

Interactive comment on “Reconciliation of measurements of hygroscopic growth and critical supersaturation of aerosol particles in Southwest Germany” by M. Irwin et al.

Anonymous Referee #1

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General Comments

This paper describes hygroscopic growth factors and CCN activities of atmospheric aerosol particles at the Hornisgrinde mountain site in Germany. Critical supersaturations and other variables related to water uptake were predicted from hygroscopic growth factors under subsaturated water vapor conditions, and were compared to those derived from CCN measurements. These comparisons were made using the κ -Köhler model. Substantial differences between the “potential CDNC” predicted from hygroscopic growth factors and those predicted from CCN data are reported in this paper. Because the use of the κ -Köhler model has become a common approach to describe

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the hygroscopicity and CCN activation of particles, assessment of the κ -Köhler model based on field observations, as conducted in this study, is highly valuable. The topic of this study is within the focus of Atmospheric Chemistry and Physics.

While the finding of this study is very interesting, this manuscript needs substantial revision. This is in part because the experimental and analytical procedures are not explained sufficiently. For example, definitions of key variables, e.g., S_{c,D_0} , are not provided explicitly. Furthermore, I could not find references that are probably important to understand the details of the procedures. There are also some other issues that need to be revised. For example, the cloud and non-cloud periods are not consistently assigned throughout the paper. The authors present different degree of agreement in the closure of potential CDNC, yet this variability depending of the calculation procedures is not addressed adequately in the discussion of Figs. 4 and 5. Possible measurement biases and the reasons for the disagreements presented in Figs. 4, 5 and 7 need more discussion.

I recommend publication of this paper only after the concerns raised by this review are adequately addressed. More specific comments and technical corrections are listed below.

Specific Comments

Abstract: Because the AMS-derived organic:sulfate ratio is used for analyses and discussion repeatedly, the AMS measurement should be addressed more in the abstract.

Experimental Methodology Section: I do not find two references that seem be important to understand the methodology. Jones et al. (2010) is missing in the reference list. I could not find Irwin et al. (2010) although it is in the reference list.

Page 17077, line 24: The definition of “dry” condition in this study should be provided.

Page 17080, lines 1–4: The definition and the calculation procedure of S_{c,D_0} is not clear, which should be provided more explicitly.

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Page 17080, line 17: It seems that $\kappa_{S_{c,D_0}}$ is the κ derived from S_{c,D_0} . This definition of $\kappa_{S_{c,D_0}}$ should be given more clearly.

Page 17080, lines 28–29: It is not clear which variables were affected only negligibly by multiple charging.

Page 17082, line 14: The definition of $\kappa_{(GF_{D_0})}$ is not clearly given. The parenthesis in $\kappa_{(GF_{D_0})}$ does not seem to be necessary.

Results and analysis section: The measurement period, and the cloud and non-cloud periods are not defined consistently throughout the paper. Fig. S2 is from 25 June, although the measurement seems to have started earlier according to the text (P17083, L15). While the authors explain that a distinct cloud event was observed on 30 June (P17083, L16-17), the decrease in RH seems to have been greater on 1 July. The boundaries of cloud and non-cloud periods in Figs. 1–3 are different from those defined in Fig. S2. Moreover, the end of CP2 in Fig. 2 is different from those in Figs. 1 and 3.

Section 3.2: Possible presence of refractory components, which are not detectable by the AMS, and its influence on the comparison of the AMS data to the hygroscopicity and the CCN activity should be addressed.

Page 17083, lines 24–25: This sentence is written as if activation of accumulation mode particles is a fact. However, it seems to be just an inference by the authors.

Page 17084, lines 12–14: The mean organic:sulfate ratios in the cloud and non-cloud periods should be given, because the difference is not clear in Figure S2.

Page 17088, lines 10–17: The model here seems to assume $\sigma = 72.5 \text{ mNm}^{-1}$. This point should be explained more clearly.

Page 17089, lines 20–24: Does this sentence mean that the variations in the hygroscopicity of organics and the surface tension were more important than the variation in the organic:sulfate ratios? More explanation may be necessary.

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Page 17089, lines 25–29: Isn't there any possibility that the hygroscopic growth of particles was not in the equilibrium in the HTDMA, which resulted in the underestimation of $\kappa_{GF_{D_0,RH,mean,corr}}$?

Page 17090, lines 11–14: As pointed out by a posted comment, the term “potential CDNC” is misleading and should be reworded.

Page 17090, line 15 – Page 17091, line 23: The explanation in the paragraphs is difficult to follow because the definitions and the calculation procedures of the variables are not explained adequately. More detailed explanation should be added.

Discussion section: Possible reasons for the disagreements presented in Figs. 4, 5, and 7 (e.g., a possible presence of slightly-soluble materials) should be discussed more, in light of the simplifications assumed in the κ -Köhler model.

Page 17091, lines 25–27: Is this statement based on the result in Fig. 7? If the degree of the agreement depends on the analytical methods for the CCNc data, how does the difference affect the discussion in this paper? For example, results in Fig. 5 is based on $\kappa_{S_{c,D_0}}$, with which the closure of potential CDNC in Fig. 7 seems to show poorer agreement than those from other approaches.

Page 17093, lines 1–6: The point of the discussion in the two sentences is not very clear.

Page 17093, lines 27–29: I do not see the point of this explanation. Because the AMS-derived composition is governed by the composition of relatively large particles, the fact that the hygroscopicity of small particles (<127 nm) did not correlate with the AMS-derived organic:sulfate ratio is not contradictory. In the case of relatively large particles ($\geq 127 \text{ nm}$), some relationship between the HTDMA data and the organic:sulfate ratio (Fig 6a) was seen.

Page 17095, line 25 – Page 17096, line 2: I do not understand the explanation in the sentences. The variables mentioned here should be given more clearly.

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Figs. 1-3: Some data seem to be omitted. What are the criteria of the data screening?

Fig. 7: The blue marker whose vertical axis value is about 20 nm in Figs. 7a and 7b is missing in Fig. 7c.

Fig. S2: Why were the aerosol number concentrations very high on 27 June? Is there any possibility of the failure of the measurement?

Technical Corrections

Page 17081, line 22: "wasmeasured" → "was measured"

Page 17082, line 11: " $GF_{D,RH}$ " → " $GF_{D0,RH}$ "

Page 17087, line 27: Should " $20 \text{ nm} \leq D_0 \leq 200 \text{ nm}$ " be changed to " $20 \text{ nm} \leq D_0 \leq 200 \text{ nm}?$ "

Page 17087, line 27: "Fig. 4)" → "Fig. 4"

Page 17095, line 21: "instrument instrument" → "instrument"

Tables 2 and 3: The expression of κ with parenthesis in the suffix is different from the expression in the text.

Fig. 1: Because both the average growth factor and the upper end of the normalized counts are drawn in red, they are not distinguishable.

Fig. 1: A space should be added before "nm."

Fig. 5: A space should be added before "nm." The letter k in the axis labels should be corrected to κ . The expression of the horizontal axis label (with parenthesis) is different from that in other parts of the manuscript.

Fig. 7: The letter k in the axis labels should be corrected to κ . The suffix of κ (k) in Fig. 7b and that of S in Fig. 7c are different from those in the text.

Figs. S1-S3: A space should be added before "m," "m/s," and "nm."

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Fig. S4: The unit of "1.92" in the annotation should be added. Two alternate expressions, " S_{c,D_0} " and " $S_{c(D_0)}$," seem to be used for the same variable.

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