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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Some insights into the condensing vapors driving new particle growth to CCN sizes on the basis of hygroscopicity measurements" *by* Z. J. Wu et al.

Anonymous Referee #2

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The paper presents a case study of three consecutive new particle formation events in a regional background station in Europe. Hygroscopicity measurements and AMS data are used to draw conclusions about the contributions of sulfuric acid and organic vapors to the growth of the newly formed particles to CCN sizes. CCN concentrations are calculated from size distributions and chemical composition data. The paper combines very useful measurements and calculations, but could make better use of its data and should be more rigorous in the presentation of the analyses. The following issues should be addressed before publication:

Major comments:

1) No particle sources other than the NPF events seem to have been taken into consideration in the analysis. Melpitz is not a pristine environment, and even if the possible contribution of local sources can be excluded during the three-day period of observation (the paper does not state how or if this was done), wind direction can still change aerosol characteristics substantially within the time it takes for newly formed particles to grow to CCN sizes. How do the authors know that the results of their calculations pertain to aerosol particles specifically originating in the nucleation events when many hours have passed, as is the case for the calculated increases in CCN concentrations in section 4.3? How do the authors account for meteorological conditions, such as concentration/dilution due to boundary layer dynamics, when observing concentration changes (such as CCN concentrations) ?

2) It seems to me that the available data should be exploited better. For example, size-resolved AMS data are available, but, as far as I can tell, are only made use of in the calculation of D(crit). In describing changes in the organic fraction during the NPF events, instead of using PM1, the size-resolved AMS data could be used to more accurately represent chemical composition of the smaller particle size range, thereby reducing the potential dominance of large particles (possibly from sources other than NPF) in the mass concentrations. Also, with those size-resolved AMS data available, why is all hygroscopicity and all condensational growth reduced to sulfate and organics? With HTDMA data available, it may be possible to derive a kappa(HTDMA), and use that for the calculation of the expected CCN concentrations – was there a specific reason the authors decided to use only the AMS chemistry + size distribution data?

3) There is important information missing, such as: What were the kappa(i) values used that entered the kappa(chem) calculation? How and over what time period was GR(obs) calculated? How were the hygroscopic growth factors derived from the HTDMA data? The description of the results is also missing information: Increases/decreases are described without giving the time period over which increases/decreases were observed, or how large the increases/decreases were. How

are the beginning, end, and duration of an NPF event defined? Whe stating that something was observed "during the events", what exact time frame does that refer to? Also, for which exact points during the particle growth were the soluble fractions calculated?

Comments on the writing:

1) There are multiple language errors, which should be corrected (singular/plural errors, missing/superfluous articles, wrong punctuations), as they can hamper the understanding of the material, e.g.: p. 8405, line 26 “sulfuric acid and organics, which fraction is the leading component in particles. . .”, p.8406, line “measurements during a NPF events”, p. 8406: “in the polluted areas, Atlanta, USA”, p. 8413 “took place from, 5 to 7 June 2008, “.

2) There are many references in the text that are missing in the list of references, e.g. p. 8408, lines 21-23.

Specific comments by section:

Introduction:

p. 8405, line 8: “the single largest uncertainty” of what? In the statement by Kerminen et al. that is most likely cited here, nucleation is not stated to be “the single largest uncertainty” in the CCN budget.

p. 8405, line 9: “radioactive” should be "radiative"

p. 8406, line 6: what are “potential chemical species”?

Experiments:

Perhaps the title “Measurements”, or “Experimental” would be better, as “Experiments” could raise the expectation of a controlled laboratory setup.

p. 8406, line 15: Could the authors provide a little more information on the full dataset that this case study is a part of, and specifically, state why were those exact three days chosen? How many events were observed in the full dataset? Please also describe the origin of SO2 and Rad, used later in this study.

Section 2.1:

The HTDMA measurements should be described better: which dry sizes and RH's were chosen? What was the sampling schedule for the chosen parameters? How were the growth factors determined from the size distribution of DMA2?

p. 8407, line 2: "described" is better than "illustrated"

Section 2.2:

Please provide a lower particle size detection limit of the AMS.

p. 8407, line 24: "typically": Since the investigated dataset is only three days, could the authors give more detail here?

p. 8408, line 8: How was the density value chosen?

p. 8409, line 3: "two groups including soluble and insoluble fractions" – this is confusing. How many groups or fractions were there?

p. 8409, line 13 and following: Should the assumption of an insoluble organic fraction not underestimate the actual soluble fraction of the particle?

p. 8409, line 17: It would be helpful if the term "equivalent soluble fraction" was used consistently from this point onward.

Section 3.2:

p. 8410, line 9: The size ranges of the AMS measurements, the chemical species used in the calculation, as well as the values used for their respective kappa(i) should be stated here.

p. 8410, line 15: "are activated", not "were activated"

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p. 8410, line 16: Please state which critical supersaturations were chosen for the calculation.

p. 8410, line 19: Integrating the size distribution from D(crit) upwards: The implicit assumption of an internal mixture of the determined (bulk) chemical composition should be stated explicitly here.

p. 8410, line 22 and following: This discussion is confusing in its current location in the section: “The critical diameters” (p. 8411, line 1) could be interpreted as the critical diameters in this study, calculated from eq. 5, rather than a general statement on the approximate range of critical diameters, which, I believe, is what the authors mean. This description of the chosen AMS size ranges should be moved up, as stated in the initial comment on this section.

Section 3.4:

What were the time frames (and particle diameter ranges) that were actually used for the determination of the growth rates in this study?

p. 8412, line 19: Weren’t the H2SO4 concentrations calculated, rather than measured?

Results:

p. 8413, line 9: How was the particle formation rate calculated?

p. 8413, line 18: average over what? What was the time frame of the GR determination? It would be helpful is “GR(obs)” were used consistently.

p. 8413, line 20: define “particle formation period”

p. 8413, line 21: “This was because...”: Has causality been established?

p. 8413, line 23: “hygroscopic” would be better than “water-soluble”

p. 8414, line 2: The processes were not observed directly. Rather, they can be inferred (to a degree) by the measurements. This should be made clear.

p. 8414, line 4: “Significant” should only be used when statistical significance is established.

p. 8414, line 6: Please substantiate the statement: “sulfuric acid condensation played a minor role in particle growth”? And what is the time period?

p. 8414, lines 6 – 10: “Lower temperature facilitates [. . .] leading to an evident decline of hygroscopicity.” It seems to me that the initial observation was the decrease in hygroscopicity and the increase in organic mass fraction, and the condensation of semi-volatile compounds is a possible reason, rather than the reverse: stating a plausible process (condensation of semi-volatiles) and treating the observations as “confirming” it.

p. 8414, lines 9 and following: “when these particles grew to. . .” How were the hygroscopicity measurements at the different particle sizes attributed to the newly formed (rather than pre-existing) particles? How was the growth traced, and which exact hygroscopicity masurements were taken at which point in the growth process?

p. 8414, line 17 and following: Would the size-resolved AMS chemistry data not be more helpful here, rather than the bulk organic fraction, which surely is more sensitive to larger particles?

p. 8414, line 20: “considering that the chemical species contributing to particle growth was similar to. . .”: This is an assumption and should be stated as such.

p. 8414, line 18: please define “later stage”

Section 4.3:

p. 8415, line 9: see comment above on significance

p. 8415, line 12: “. . ...step-wisely enhanced due to the accumulation processing” It is not completely clear to me what is meant by that.

p. 8415, line 17 “same time period before the end of the event”: please define “end of event”

p. 8416, lines 22 – 24: Please substantiate the statement on BVOCs.

p. 8417, line 14: “The observation showed that. . ..” This does not follow from the observations presented in this study.

Comments on the Tables and Figures:

Table 1: What measurement points or what averages over which time frames are shown here?

Figure 1: The figure is overloaded. It is vital to be able to read details out of Figure 1 to understand the text, but everything is very small. My suggestion would be to make three Figures (total): one with size distributions, critical diameter, and CCN concentrations, a second one with chemical composition and hygroscopic growth factors, and a third with the auxiliary meteorological and gas-phase data.

a) The plot should not be this saturated (in both the upper and the lower concentration limit); a lot of detail is lost. A logarithmic concentration scale would help tremendously. What are the white circles? The diameter axis should have minor ticks (numerous references to specific particle sizes in the text). The color bar label is in an odd place. b) What is the unit of H2SO4? c) The RH axis should not extend to 120%.

Figure 2: It would be helpful if the beginning and end of the nucleation event were marked in the plot, in addition to t1 and t2.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 8403, 2015.

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