

Interactive comment on "Hygroscopic properties and CCN activity of atmospheric aerosols under the influences of Asian continental outflow and new particle formation at a coastal site in East Asia" by Hing Cho Cheung et al.

Anonymous Referee #1

Received and published: 16 September 2019

The manuscript by Hing Cho Cheung et al. entitled "Hygroscopic properties and CCN activity of atmospheric aerosols under the influences of Asian continental outflow and new particle formation at a coastal site in East Asia" presents aerosol and cloud condensation nuclei (CCN) properties measured at the northern tip of Taiwan Island during a campaign from April 2017 to March 2018. Strong size dependence in the hygroscopicity parameter kappa was observed. Cluster analysis of the back trajectories shows a significant variation in the CCN number concentration (NCCN), number concentration of aerosol particles (NCN), and in the hygroscopicity parameter kappa for different

C1

pollution sources.

The manuscript reports interesting results and is well structured but exhibits some severe deficiencies as detailed below. In principle the paper could become acceptable for publication in ACP, but only after major revisions resolving the open questions and problems outlined below. From a formal perspective, the manuscript is well structured, but the individual arguments and conclusions are not always clear and properly substantiated. The manuscript could be greatly improved by reference and orientation with regard to earlier high-quality CCN measurement studies (methods, analysis, results and quality assurance).

Major and General Comments:

1) Page 5 line 17: The measurement intervals are described. The CCN counter need several minutes after supersaturation changes to stabilize. It is of high importance to remove these time periods from the dataset. It is unclear if this correction has been done for the current analysis. This correction is absolutely needed.

2) Page 5 line 20-21: The authors mention that the CCNC was calibrated, was the CCN calibrat-ed according to Rose et al. (2008)? If not this needs to be further explained. If yes the manuscript needs to be cited. Was the data corrected of the maximal activated fraction, which is of high importance in particular for total CCN measurements (Paramonov et al., 2013; Rose et al., 2010)? In addition, it would be good to refer to a manuscript which describe operation of the DMT CCNC (e.g. Lance et al., 2006).

3) Page 6 line 13-14: The hygroscopicity parameter kappa is very sensible and can be easily calculated wrong using equation 1. The common way to calculate kappa by total CCN number concentration is the numerators of equation 1. This fits very well to the kappa calculated by size resolved CCN measurements (e.g. Rose et al. 2010; Irwin 2011; Gunthe et al. 2011; Jurányi et al., 2011; Pöhlker et al., 2016). Equation 1 can be used if the CPC and the integral of SMPS is the same for the whole measurement period, however, in this case the denomi-nator of equation 1 will be cut out.

4) Page 8 line 2 and Table 2: The values for kappa reported in this study are different to former studies; this is also described by the author. It should be discussed if the different to other studies is coast by the method which has been used and if yes the analysis should be redone.

5) Page 8 line 5: By comparing the calculated kappa to former studies, the difference in the methods to calculate kappa needs to be discussed. Meng et al. (2014)and lwamoto et al. (2016) measured size resolved CCN properties and Schmale et al. (2018) calculated kappa by using AMS measurements. These findings should also be compared to Irwin et al (2011).

6) Page 10 line 25 and Figure 6: The author describe new particle formation events (NFP) in figure 6. I cannot see a significant nucleation mode in Figure 6 in the NFP case. The NFP case seems more like an Aitken mode aerosol size distribution while the non-NPF case is a bimodal Aitken and accumulation mode aerosol size distribution. Also the fitting is not convincing.

7) Page 11 line 5-7: The number concentration of particles smaller 30 nm is reported. How was this number calculated? It can be quite difficult to measure particles smaller 10 nm. It would be important to know the lower cut off for particles smaller 30 nm.

8) Page 11 line 25: The authors report that the increase in CCN cannot be explained by the growth of NPF because the additional CCN were observed at an initial stage of the NPF event. Everything which can be extracted from the manuscript in its present form does not indicate a NPF in an initial state. This is, however, very difficult to detect and need special care in the inlet system. The findings of the manuscript leads to the summery that growth of newly formed particle can explain the increase in CCN.

9) Page 12 line 33-34: This sentence is not clear. The sentence needs to be restructured.

10) Overall the discussion and the final finding is very much focusing on the NPF. This

C3

is not well described in the method section and the NPF events are not clearly shown. I would recom-mend to concentrate on the CCN key parameters in the discussion and do not focus on NPF. If the author is willing to stick to the NPF discussion the method section need to include the losses in the sampling line, the well calibrated detection limit of the instruments and the NFP events need to be presented very clearly and convincing in the manuscript.

Minor Comments

11) The abstract would benefid from representative values for kappa, NCCN and NCN.

12) Page 4 line 20: it would be helpful if the author could cite a reference for the station.

13) Page 8 line 16: "It is noteworthy that both the κ and DSS decrease with the SS". Even if kappa would stay the same Dss would still decrease with SS. And when kappa decrease for a given SS, Dss is increasing not decreasing in comparison relative to the Dss of the same kappa.

14) Figure 7: The axes of the particle size distribution have only one number this make it impos-sible to interpreted the figure.

15) Figure 8: The condensation vapors should be located left of the NPF. The process described in the figure is not the only way to increase CCN by NPF. The condensation vapors for example would usually first condense on bigger particles. Following I would not show this figure in the pressed form as a final finding in the manuscript.

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., 11(21), 11023–11039, doi:10.5194/acp-11-11023-2011, 2011.

Irwin, M., Robinson, N., Allan, J. D., Coe, H. and McFiggans, G.: Size-resolved aerosol water uptake and cloud condensation nuclei measurements as measured above a

Southeast Asian rainforest during OP3, Atmos. Chem. Phys., 11(21), 11157–11174, doi:10.5194/acp-11-11157-2011, 2011.

Iwamoto, Y., Kinouchi, K., Watanabe, K., Yamazaki, N. and Matsuki, A.: Simultaneous Measurement of CCN Activity and Chemical Composition of Fine-Mode Aerosols at Noto Peninsula, Japan, in Autumn 2012, Aerosol Air Qual. Res., 16(9), 2107–2118, doi:10.4209/aaqr.2015.09.0545, 2016.

Jurányi, Z., Gysel, M., Weingartner, E., Bukowiecki, N., Kammermann, L. and Baltensperger, U.: A 17 month climatology of the cloud condensation nuclei number concentration at the high alpine site Jungfraujoch, J. Geophys. Res. Atmos., 116(10), 1–16, doi:10.1029/2010JD015199, 2011.

Lance, S., Medina, J., Smith, J. and Nenes, A.: Mapping the operation of the DMT continuous flow CCN counter, Aerosol Sci. Technol., 40(4), 242–254, doi:10.1080/02786820500543290, 2006.

Meng, J. W., Yeung, M. C., Li, Y. J., Lee, B. Y. L. and Chan, C. K.: Size-resolved cloud condensation nuclei (CCN) activity and closure analysis at the HKUST Supersite in Hong Kong, Atmos. Chem. Phys., 14(18), 10267–10282, doi:10.5194/acp-14-10267-2014, 2014.

Paramonov, M., Aalto, P. P., Asmi, A., Prisle, N., Kerminen, V.-M., Kulmala, M. and Petäjä, T.: The analysis of size-segregated cloud condensation nuclei counter (CCNC) data and its implications for cloud droplet activation, Atmos. Chem. Phys., 13(20), 10285–10301, doi:10.5194/acp-13-10285-2013, 2013.

Pöhlker, M. L., Pöhlker, C., Ditas, F., Klimach, T., Hrabe de Angelis, I., Araújo, A., Brito, J., Carbone, S., Cheng, Y., Chi, X., Ditz, R., Gunthe, S. S., Kesselmeier, J., Könemann, T., Lavrič, J. V., Martin, S. T., Mikhailov, E., Moran-Zuloaga, D., Rose, D., Saturno, J., Su, H., Thalman, R., Walter, D., Wang, J., Wolff, S., Barbosa, H. M. J., Artaxo, P., Andreae, M. O. and Pöschl, U.: Long-term observations of cloud condensation nuclei

C5

in the Amazon rain forest – Part 1: Aerosol size distribution, hygroscopicity, and new model parametrizations for CCN prediction, Atmos. Chem. Phys., 16(24), 15709–15740, doi:10.5194/acp-16-15709-2016, 2016.

Rose, D., Gunthe, S. S., Mikhailov, E., Frank, G. P., Dusek, U., Andreae, M. O. and Pöschl, U.: Calibration and measurement uncertainties of a continuous-flow cloud condensation nuclei counter (DMT-CCNC): CCN activation of ammonium sulfate and sodium chloride aerosol particles in theory and experiment, Atmos. Chem. Phys., 8(5), 1153–1179, doi:10.5194/acp-8-1153-2008, 2008.

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(7), 3365–3383, doi:10.5194/acp-10-3365-2010, 2010.

Schmale, J., Henning, S., Decesari, S., Henzing, B., Keskinen, H., Sellegri, K., Ovadnevaite, J., Pöhlker, M. L., Brito, J., Bougiatioti, A., Kristensson, A., Kalivitis, N., Stavroulas, I., Carbone, S., Jefferson, A., Park, M., Schlag, P., Iwamoto, Y., Aalto, P., Äijälä, M., Bukowiecki, N., Ehn, M., Frank, G., Fröhlich, R., Frumau, A., Herrmann, E., Herrmann, H., Holzinger, R., Kos, G., Kulmala, M., Mihalopoulos, N., Nenes, A., O'Dowd, C., Petäjä, T., Picard, D., Pöhlker, C., Pöschl, U., Poulain, L., Prévôt, A. S. H., Swietlicki, E., Andreae, M. O., Artaxo, P., Wiedensohler, A., Ogren, J., Matsuki, A., Yum, S. S., Stratmann, F., Baltensperger, U. and Gysel, M.: Long-term cloud condensation nuclei number concentration, particle number size distribution and chemical composition measurements at regionally representative observatories, Atmos. Chem. Phys., 18(4), 2853–2881, doi:10.5194/acp-18-2853-2018, 2018.

Interactive comment on Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2019-519, 2019.