

Dear Editor,

Your continuous patience and invaluable assistance throughout the review process are deeply appreciated. We would like to express our gratitude for the comments provided by the reviewer. We have revised our manuscript to address these comments and improve the overall quality of our work.

Your sincerely,

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Response to comments:

1. *In the revised manuscript, the figures in the main text exclusively present NCCN and kappa values without accounting for the deviation of the SS calibration, which is referred to as "lab-calibrated" in the manuscript. Conversely, NCCN and kappa values that take into consideration the deviation of the SS calibration, referred to as "approximated on-site" in the manuscript, are only depicted in the supplementary material. I believe that this presentation may potentially lead to misconceptions, suggesting that the "lab-calibrated" values are more accurate than the "approximated on-site" values. This is particularly noteworthy when it comes to estimating kappa because in cases where NPF due to the SS in the CCN counter results in overestimation, kappa could be underestimated by approximately 20%. Therefore, I encourage the authors to include both "lab-calibrated" and "approximated on-site" values of NCCN and kappa in the figures within the main text. Doing so would serve as a reminder to readers to exercise caution when interpreting the NCCN and kappa values reported in this study.*

Response: We appreciate the valuable suggestions, which are helpful enhancing the overall quality of our work. In our revision, both "lab-calibrated" and "approximated on-site" values of N_{ccn} and κ values for the case studies have been added into the figures within the manuscript and Supporting Information.

2. *In response to the minor comments from my previous review, the authors asserted that the utilization of Equation (6) depends on a growth factor derived from HTDMA measurements, which were unavailable in this study. They also mentioned that the absence of concurrent HTDMA measurements was the primary reason why most studies rely on Equation (10). However, this argument is incorrect. In Petters and Kreidenweis (2007), it is explicitly stated that this equation can be applied under conditions of cloud droplet activation without any mention of the necessity for a growth factor or HTDMA data: "Equation (6) applies over the entire range of relative humidity and solution hygroscopicity. It can thus be used to predict particle water content in the subsaturated ($S < 1$) regime, as well as to predict the conditions for cloud droplet activation. The critical supersaturation (s_c , where $s_c = S_c - 1$ and is usually expressed as a percentage) for a selected dry diameter of a particle having hygroscopicity κ is computed from the maximum of the κ -Kohler curve (Eq. 6). Figure 1 shows the relationship between dry diameter and critical supersaturation for a range of constant κ values, computed for $\sigma_{s/a} = 0.072 \text{ J m}^{-2}$ and $T = 298.15 \text{ K}$." Moreover, this equation has been employed in other research as well. For instance, in Kerminen et al. (2012), it was referred to as Equation (1) and utilized to calculate the relationship between particle dry size and critical supersaturation, as demonstrated in Figure 1 of that work, without any indication of the need for a growth factor or HTDMA data. I strongly urge the authors to rectify this matter.*

Response: We are sorry that our previous response on this issue was not clear enough and caused your concerns. We try to provide a more comprehensive explanation here:

Eq. (6) in Petters and Kreidenweis (2007) is derived from the Köhler theory, in which the hygroscopicity parameter κ replaces the solution activity. However, Petters and Kreidenweis (2007) didn't use this method for κ calculation. Instead, they employed a simple rule, Eq. (7), to get a range of constant κ values, where κ is defined as $\kappa = \sum \varepsilon_i \kappa_i$. Here, ε_i represents the volume fraction of a certain chemical compound in aerosol and the κ_i represents the hygroscopicity parameter of that compound. When utilizing Eq. (6) to calculate κ values, the inclusion of a wet diameter (D_{wet}) or growth factor is necessary. These parameters need to be obtained from HTDMA measurements, a practice employed in numerous studies (e.g., Kawana et al., 2017; Cerully et al., 2011). Unfortunately, since HTDMA data was not available in our study, we selected for Eq. (10) from Petters and Kreidenweis (2007) for κ value calculations.

Moreover, Fig. 1 in Professor Kerminen's research (Kerminen et al., 2012) shows the relationship between the calculated critical supersaturation (S_c) and dry diameter (D_{dry}), in which κ values serve as constant input values.

In fact, there were three primary approaches to determine κ values in the literature, i.e., 1) Using Eq. (7) as showed in Petters and Kreidenweis (2007); 2) The use of HTDMA measurements; and 3) Employing either directly measured critical diameters at a super saturation (Rose et al., 2010) or estimated ones (in our study).

We hope that our explanation can address your concerns.

References:

- [1] Kerminen, V. M., Paramonov, M., Anttila, T., Riipinen, I., Fountoukis, C., Korhonen, H., Asmi, E., Laakso, L., Lihavainen, H., Swietlicki, E., Svenningsson, B., Asmi, A., Pandis, S. N., Kulmala, M., and Petäjä, T.: *Cloud condensation nuclei production associated with atmospheric nucleation: A synthesis based on existing literature and new results*, *Atmos. Chem. Phys.*, 12, 12037–12059, <https://doi.org/10.5194/acp-12-12037-2012>, 2012.
- [2] Petters, M. D. and Kreidenweis, S. M.: *A single parameter representation of hygroscopic growth and cloud condensation nucleus activity*, *Atmos. Chem. Phys.*, 7, 1961–1971, <https://doi.org/10.5194/acp-7-1961-2007>, 2007.
- [3] Kawana, K., Nakayama, T., Kuba, N., and Mochida, M.: *Hygroscopicity and cloud condensation nucleus activity of forest aerosol particles during summer in Wakayama, Japan*, *J. Geophys. Res.-Atmos.*, 122, 3042–3064, <https://doi.org/10.1002/2016jd025660>, 2017.
- [4] Cerully, K. M., Raatikainen, T., Lance, S., Tkacik, D., Tiitta, P., Petäjä, T., Ehn, M., Kulmala, M., Worsnop, D. R., Laaksonen, A., Smith, J. N., and Nenes, A.: *Aerosol hygroscopicity and CCN activation kinetics in a boreal forest environment during the 2007 EUCAARI campaign*, *Atmos. Chem. Phys.*, 11, 12369–12386, <https://doi.org/10.5194/acp-11-12369-2011>, 2011.

[5] Rose, D., Nowak, A., Achtert, P., Wiedensohler, A., Hu, M., Shao, M., Zhang, Y., Andreae, M. O., and Pöschl, U.: *Cloud condensation nuclei in polluted air and biomass burning smoke near the mega-city Guangzhou, China – Part 1: Size-resolved measurements and implications for the modeling of aerosol particle hygroscopicity and CCN activity*, *Atmos. Chem. Phys.*, 10, 3365–3383, <https://doi.org/10.5194/acp-10-3365-2010>, 2010.