As the Reviewer#2 commented, the authors present a 4 year-long size resolved CCN measurement data at a central European rural background station. This type of data is very valuable for the atmospheric science community. The authors have focused on analyzing the seasonal trends and test multiple approaches to predict the number of activated particles NCCN. Based on the suggestions and comments from reviewer#2, the authors have made corresponding revisions and edits on the manuscript and making the current version of the paper being improved.

Comment 1:

However, I still have some concerns here, since the data are based on a 4-year measurement, I would like to suggest the authors may present a yearly trend or changes of the aerosol CCN activation, which is not shown in the revised manuscript. It would be interesting to study and analyze such trends over a regional scale.

Response: Thanks for your comment. We further investigated the yearly trends of CCN activation properties. The CCN number concentration (N_{CCN}) and hygroscopicity factor calculated from monodisperse CCN measurements (κ_{CCN}) measured at supersaturation (*SS*) of 0.1% and 0.7% are chosen to represent the CCN activation characteristics. The results are shown in the following Figure S7. The yearly trends in N_{CCN} and κ_{CCN} are not significant (without significant increase or decrease trends) during the measurements from August 2012 to October 2016. However, it is interesting to see that the N_{CCN} measured in 2015 was significantly lower than it measured in the other four years. One of the reasons could be that the CCN measurements in 2015 concentrated in summer and autumn, lacking measurements in the spring and winter months (Figure S1). As shown in Figure 3b, N_{CCN} measured at summer and autumn are lower than those measured in spring and winter due to its seasonal trend, causing the lowest N_{CCN} values in 2015. Thus, the CCN measurements in 2015 may not be representative of the CCN characteristics of the whole year. Similarly, the 2012 measurements may not be representative of year-round CCN characteristics because of the lacking spring and summer measurements. Additionally, it is also hard to see the yearly trends using the only 4-year data. In order to investigate the yearly trends of CCN activation characteristics, longer periods measurements are required.

We clarify it in the text and SI as follows.

"Additionally, no significant yearly trends of the CCN activation characteristics are found during the 4-year measurements and the results are provided in SI (Text S2 and Figure S7)." Lines 314 to 316.

"**Text S2. Yearly variations of CCN activation characteristics.**

Yearly trends of CCN activation properties are investigated. The CCN number concentration (N_{CCN}) and hygroscopicity factor calculated from monodisperse CCN measurements (κ_{CCN}) measured at supersaturation (SS) of 0.1% and 0.7% are chosen to represent the CCN activation characteristics. The results are shown in Figure S7. The yearly trends in N_{CCN} and κ_{CCN} are not significant (without significant increase or decrease trends) during the measurements from August 2012 to October 2016. However, it is interesting to see that the N_{CCN} measured in 2015 was significantly lower than it measured in other four years. One of the reasons could be that the CCN measurements in 2015 concentrated in summer and autumn, lacking measurements in the spring and winter months (Figure S1). As shown in Figure 3b, N_{CCN} measured at summer and autumn are lower than those measured in spring and winter due to its seasonal trend, causing the lowest median NCCN values in 2015. Thus, the CCN measurements in 2015 may not be representative of the CCN characteristics of the whole year. Similarly, the 2012 measurements may not be representative of year-round CCN characteristics because of the lacking spring and summer measurements. Additionally, it is also hard to see the yearly trends of CCN activation characteristics using the only 4-year data. In order to investigate the yearly trends of CCN activation characteristics, longer-term measurements are required." in lines of 42 to 60 in SI.

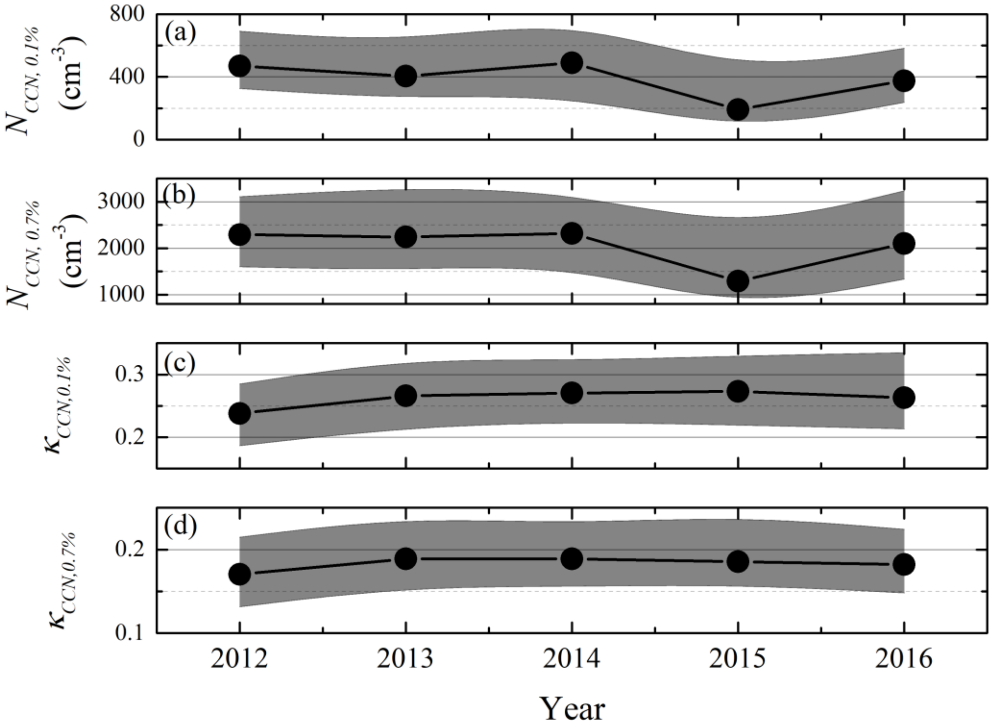

Figure S7. Yearly variations of (a) CCN number concentration (N_{CCN}) at supersaturation (SS) of 0.1% ($N_{CCN,0.1\%}$), (b) N_{CCN} at SS of 0.7% ($N_{CCN,0.7\%}$), (c) hygroscopicity factor calculated from monodisperse CCN measurements (κ_{CCN}) at SS of 0.1% ($\kappa_{\mathrm{CCN},\,0.1\%}$), and (d) κ_{CCN} at SS of 0.7% ($\kappa_{\mathrm{CCN},\,0.7\%}$). Dots represent the median values. Shaded areas represent the values in the range from 25th to 75th percent.

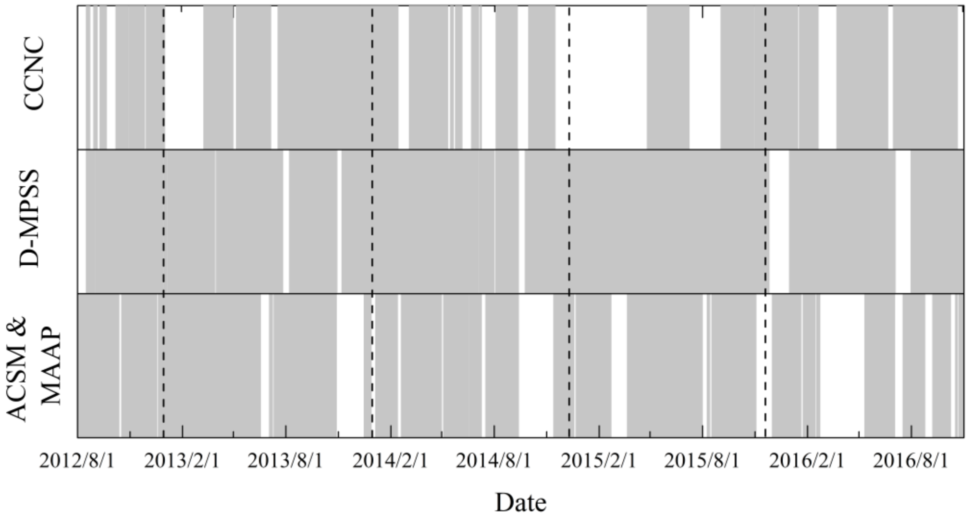

Figure S1. Coverage of the effective data represented by the gray columns during the long-term experiment at Melpitz. CCNC — cloud condensation nuclei counter, D-MPSS — Dual-mobility particle size spectrometer, ACSM — aerosol chemical species monitor, MAAP — multi-angle absorption photometer.

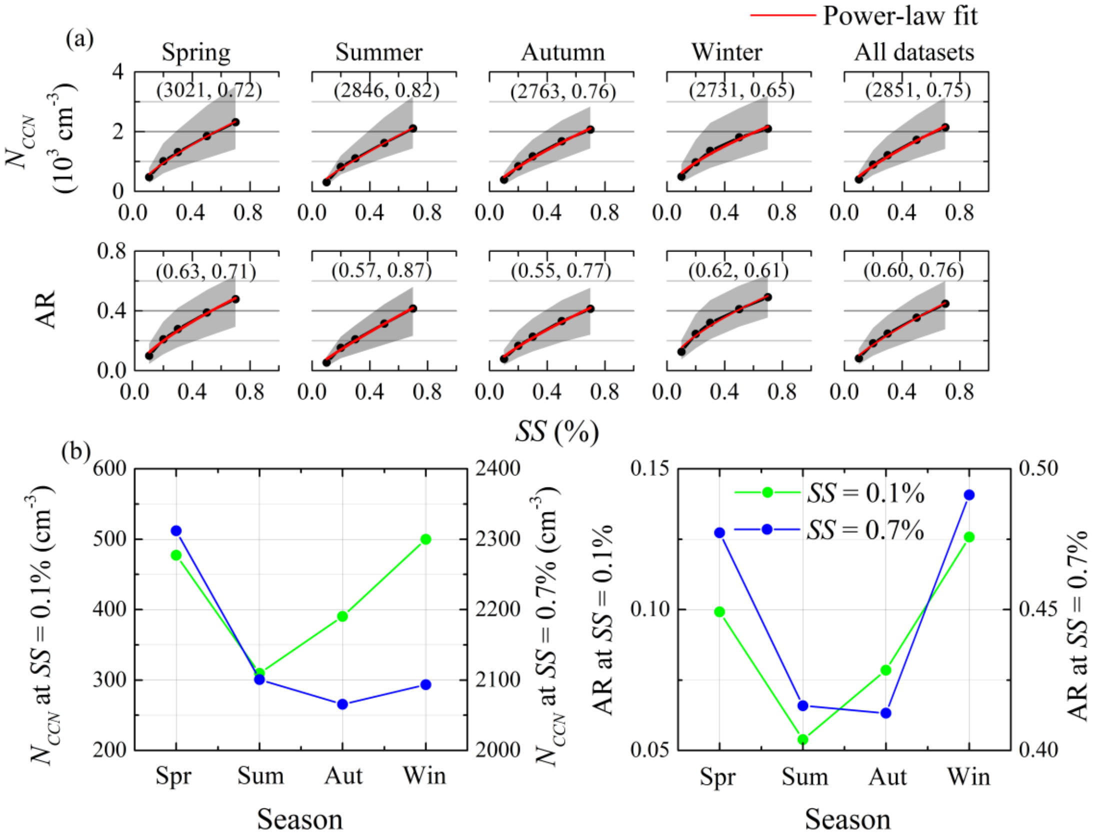

Figure 3. (a) Relationships between CCN number concentration (N_{CCN}) and supersaturation (*SS*), and relationship between activation ratios (AR) and *SS* for different seasons. (b) Seasonal trends of N_{CCN} and AR at *SS* = 0.1% and 0.7%. Dots represent the median values of N_{CCN} and AR. Shaded areas represent the values in the range from 25th to 75th percent. Red lines are power-law fittings for N_{CCN} (and AR) vs. *SS*. Two parameters of the fitting results are shown in brackets.

Comment 2:

In addition, the authors concluded that "…. the kappa - Dp power-law fit measured at Melpitz could be applied to predict NCCN for other rural regions…." I wonder how did they can derive this, and it would be better to include a CCN closure test or if the authors can present a result by applying the kappa - Dp power-law fit measured at Melpitz to predict the NCCN at other rural sites.

Response: Thanks for your suggestion.

For predicting the NCCN using a parameterized kappa (2nd category in Table 3), the real-time particle number size distribution (PNSD) is an input and its effect on the prediction bias is not considered. As shown in Figurer 8, the κ - D_p relationship measured at Melpitz (black line) is similar to that measured at other rural regions with similar κ - D_p power-law fitting results, e.g., the Vavihill station in Sweden (Fors et al., 2011) and the Xinken station in China (Eichler et al., 2008). Thus, we concluded that the kappa - Dp power-law fit measured at Melpitz could be applied to predict NCCN for other rural regions.

Following your suggestion, we conduct a CCN closure test to support the conclusion. However, the data of PNSD and CCN measurements in Vavihill station in Sweden (Fors et al., 2011) and the Xinken station in China (Eichler et al., 2008) are not available. Let's consider it the other way around. If the κ - D_p power-law fitting measured at other rural stations can well predict the NCCN at Melpitz, it also helps to support our conclusion. Thus, we applied the κ - D_p power-law fitting measured at Vavihill and the Xinken (green and purple lines in Figure 8) to predict the NCCN at Melpitz. The results are as shown in Figure S9. Good prediction results were obtained with mean deviations between the predicted and measured NCCN of ~1%.

We clarify it in the text as follows.

"We conducted a CCN closure test to support this conclusion. Due to lacking the data of PNSD and CCN measurements at Vavihill and Xinken stations, we applied the κ - D_p power-law fitting measured at the two rural stations (green and purple lines in Figure 8) to predict the N_{CCN} at Melpitz. Good prediction results were obtained with mean deviations of ~1% (Figure S9 in SI)." Lines 459 to 463.

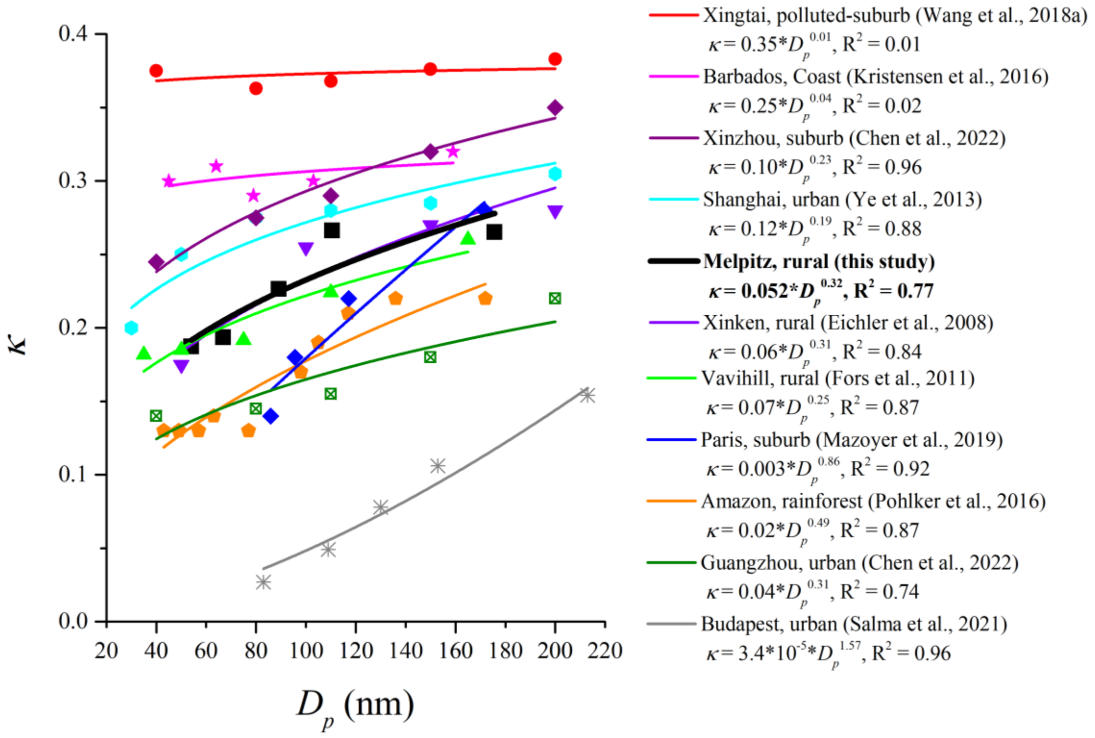

Figure 8. Relationships between the particle hygroscopicity factor (κ) and diameter (D_p) observed at different stations. Lines are power-law fits of κ vs. D_p.

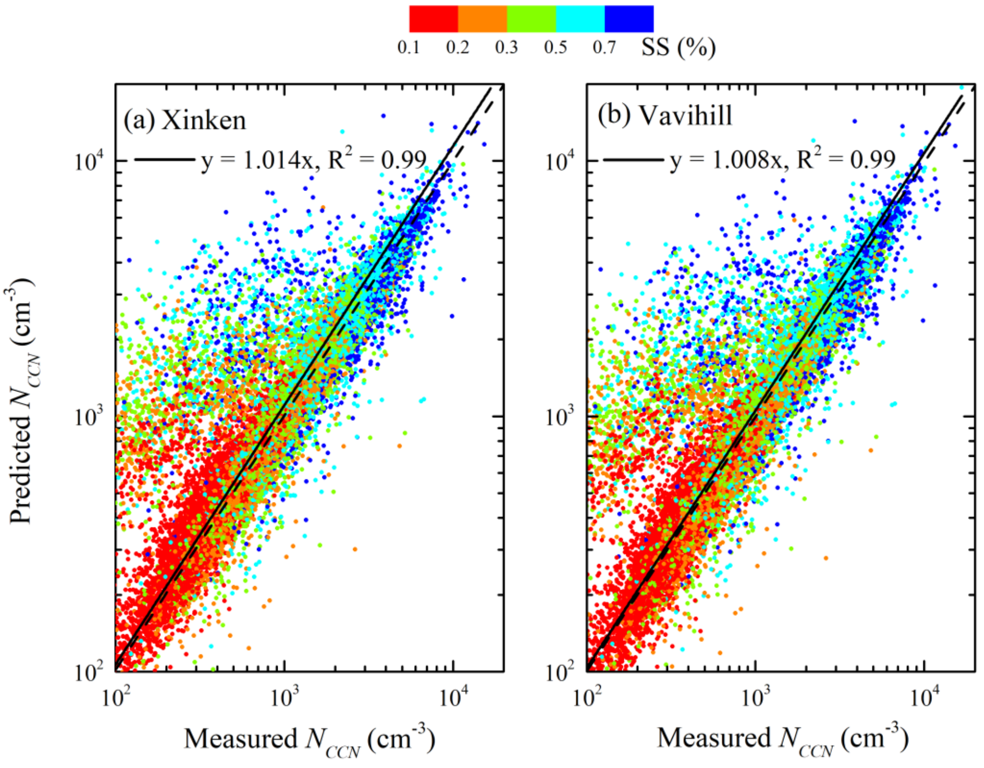

Figure S9. Predicted vs. measured CCN number concentration (N_{CCN}) at Melpitz. (a) using the κ-D_p power-law fitting measured at Xinken station in China (Eichler et al., 2008) to predict the Melpitz

N_{CCN}; (b) using the κ - D_p power-law fitting measured at Vavihill station in Sweden (Fors et al., 2011). Dashed line is the 1:1 line and solid line is the linear fitting.

Comment 3:

In Figure 8, some important references relevant to aerosols hygroscopicity dependence on Dp may be missing (Chen et al., 2022, ACP, 22, 6773–6786, https://doi.org/10.5194/acp-22-6773-2022, 2022). Adding these necessary discussions constitutes major revisions. I recommend publication after these are done, and the remaining issues listed below have been addressed.

Response: Thanks for your suggestion. The κ vs. D_p relationships measured at Guangzhou and Xinzhou stations (Chen et al., 2022) are added in Figure 8, which are in wine red and dark green. We clarify it in the text as follows.

"However, it may cause considerable deviations for different aerosol background regions, e.g., the suburb stations in Xingtai, China (Wang et al., 2018a), Xinzhou, China (Chen et al., 2022), and Paris, France (Mazoyer et al., 2019), the coast of Barbados (Kristensen et al., 2016), the amazon rainforest (Pöhlker et al., 2016), and the urban stations in Budapest, Hungary (Salma et al., 2021), Guangzhou, China (Chen et al., 2022), and Shanghai, China (Ye et al., 2013), because their κ - D_p relationships are different from that measured at Melpitz." Lines 459 to 465.

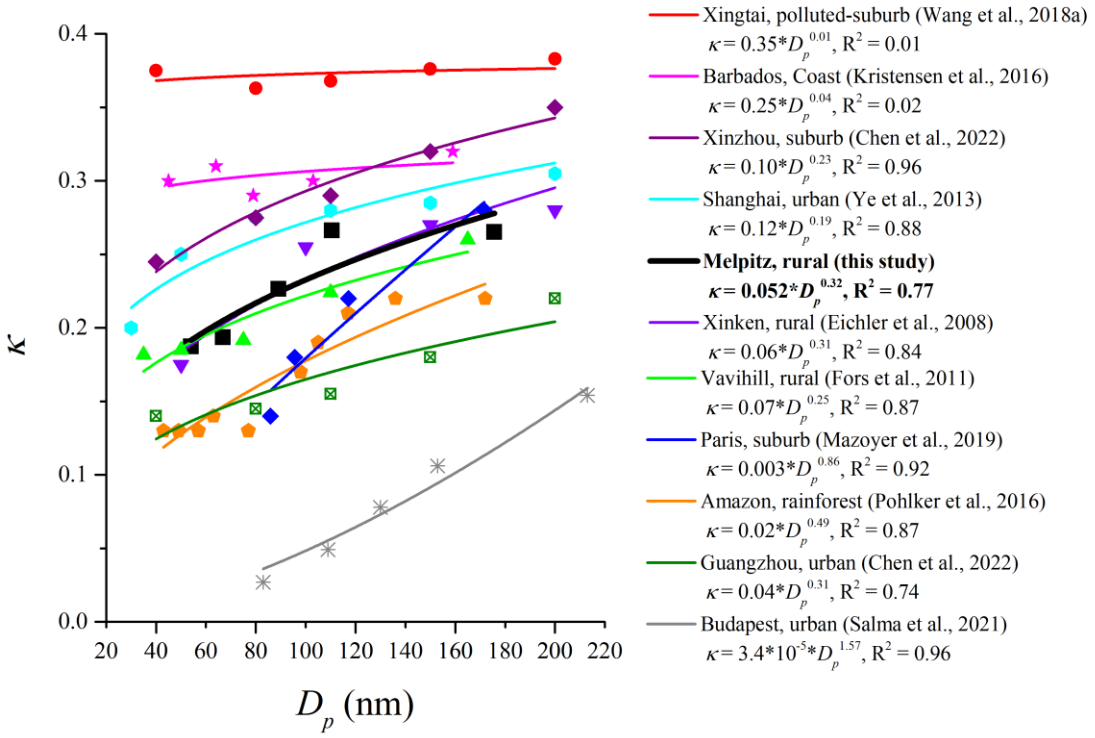

Figure 8. Relationships between the particle hygroscopicity factor (κ) and diameter (D_p) observed at different stations. Lines are power-law fits of κ vs. D_p.

Chen, L., Zhang, F., Zhang, D., Wang, X., Song, W., Liu, J., Ren, J., Jiang, S., Li, X., and Li, Z.: Measurement report: Hygroscopic growth of ambient fine particles measured at five sites in China, Atmos. Chem. Phys., 22, 6773–6786, https://doi.org/10.5194/acp-22-6773-2022, 2022.

Comment 4:

The authors need to present the key findings in the abstract, so as to highlight their main points.

Response: Thanks for your comment. The following four points are what we would like to highlight in abstract:

1) We provide the overall CCN activation characteristics at Melpitz.
2) Aerosol particle activation is highly variable across seasons.
3) Both κ and the mixing state are size dependent.
4) Size-resolved κ improves the N_{CCN} prediction.

In Abstract, to help readers easily see the four points, we highlight these four points by adding the serial number before each point, as follows.

"Understanding aerosol particle activation is essential for evaluating aerosol indirect effects (AIEs) on climate. Long-term measurements of aerosol particle activation help to understand the AIEs and narrow down the uncertainties of AIEs simulation. However, they are still scarce. In this study, more than 4-year aerosol comprehensive measurements were utilized at the central European research station Melpitz, Germany, to gain insight into the aerosol particle activation and provide recommendations on improving the prediction of number concentration of cloud condensation nuclei (CCN, N_{CCN}). (1) The overall CCN activation characteristics at Melpitz is provided. As supersaturation (SS) increases from 0.1% to 0.7%, the median N_{CCN} increases from 399 to 2144 cm^{-3}, which represents 10% to 48% of the total particle number concentration with a diameter range of 10 – 800 nm, while the median hygroscopicity factor (κ) and critical diameter (D_c) decrease from 0.27 to 0.19 and from 176 to 54 nm, respectively. (2) Aerosol particle activation is highly variable across seasons, especially at low SS conditions. At SS = 0.1%, the median N_{CCN} and activation ratio (AR) in winter are 1.6 and 2.3 times higher than the summer values, respectively. (3) Both κ and the mixing state are size dependent. As the particle diameter (D_p) increases, κ increases at D_p of ~40 to 100 nm and almost stays constant at D_p of 100 to 200 nm, whereas the degree of the external mixture keeps decreasing at D_p of ~40 to 200 nm. The relationships of κ vs. D_p and degree of mixing vs. D_p were both fitted well by a power-law function. (4) Size-resolved κ improves the N_{CCN} prediction. We recommend applying the κ - D_p power-law fit for N_{CCN} prediction at Melpitz, which performs better than using the constant κ of 0.3 and the κ derived from particle chemical compositions and much better than using the N_{CCN} (AR) vs. SS relationships. The κ - D_p power-law fit measured at Melpitz could be applied to predict N_{CCN} for other rural regions. For the purpose of improving the prediction of N_{CCN}, long-term monodisperse CCN measurements are still needed to obtain the κ - D_p relationships for different regions and their seasonal variations."