Review of Kim et al. (2011)

Kim et al. present a comprehensive data set of hygroscopicity and CCN activity measurements, which were performed during several field campaigns on two islands of Korea in summer 2006-2009. Parallel measurements of the total aerosol mass concentration and concentrations of several gaseous species indicated that the two islands were under anthropogenic influence from the Asian continent. These data sets allowed classification into pollution and non-pollution days, though no significant differences between those days could be observed for the particle hygroscopicity.

In a closure study, the measured total CCN concentrations were compared with the values predicted by several different methods using the dry particle size distribution and the κ-Köhler model by Petters and Kreidenweis (2007), in which the hygroscopicity parameter κ is derived from either the hygroscopic growth factor or the critical supersaturation. Similar as other earlier studies, Kim et al. found that using size-resolved and temporally varying κ values provided better agreement between predictions and measurements than using a constant average κ of 0.3. They conclude that the hygroscopicity of particles smaller than 100 nm must be known for accurate prediction of the CCN concentration.

The current study yields another important data set of size-resolved hygroscopicity and CCN activity measurements in Asia, which may help to understand how the emission of anthropogenic aerosol particles change cloud microphysical and radiative properties. The results of this study confirm and complement the findings of earlier studies and provide important knowledge on how the CCN activity can be described and implemented in numerical models. The manuscript therefore represents a substantial contribution to scientific questions. It is within the scope of ACP and I recommend its publication after the following questions, comments and suggestions for correction/improvement have been addressed.

General comments:

1) The closure study was performed only for the Gosan 2008 campaign since this was the only one with dry particle size distribution measurements. Your HTDMA results show increasing hygroscopicity with particle size (e.g., Tab. 2), but your CCN results show the opposite (e.g., Fig. 11, and 8; larger κ for 50 nm than for 100 nm). Also, an earlier study by Kuwata et al. (2008) presented slightly increasing κ with increasing size and generally higher κ for the same location (Tab. 3). I am wondering if this discrepancy might be due to an artifact in the actual supersaturation of your CCN counter. As you write in Sect. 2.1, the CCN counter was calibrated with ammonium sulfate or sodium chloride using the Köhler model denoted as AA1 in Rose et al. (2008). As shown in Fig. 11 of that paper, however, the AA1 model gives significantly higher critical supersaturations for the same particle diameter and composition than the more precise (and recommended) Köhler model AP3. Moreover, the relative deviation between AA1 and AP3 is strongly increasing with decreasing critical supersaturation.

That means, when your CCN counter measures the CCN concentration at a low supersaturation (e.g., 0.2%, determined with AA1), the actual supersaturation according to model AP3 would be ~18% smaller (i.e., 0.16%). Assuming that from your CCN measurements you observe for 100 nm particles a critical supersaturation of 0.16% (AP3) instead of 0.2% (AA1), you would calculate, according to Petters and Kreidenweis (2007), a κ value of 0.54 (AP3) instead of 0.34 (AA1). Similarly, at a higher supersaturation (e.g., 0.55%, determined with AA1), the actual supersaturation according to AP3 would be ~14% smaller (i.e., 0.47%). Assuming that you observe for 50 nm particles a critical supersaturation of 0.47% (AP3) instead of 0.55% (AA1), you would calculate a κ value of 0.5 (AP3) instead of 0.36 (AA1). So, only by using a different Köhler model, your results will change a lot. It would be good if you could discuss this issue upon revision of your manuscript and estimate the possible influence on your further results (CCN closure, etc.)

2) For better comparison of your data set with other studies it would be helpful if you could give more general averages of the observed parameters (e.g., not only the average GF for the

individual hygroscopicity categories, but also an overall average GF (weighted by the number fraction in each class); average S_c and κ for individual particle sizes during each campaign).

Specific comments:

P 19684, L 2: better write "total particle concentration"

P 19684, L 22: If you include "κ" after "Hygroscopicity parameters" one can expect already from the abstract that you used the terminology according to Petters and Kreidenweis (2007).

P 19685 and elsewhere: Be more careful in the use of the word "aerosol". Rather use "aerosol particle" or just "particle" when you explicitly talk of the particulate matter (e.g., L13: "particle within the droplet", L 25: "vapor … condenses onto the particle", etc.), since "aerosol" denotes the suspension of particles in a gas.

P 19685, L 18: Asmi et al. (2011) gave a comprehensive overview of particle size distributions in Europe.

P 19686: Please consider adding a few recent publications in the introduction, e.g., L 4: Cerully et al. (2011), L 13: Rose et al. (2011), L 19: Gunthe et al. (2011).

P 19687, L11-14: Was the CPC integrated in the SMPS or operated separately? How was the total particle number concentration measured? By the CPC only (measuring the polydisperse concentration) or by integrating the size distribution measured by the SMPS? Please clarify.

P 19687, L 14-17: Can you please say something on the comparability of the two different CCN spectrometers. Did you perform any direct comparison of the instruments?

P 19688, L 6-9: Do you mean the total CCN concentration with "N_{CCN}" or the concentration of the monodisperse particles? How did you measure the total CCN concentration? Please describe the second path of the scheme in Fig. 2. Did you regularly switch between a polydisperse and monodisperse measurement? What kind of dryer did you use and what was the maximum relative humidity of the aerosol?

P 19688, L 9-11: For how long did you measure at each supersaturation? What was the settling time of the supersaturation (time between setting a new supersaturation level and starting measurement at this level)? How long did it take to finish one complete spectrum of supersaturations?

P 19688, L 20-21: The HTDMA setup is not shown in Fig. 2.

P 19689, L 21-23: Please change to "BCMO 2009 had the lowest $PM_{2.5}$ … concentrations, but its SO_2…"

P 19695, L 11 ff: Do you have any reference for other studies using the same power law approximation? Please compare your results with Rose et al. (2010) who used a similar approach for a data set in south China.

P 19696, L 26: include "could" before "be used for"

P 19696 L 25 – P 19697 L 2: A modeling study by Pringle et al. (2010) also suggested an average $\kappa = 0.3$ for aged continental aerosol.

P 19697 L 3-5: I agree that in your study the particles are mainly of continental origin but since you measured on an island I would expect also marine influence, i.e. higher hygroscopicity than for aged continental aerosol (i.e., on average $\kappa > 0.3$). Can you comment on that? As already mentioned in my general comments, Kuwata et al. (2008) did observe higher κ for the same location. What is the distance (km) between Jeju Island and the mainland?

P 19702, L 2 and 10: write "size-resolved" instead of "size-resolving"

Tab. 2: Is it possible to include a column with the overall average GF at each particle size?

Tab. 3: Why do you list only CCN results for Gosan 2008 and BCMO although you write in Sect. 2.1 that you did CCN measurements during all campaigns? For Gosan 2008 also the results at 50nm are missing.

Tab. 5: It might be good to mention here also whether the prediction over- or under-estimates the measurement (i.e., is there a positive or negative bias in the prediction) rather than giving only the relative deviation.

Fig. 2: What was the setup like during the other Gosan campaigns?

Fig. 3: Please indicate that the supersaturation at h/2 is S_c.

Fig. 4 and 5: What are the error bars?

Fig. 4: Please change the caption to something like "Average concentrations of PM mass and of various gaseous species…"

Fig. 9: What are the parameters of the regression lines and what is the correlation coefficient?

Fig. 11 and 13: Are these plots showing the statistical distribution (median, percentiles, etc.) of κ? If so, please indicate which line of the box and the bar belong to which percentile.

Fig. 13: Should the x-axis label be "N_{CCN_meas}"? Is "_meas" also missing in the caption? Why and how do you merge the data in individual bins? A simple scatter plot (rel. dev. vs. N_{CCN_meas}) would show the same I guess.

References:

Asmi, A., Wiedensohler, A., Laj, P., Fjaeraa, A.-M., Sellegri, K., Birmili, W., Weingartner, E., Baltensperger, U., Zdimal, V., Zikova, N., Putaud, J.-P., Marinoni, A., Tunved, P., Hansson, H.-C., Fiebig, M., Kivekäs, N., Lihavainen, H., Asmi, E., Ulevicius, V., Aalto, P. P., Swietlicki, E., Kristensson, A., Mihalopoulos, N., Kalivitis, N., Kalapov, I., Kiss, G., de Leeuw, G., Henzing, B., Harrison, R. M., Beddows, D., O'Dowd, C., Jennings, S. G., Flentje, H., Weinhold, K., Meinhardt, F., Ries, L., and Kulmala, M.: Number size distributions and seasonality of submicron particles in Europe 2008–2009, Atmos. Chem. Phys., 11, 5505-5538, doi:10.5194/acp-11-5505-2011, 2011.

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. Discuss., 11, 15029-15074, doi:10.5194/acpd-11-15029-2011, 2011.

Gunthe, S. S., Rose, D., Su, H., Garland, R. M., Achtert, P., Nowak, A., Wiedensohler, A., Kuwata, M., Takegawa, N., Kondo, Y., Hu, M., Shao, M., Zhu, T., Andreae, M. O., and Pöschl, U.: Cloud condensation nuclei (CCN) from fresh and aged air pollution in the megacity region of Beijing, Atmos. Chem. Phys. Discuss., 11, 9959-9997, doi:10.5194/acpd-11-9959-2011, 2011.

Pringle, K. J., Tost, H., Pozzer, A., Pöschl, U., and Lelieveld, J.: Global distribution of the effective aerosol hygroscopicity parameter for CCN activation, Atmos. Chem. Phys., 10, 5241-5255, doi:10.5194/acp-10-5241-2010, 2010.

Rose, D., Gunthe, S. S., Su, H., Garland, R. M., Yang, H., Berghof, M., Cheng, Y. F., Wehner, B., Achtert, P., Nowak, A., Wiedensohler, A., Takegawa, N., Kondo, Y., Hu, 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 2: Size-resolved aerosol chemical composition, diurnal cycles, and externally mixed weakly CCN-active soot particles, Atmos. Chem. Phys., 11, 2817-2836, doi:10.5194/acp-11-2817-2011, 2011.