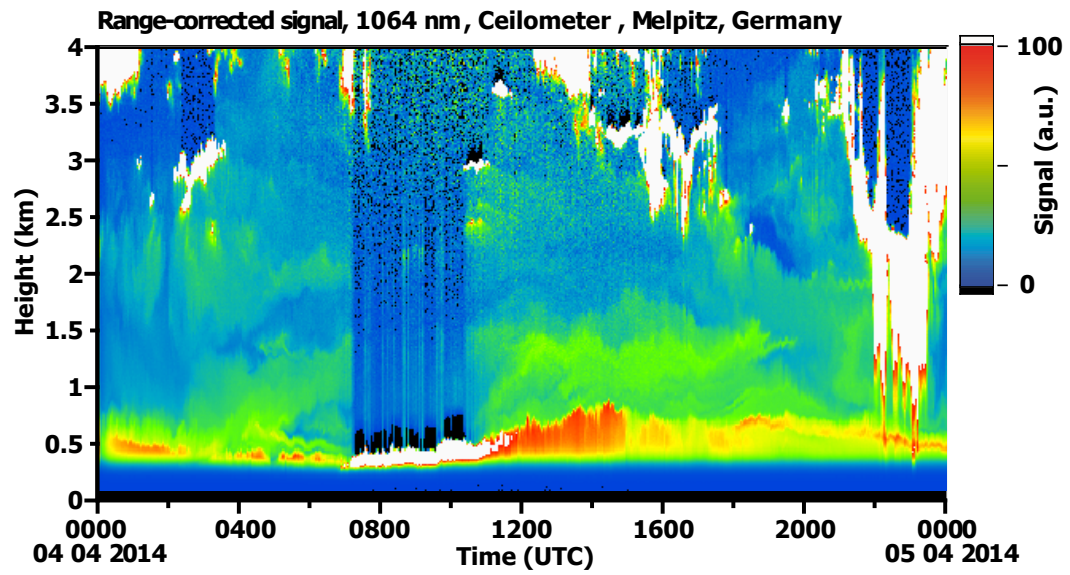


Figure 3. Backscatter signals (range corrected at the wavelength of 1064 nm) of the ceilometer installed at Melpitz on April 4, 2014. White parts show clouds and colour scales in green up to red the existence of atmospheric particles. [More details of ceilometer can be taken from Heese et al.\(2010\) and Wiegner and Geiß \(2012\).](#)

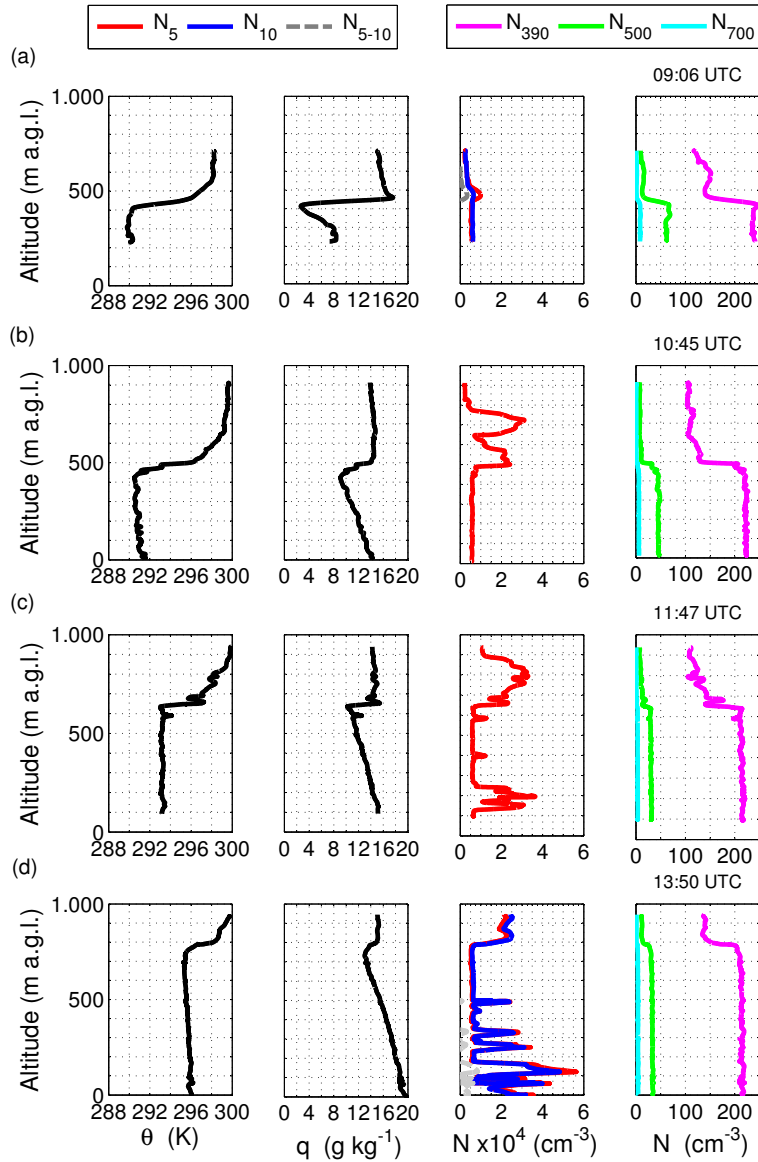


Figure 4. Vertical profiles of potential temperature θ , water vapour mixing ratio q , total aerosol particle number concentration of ultrafine particles measured with two CPCs in the particle diameter above 5 nm (red) and above 10 nm (blue) and total aerosol particle number concentration measured with OPC in the particle diameter of 390 nm (pink), 500 nm (green) and 700 nm (turquoise). All data were measured with ALADINA at (a) 09:06, (b) 10:45, (c) 11:47 and (d) 13:50 UTC on April 4, 2014. [Please note, colour scale will be changed in the new manuscript and logarithmic scale, as suggested by the referees.](#)

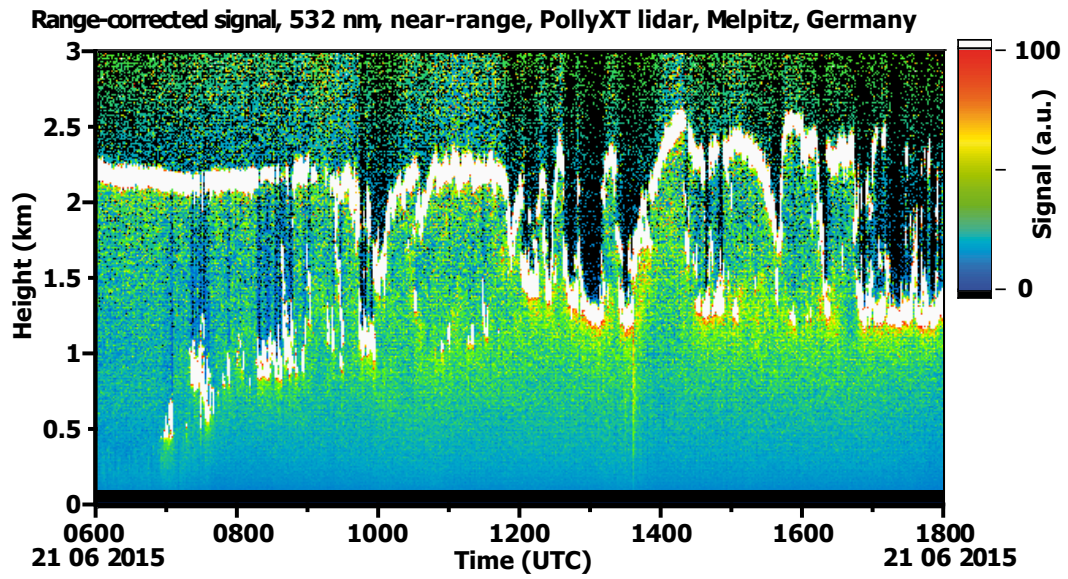


Figure 5. Backscatter signals (range corrected at the wavelength of 532 nm) of Polly^{XT} lidar installed at Melpitz on June 21, 2015. White parts show clouds and colour scales in green up to red the existence of atmospheric particles. [More details of the lidar can be taken from Engelmann et al. \(2016\).](#)

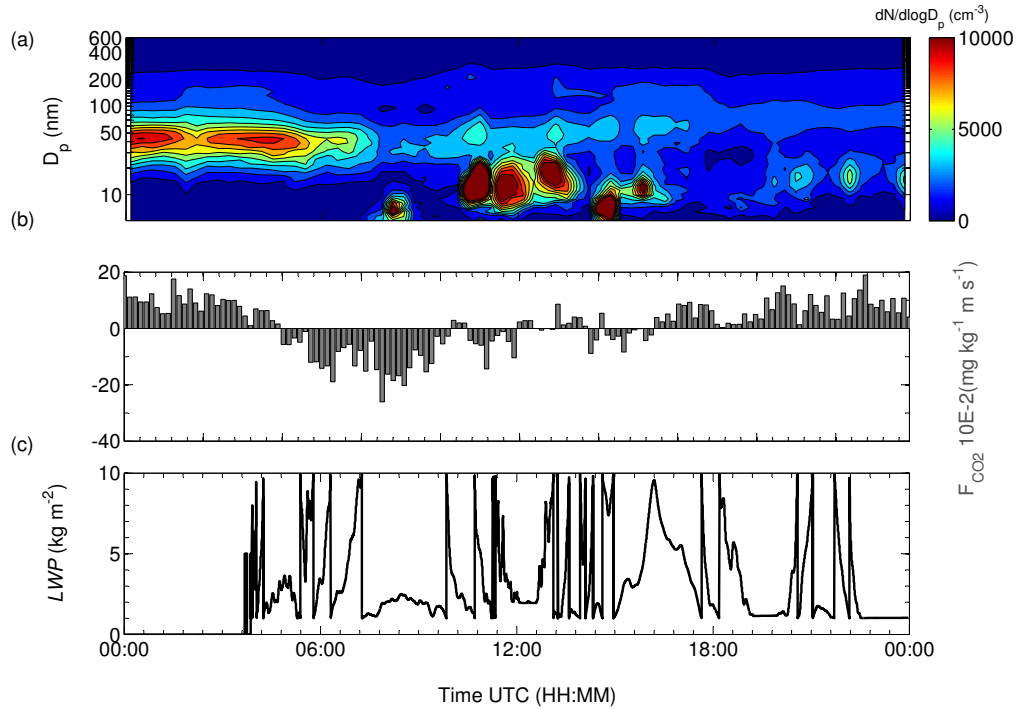


Figure 6. (a) Particle size distribution measured with TSMPS, (b) fluxes of CO_2 , F_{CO_2} , and (c) liquid water path, LWP, during MelCol experiment on June 21, 2015. The homogeneous aerosol load was affected by ABL growth after 07:30 UTC and six sporadic formation events (seen in red dots in the size range of 10 to 20 nm) were indicated during daytime between 08:10 and 16:10 UTC under cloudy conditions and SW wind.

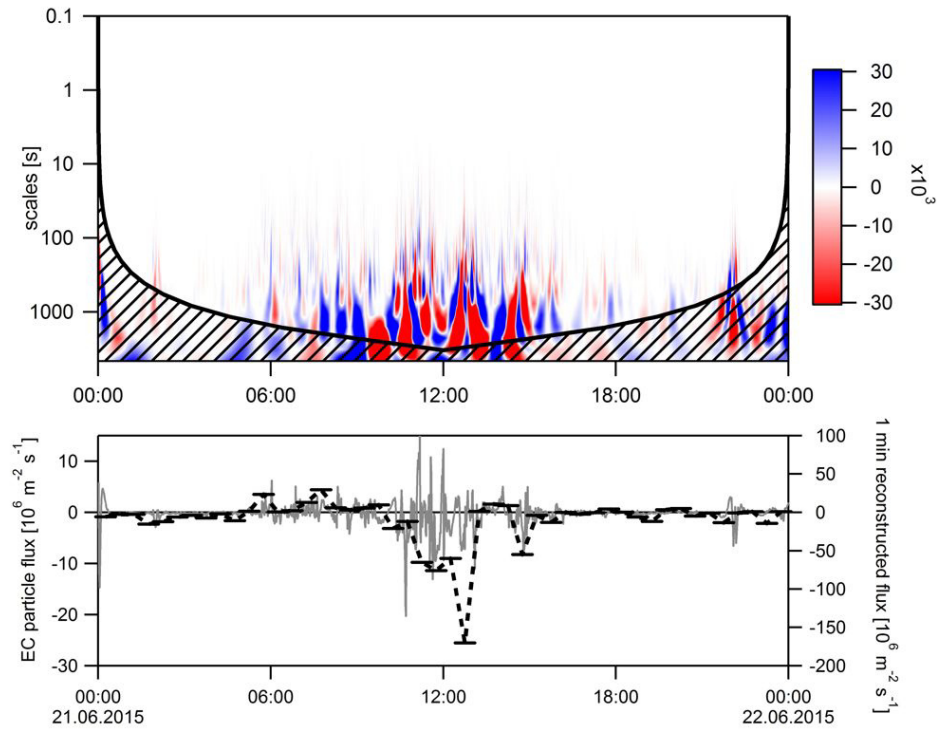


Figure 7. Wavelet analyses of EC station in time scale on top and below particle fluxes estimated during MelCol experiment on June 21, 2015. During sporadic new particle formation, deposition was observed (red), in contrast to emission by non-event sections (blue). The 30 s average of EC particle fluxes (dashed line) showed the significant deposition affected by NPF at 12:30 UTC.

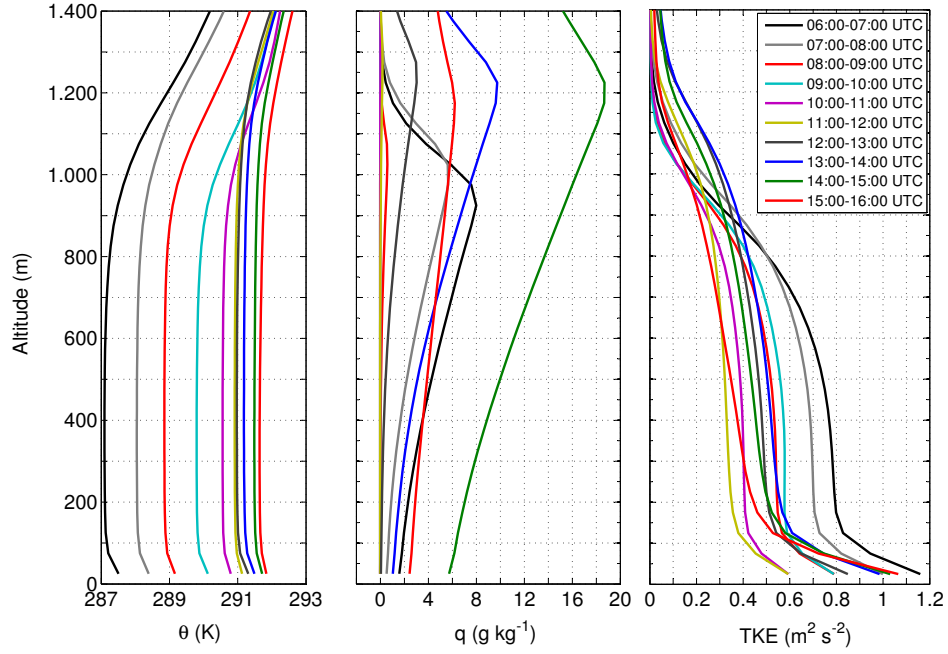


Figure 8. Potential temperature θ , water vapour mixing ratio q and turbulent kinetic energy TKE depending on the altitude, estimated with the LES model in 1 h time intervals starting from 06:00 until 16:00 UTC during the MelCol experiment on June 21, 2015. Highest values of q were estimated between the height of 950 and 1210 m close to inversion layer, respectively and appeared before sporadic NPF occurrence at ground.

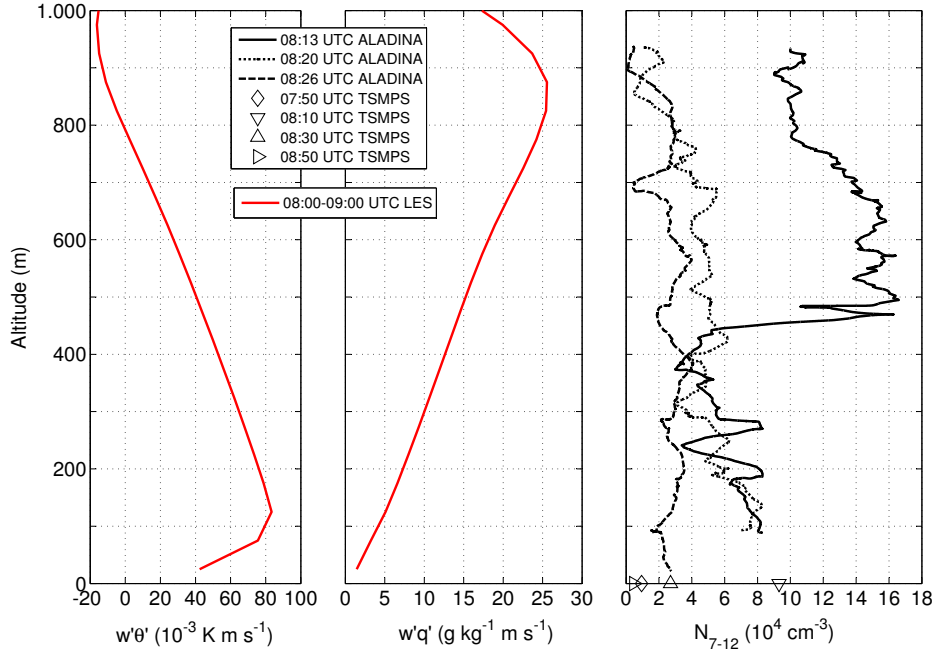


Figure 9. Vertical profiles of sensible heat flux $w'\theta'$ in K m s^{-1} and latent heat flux $w'q'$ in $\text{g kg}^{-1} \text{m s}^{-1}$ obtained by LES output in the interval of 08:00–09:00 UTC (red lines). Besides, three vertical profiles of freshly formed boundary layer aerosol in the particle diameter between 7 and 12 nm (N_{7-12}) measured with the CPCs on the UAS ALADINA at 08:13 UTC (solid line), 08:20 UTC (dotted line) and 08:26 UTC (dashed line) are compared with the total aerosol particle number concentration derived by TSMPS in the diameter range of 7 and 12 nm at 07:50 UTC, 08:10 UTC, 08:30 UTC and 08:50 UTC. All data were derived during the MelCol experiment on June 21, 2015.

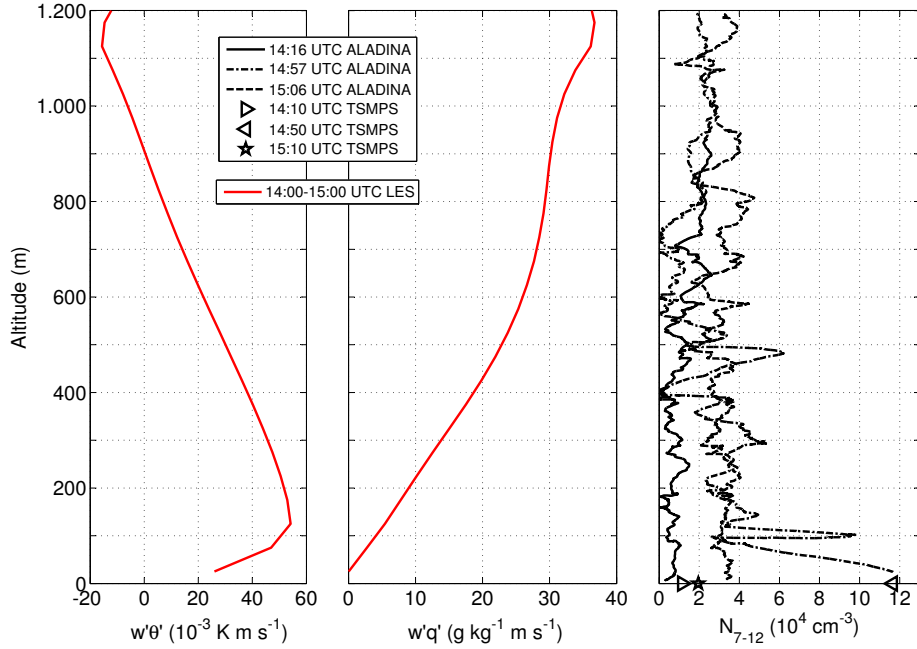


Figure 10. Vertical profiles of sensible heat flux $w'\theta'$ and latent heat flux $w'q'$ obtained by LES output in the interval of 14:00–15:00 UTC (red lines). Besides, three vertical profiles of freshly formed boundary aerosol in the particle diameter between 7 and 12 nm (N_{7-12}) measured with the CPCs on the UAS ALADINA at 14:16 UTC (solid line), 14:57 UTC (dotted line) and 15:06 UTC (dashed line) are compared with the total aerosol particle number concentration derived by TSMPS in the diameter range of 7 and 12 nm at 14:10 UTC, 14:50 UTC and 15:10 UTC. All data were derived during the MelCol experiment on June 21, 2015.

Table 1. Airborne measurements performed with the UAS ALADINA during Case I on 4 April, 2014. The flight time of ALADINA between take-off and landing and the maximum altitude of flights. In addition, prevailing conditions of clouds, SO₂ gas concentration, pressure at surface p_0 and temperature T_{1m} from mast in 1 m height are presented in relation to the corresponding flights.

Flight	Flight time (UTC)	Max. height (m)	Sky conditions	SO ₂ ($\mu\text{g m}^{-3}$)	p_0 (hPa)	T_{1m} ($^{\circ}\text{C}$)
1	06:15-06:50	950	StCu	1.7–2.4	998	7.7–8.8
2	07:50-08:17	700	ABL clouds	1.1–2.2	999	5.3–6.1
3	08:55-09:30	750	ABL clouds	1.9–2.9	999	7.0–9.1
4	10:41-11:28	1000	ABL clouds	3.7–4.6	1000	10.8–11.3
5	11:45-12:27	950	ABL clouds–StCu	2.7–4.4	1000	10.8–12.1
6	13:20-13:58	950	StCu	3.6–5.0	1001	11.8–12.3

Table 2. Performed airborne measurements with the UAS ALADINA during MelCol experiment Case II on June 21, 2015. The flight time of ALADINA between take-off and landing and the maximum altitude of flights. In addition, prevailing conditions of clouds, SO₂ gas concentration, pressure at surface p_0 and temperature T_{1m} from mast in 1 m height are presented in relation to corresponding flight time series.

Flight	Flight time (UTC)	Max. height (m)	Sky conditions	SO ₂ ($\mu\text{g m}^{-3}$)	p_0 (hPa)	T_{1m} ($^{\circ}\text{C}$)
1	08:00-08:35	950	ABL clouds–StCu	1.2–1.6	1007	14.8–15.9
2	08:46-09:15	950	StCu	0.9–1.2	1007	15.4–17.4
3	09:28-10:01	950	StCu	0.8–0.9	1007	14.7–17.4
4	12:03-12:38	1100	StCu	1.2–1.5	1007	15.7–17.1
5	12:51-13:15	1200	StCu	1.2–1.3	1006	15.7–16.3
6	13:51-14:23	1200	StCu	1.4–1.5	1005	15.6–18.3
7	14:57-15:32	1200	StCu	1.4–3.7	1005	16.9–17.7