Atmos. Chem. Phys. Discuss.,
https://doi.org/10.5194/acp-2019-519-AC2, 2020

Atmospheric Chemistry and Physics Discussions

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.

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Received and published: 22 January 2020

Thank you for the valuable comments. We have further revised our manuscript according to the reviewers' comments. In response to the comments, the data has been recalculated to account for the maximum activated fraction.

Major and specific comments: 1) Page 6 line 22. The eq2. and eq3. Are originally proposed by Petters and Kreidenweis (2007). They suggest this relationship only exist when kappa > 0.2. However, in this study, I noticed kappa < 0.2 often occurs. The

following equation (eq.6. in their paper) can be used for derive more appeasable kappa values:

S(D) = (Dˆ3 – dˆ3)/ (Dˆ3 – D_dˆ3 (1-k)) expaAa ((4sigma_(s/a) M_w)/ (RT)W D)) (1)

Ans: The kappa value has been re-calculated by this equation, with the cut-off diameter (Dcut), which used to represent average hygroscopic of particle with size above the Dcut.

2) Page6 line 26-27. The author assume Dd equals to Dss calculated by eq.1. This approach is simple but not being widely used. The Dss in this study is often referred d_act is related to the chemical composition. But to my best knowledge, most studies use such diameter (d_act or Dss as) didn't further calculate into kappa (Furutani et al., 2008, Quinn et al., 2008, Burkart et al., 2011, Leng et al., 2013). In my personal understanding, Dss calculated by eq.1. contains too much uncertainty, if further calculated into kappa and intercompare with other studies, there might be misleading results. If the authors still like to use the approach in the current version of manuscript, I suggest authors provide a thoroughly discussion on why this method works (e.g. include high-quality references, compare the kappa with those derived from size-resolved measurement at same location, etc).

Ans: The Dd calculated by eq (1) was named cut-off diameter by Rose et al. 2010 which can be represent the average hygroscopic of particles above the Dcut. The discussion about the kappa calculated by Dcut (integral CCN data) and Dss (from size-resolved CCNs data) has been discussed in Section 2.

3) Page 9 line 19-20: The author claims the lower kappa in July-August 2017 in consist with chemical composition measurement. However, I noticed that there were many inconsistencies for the rest of the period. One clearly inconsistency is that in June 2017, the kappa is relatively higher while OC fraction is also very high. I suggest the author give a proper explanation of why kappa and chemical composition shows many inconsistencies otherwise I will suspect the kappa value derived from non-size-resolved

measurement.

Ans: The data used in Figure 5 was PM2.5 chemical composition, while the kappa values were calculated by the integral CCN data with Dcut which mentioned in the reply of previous comments. The presented kappa ($\sim$0.5) was obtained at the setting of SS: 0.3% with D: 70nm while the kappa is around 0.2 for the SS: $\sim$0.5% with D: 52nm (see Figure 4). In previous size-resolved chemical composition of aerosol in northern Taiwan. The higher organic carbon fraction was obtained for UFPs. Therefore, the lower kappa values ($\sim$0.2) for smaller Dcut was reasonable.

4) Section 3.3. The authors suggested NPF enhance NCCN due to coagulation and give a thorough discussion of why it is possible and logical. However, without any quantitively and semi-quantitively estimation and without comparison with other possible pathways (e.g. vapor condensed on sub-CCN, coagulation between sub-CCNs, Oxidation process etc.), it is hard to say coagulation between small particle and sub-CCN is the major cause of CCN enhancement without additional evidence.

Ans: We agreed with the comment that it is hard to proof our hypothesis without quantitative data. However, a significant increase in CCN has been observed accompanying the growth of particles at the later stage of NPF process (see Figure 7), but it is less conclusive to link the increased of CCN to the initial growth of newly formed particles, since the smallest particle size measured in this study is around 13nm. The discussion has been revised accordingly.

Page 12, line 9. The sentence has been added. “Nevertheless, the observed increased in CCN accompanying with the growth of particles could due to various mechanisms (e.g. vapor condensed on existing sub-CCN, coagulation between CCNs, and other oxidation process), and the cause to the increase in CCN and its relation to NPF need to be further studied.”

5) Page5 line1-7. How many data points has been removed? Are those points accounted for a large proportion?

Ans: Page 5, line 1-7 which is mentioned a diffusional loss correction for SMPS data, which does not remove any data. In Page 7, line 1-7: the data point removed by by eq(4) were about 12% for each SS condition.

Page 7, line 19. This sentence has been added. “About 12% of data point has been removed according to eq (4).”

6) Page5 line18-19. Have you also check the sample of CCNC and how good was that? Considering your way of calculating kappa may be very sensitive to accurate reading of number concentration. The total flow (flow entering the CCNC, which then split into sample flow and sheath flow) is important for the accuracy of SS while the sample flow will affect the NCCN reading.

Ans: The flow ratio between sheath and sample flows of the CCNC was within 10+/-0.3, for the period of August and September during which the flooding occurred, and the NCCN data has been removed.

7) Page7 line14-15. The end-point of the trajectories was 200m above ground level. Is the result from such setting consist with those for a lower (e.g. 20m a.g.l.) altitude? If not, what is the specific reason for choosing 200m?

Ans: There are mountains near the coastal CAFÉ station, which may induce higher uncertainty for trajectories calculation under complex terrain. Therefore, we choose the end-point height at 200m, which was referenced to previous studies in norther Taiwan (Cheung et al. 2013, 2016). Also the characteristics of the CO/NOx and O3 were reasonable of the current cluster analysis.

8) Page8 line 32-33. It is too arbitrary to say NPF contribute NCCN only because NCN and NCCN are consist. If the aerosol loading is higher, then both NCN and NCCN are expected to be higher. Please show PM2.5 value of these months to rule out such possibility.

Ans: The PM2.5 data measured for NPF and non-NPF events have been discussed.

Also the hourly data of PM2.5 was included in Figure 7 to show the diurnal variation of PM2.5 for NPF and non-NPF events. In general, the increase of PM2.5 may cause the increase of NCN and NCCN, however, the NCCN was not always follows the trend of PM2.5 at the early stage of NPF. NCCN increased gradually during the growth process, however, the PM2.5 decreased at 0700LT (see Figure 7a) while NCCN and kappa keep increasing with N30-100, and N100-736. Also, the PM2.5 mainly contribute to the mass concentration, rather then number concentration. Therefore, the significant increase of NCCN is not likely due to increase of PM2.5 in this study.

Page 12, line 4. Following sentences have been added. "There is a case that the increase of PM2.5 may cause the increase of NCN and NCCN, however, the NCCN does not follows the trend of PM2.5 at the early stage of NPF. NCCN increased gradually during the growth process, however, the PM2.5 decreased at 10:00 LT (see Figure 7a) while NCCN and kappa keep increasing with N30-100, and N100-736. Also, the PM2.5 mainly contribute to the mass concentration, rather then number concentration. Therefore, the significant increase of NCCN is not likely due to increase of PM2.5 in this study."

9) Figure 4. The plots should be improved. It is very difficult for the readers to grasp the variation from these plots. I suggest the author totally redesign the whole figure or at least adding some vertical grid lines in each plot.

Ans: Figure 4 has been modified, also included the result of all SS settings.

10) Figure 5. Sea salt is a type of aerosol particle, it contains multiple components and some of these components have various possible sources. Please define what is "sea-salt" refers to in this figure and clarify how it is derived from measurement.

Ans: The sea-salt particles in Figure 5 was calculated by the 1.47*[Na+] + [Cl-]. The calculation of sea-salt particles has been added in text.

11) Figure 5. I notice kappa significate increase between 21:00 LT and 2:00 LT. Do you

have any explanation for that?

Ans: As I mentioned in text that most of the NPF events occurred during warm seasons while southernly wind associated with urban pollution dominated in daytime. However, a land and sea breeze circulation also usually observed at CAFÉ station. A aged pollution plume may recirculated back to CAFÉ station from marine boundary. But without high temporal resolution data of aerosol composition, this explanation just my speculation.

Page 12, line 33. the sentences have been added "Furthermore, it was noted that kappa values significantly increased between 21:00 LT and 2:00 LT (Figure 5), this could due to the influence of land-sea breeze circulation during which an aged pollution may be recirculated back to CAFÉ station from marine boundary."

Minor comments: 12) Page 2 line4. I suggest not use the word 'campaign', it is more like a continuous measurement.

Ans: Page 2, line 4. The word 'campaign' has been revised to 'continuous measurement'.

13) Page4 line9-10. Yue et al., (2011) is not a short-term intensive study.

Ans: The term 'a few short-term intensive studies' has been revised to 'a few studies with 1-3 months measurement periods.

14) Page6 line10-11. When NPF occurs, NCN for size<13nm is not negligible. It is more logical to say 'the particles out of the measured particle size range has negligible contribution to NCCN'.

Ans: Page 6, line 23. The sentence has bee revised to 'Also, the number concentration of particles out of the measured particle size range is assumed negligible contribution to NCCN.'

15) Page 10 line 22. The author report 31 NPF events during warm season with an

occurrence frequency of 58.5%. The occurrence frequency should be number of NPF days divided by total days. In such case, did you mean there are only 53 days with PSD data during 4 months?

Ans: There are a total of 53 NPF events throughout the one-year measurement, and 31 out of 53 NPF events were observed during the warm seasons. The total PSD data during June to September is 217 days, hence the occurrence frequency should be 14%.

Page 11, line 17. The sentence has been revised to '. . .representing an occurrence frequency of 14%'.

16) Page 15 line13-21 DOI links are incorrect, please check carefully. Ans: The doi links have been revised.

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