Final revision of J. Größ et al.: Evolution of gaseous precursors and meteorological parameters during new particle formation events in the Central European boundary layer, ACP manuscript acp-2014-990

General Remarks

We deeply apologise for the delays that have occurred with the production of this final manuscript. Furthermore, we thank the Editor very much for giving us the opportunity to finish this task, after all.

This file documents, for the editor, the changes that we made in the final manuscript in response to Anonymous Referees #1 and #2. Since some time has passed since the reviews were published on the ACPD website, we made a few updates to the text, which are also highlighted in red in the manuscript.

We hope that this version satisfies the criteria for a final publication in ACP and remain very grateful to the Editor, the Anonymous Referees, and the ACP production staff.

Changes made in response to Anonymous Referee #1

Referee #1: The authors develop an automated method to classify the strength of NPF (unfortunately with no explicit influence of growth in the classification) ...

The focus of this work has been to examine the circumstances of fresh particle formation (i.e. focussing on diameters less than 20 nm). The importance of the production of Aitken particles, cloud condensation nuclei (CCN) and optically active particles has not escaped us, but would require a much more extended analysis. This work makes use of the advantage of the NAIS instrument, which provides readings of particles down to 2 nm. This is now clarified in a revamped Section 4.1.

Referee #1: The writing is often sloppy (e.g. comma usage) and could use additionally proofreading.

The article has now been proofread by several authors again. Although there was no native English speaker among us, we have done the best to make the article more readable. We hope that the current version satisfies the standards set by the Editor and the Referees.

Referee #1: I have specific scientific and writing comments below that should be addressed before publication in ACP.

P2309 L12: I'm not sure how large-scale atmospheric models currently pa-

rameterize growth.

Some sentences were added to the Introduction for clarity: "For computational reasons, large-scale atmospheric models usually depend on parameterizations of particle nucleation and growth processes (Spracklen et al., 2010). Due to limitations of computing power, global chemistry transport models can treat aerosol particle growth due to condensation of organic precursors only in highly simplified form. [...] Overall, the degree to which particle nucleation is actually able to influence the budget of CCNs and thus terrestrial climate has to be considered highly uncertain (Kerminen et al., 2012; Westervelt et al., 2014)." These are highlighted in red in the text.

Referee #1: P2309 L17: Percentages are fractions, not frequencies.

This has been corrected accordingly.

Referee #1: P2311 L6 vs. L2312 L23: is the lower limit of the APS 0.5 or 0.8 microns?

This issue is now clarified in Sect. 2.2.3 in form of an additional footnote.

Referee #1: P2314 L20: Shouldn't the units for A be m2 W-1 cm3 in order to get the concentration of OH correct?

This issue in Sect. 2.3 is now clarified. The correct value 6110 $m^2\,W^{-1}\,cm^{-3}$ (cf. Fig. 1) has been inserted as well.

*Referee #1: P2316 L18-19: *By definition* H2SO4 production depends equally on [OH] and [SO2] (k[OH][SO2] for the rate-limiting step), it's just that here [OH] varies more. Please be more accurate in this description.*

This has been reformulated as "This case is an example where variations in the production rate of H2SO4 correlate with the variations in OH while the concentrations in SO2 remain almost constant." in the text.

Referee #1: P2317 L14: "maxima" should be "maximum" here (not plural).

This has been corrected.

Referee #1: P2319 L9: I'm guessing that g(tau) should be g(t) since you're defining "g" in a general sense (as a function of time).

This has been corrected as well.

Referee #1: L12: The event in 2a doesn't grow to sizes close to 100 nm.

This number was checked by lognormal fitting, yielding a nucleation mode diameter of 70 nm at midnight and 90 nm on the next day (not shown in the Figure). These diameters are now mentioned in the text.

Referee #1: L23-24: How does the event in 2d do in the filter? It extends for a long time but does not grow. It seems like these events could have strong responses from the filter too, but aren't necessarily the events you want to find.

The Referee pointed out correctly that there are different types of NPF events. Identifying and classifying NPF events can be done with two major intentions in mind: (1) Examining the circumstances and possible causes of fresh particle formation and (2) Evaluating the potential of NPF events to deliver total particle number concentration, CCN number concentration, and radiative forcing effects. The main objective in this paper is to examine aspect (1). This issue is now better explained in a completely rewritten Sect. 4.1.

Referee #1: P2324 L15-17: You bring up the turbulence and flux here but you haven't introduced Figure 5 yet.

Figure 5 is now introduced earlier.

Referee #1: Figures 4 and 5. What are the sigma values on the plot. They are never defined or discussed, I [...] believe.

This is now explained in the Figure captions as "The σ values, indicated by whiskers, represent the standard deviation of the mean for each subpopulation.". The purpose of the sigma values is to obtain a feeling for the significance of the difference that the shown atmospheric parameters exhibit between the three event classes. They correspond to the whiskers depicted in the plot. Technically the whisker interval represents the range in which the true mean value can be expected with 66% probability.

Referee #1: P2324 L22-24 and P2325 L6-8: Why can't the high CS overnight be due to primary emissions into a shallow night time BL that?

We have checked that issue carefully. In the surroundings of Melpitz, primary particles may be emitted by traffic sources. We found that that black carbon (BC), which is representative for such primary particle emissions in the surroundings of Melpitz, makes up less than 10% of the total particle mass at Melpitz (cf. our extended reply to Referee #1 in the discussion forum). We conclude that this is not sufficient to account for the diurnal effect in CS. As an effect, we weakened our original statement about the importance of CS for new particle formation. Also, a new paragraph was added in the corresponding Sect. 5.1, paragraph "First indications of NPF event (-3 h)".

Referee #1: P2326 L9: How can something be proportional to a class?

This text was reformulated, to "[...] rise with ascending event class".

Referee #1: P2326 L22-23: Did you check that the small particles (Dp<20 nm) actually don't contribute significantly to the CS? Sometimes they can if the nucleation event is strong and the background is clean.

This has been thoroughly checked (cf. our extended reply to Referee #1 in the discussion forum). We revised the text as follows: "CS correlates most strongly with the number of bigger particles, i.e. in the Aitken and accumulation mode. It is our interpretation that in these cases, CS originates from the same or similar pollution sources that emit SO2. The newly formed particles < 20 nm contribute only little to CS, at most 15% during event peak time for event class I, and much less outside that period."

Referee #1: P2326 L26 and other places: What do you mean by "mean levels"? Mean for the day for each class individually? Mean across all three classes?

This is now clarified to "...decrease to their pre-event levels within a matter of a few hours ...".

Referee #1: Section 5.3: I'm very confused by this section. Where are the results of these tests? Are these the sigma values in the figures? What do they mean? Why no discussion?

Sorry for this oversight from our side. We have completely rewritten Sect. 5.4 that now clarifies the matter.

Referee #1: Section 5.4: I have concerns with using only the 2-3nm bin for determining J2. (1) If particles are formed above and brought down through mixing, couldn't the particles be larger than 2-3nm before reaching the surface. Events being first observed at sizes larger than 3 nm are not uncommon at some sites. (2) Are the counting statistics good enough for a single bin (esp. the smallest one)?

We answered this question in our reply to Referee #1 in the discussion forum. Question (1) is adressed in a new section 5.3 "Reasons for the different peak times in N2-20". Issue (2) is addressed by a new sentence in Sect. 5.4.

Referee #1: Section 5.4 second paragraph: What to take from this? What might a correlation of N with [H2SO4] but not N with J2 mean?

We entirely agree that the presentation of the matter has been confusing in the ACPD version of the manuscript. As a solution we conceived a new Section 5.3 "Reasons for the different peak times in N2-20" and a new Discussions subsection "Where does nucleation take place?". These sections also refer to new experimental evidence on the spatial evolution of NPF events from airborne measurements (Platis et al., 2016).

*Referee #1: P2330 L6: Similar to a comment as before... CS is *by definition* important for [H2SO4]; however, it's the lack of CS variability (relative to [SO2] and [OH]) that you observe.*

Again, this is now clarified in the text.

Referee #1: P2330 L18: What do you mean by "larger-scale particles"? Larger-diameter particles or particles observed on a regional scale?

We meant "larger particles (10-20 nm)", which was also amended in the text.

Changes made in response to Anonymous Referee #2

Referee #2: SPECIFIC COMMENTS. To help the reader to understand results, my suggestion is to better explain the physical meaning of the convolution integral used here. (Is it similar to a cross-correlation between PNSDs and the selected 27 NPF events?)

In the light of these remarks, Sect. 4 was expanded, and subdividied into three subsections 4.1, 4.2 and 4.3.

In particular response to the question, we now explain in Sect. 4.2: "The motivation behind the Convolution Integral (CI) Method is to enable automatic detection and classification of the NPF events. [...] The CI method will even our such deficiencies in that it yields a standardised CI function, on a regular time grid, which can be compared, for example, among different sites."

Referee #2: I also suggest to explain: (i) how results rely on the manual selection of the 27 NPF events, (ii) how the CI thresholds were selected (Table 2), ...

We added the following explanation to Sect. 4.2: "The 27 NPF events were selected to provide a realistic initialisation to the CI method. [...] Such a choice would push NPF events with higher peak $N_{[2;20]}$ concentrations (even if only short-lived) higher in the ranking."

Referee #2: (iii) reasons for the different average time of peak N2-20 for the three classes (Table 2).

As a solution we conceived a new Section 5.3 "Reasons for the different peak times in N2-20" and a new Discussions subsection "Where does nucleation take place?". These sections also refer to new experimental evidence on the spatial evolution of NPF events from newly acquired experimental evidence from airborne measurements (Platis et al., 2016).

Referee #2: To reinforce findings, it would be worth to discuss how depen-

dant results are on the observation site (Melpitz), or conversely how they can be considered as general findings. For instance, a large dependence of NPF events on solar radiation and [SO2] was found: can this be considered a general finding or a result specific of the Melpitz station (due to local availability of [OH], relative humidity, H2SO4 parameterization)? Also, both the condensational sink (as a factor inhibiting NPF events) and [NH3] (as a precursor of particle nucleation) were found to have a subordinate role: is that a general finding or a finding due to the low road traffic emissions and available agricultural emissions, respectively, at the Melpitz station?

We agree that this aspect has been a shortcoming in the ACPD paper. To answer the Referee's question, we compared our work more specifically with a number of previous studies. As a result, the Discussions section has been expanded and re-organised. A new subsection 6.2 "Comparison with findings worldwide" addresses the questions raised above.

Referee #2: TECHNICAL CORRECTIONS: - Caption fig.4: is "time series" correct?

This has been corrected to "diurnal cycles".

Referee #2: pag.21 line 3: there is an "and" missing after "solar radiation", and pag.24 line 1: there is an "and" missing between "radiation" and "[OH]".

This has been corrected.

- Figure 4: I would better explain the panel f of [NH3].

This has been corrected, pointing out that a constant value of NH3 was used.

- Title: I suggest some modification to clearly reflect the contents of the paper.

We have come up with a new title, "Atmospheric new particle formation at the research station Melpitz, Germany: Connection with gaseous precursors and meteorological parameters".

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