Review Report 1

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 2018 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 pollution sources.

The manuscript reports interesting results and is well structed 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 structed, but the individual arguments and conclusion 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.

Ans: For CCN measurement, only last 5 mins data were used to represent the NCCN at different SS settings, and in kappa calculation as well. This measure was mentioned in text.

Page 5, line 22. "Since CCNC needs several minutes to stable after changing the SS setting, therefore only the last 5 minutes data were used in kappa calculation." has been added.

2) Page 5 line 20-21: The authors mention that the CCNC was calibrated, was the CCN calibrated 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).

Ans: The CCNC was not calibrated exactly according to Rose et al. (2008) and the methodology was further explained. The data was corrected of the maximal activated fraction according to Rose et al. 2010.

Page 5, line 29. The following sentences have been added:

"Since the counting efficiencies of CCNC were lower than CPC, thus the maximum activated fraction of N_{CCN}/N_{CN} would be smaller than 1. Therefore, the activation diameters used to calculate the SS values were determined by using half of maximum activated fraction of N_{CCN}/N_{CN} (Rose et al. 2010). The operation of the DMT CCNC adopted in this study can be referred to 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 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 denominator of equation 1 will be cut out.

Ans: The kappa values were recalculated by using the equation (6) proposed by Petters and Kreidenweis, with the apparent cut-off diameter of CCN activation (D_{cut}) which is diameter above the integral N_{CN} equals the observed N_{CCN}. The correspond kappa values (K_{cut}) was suggested to represent effective average hygroscopicity of CCN-active particles in the size range above D_{cut} (Rose et al. 2010).

The equation (1) in manuscript has been replaced by equation (6) in Petters ad Kreidenweis (2007). The description about the kappa calculation has been revised in Section 2.2 in Page 6.

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.

Ans: More discussion about the difference between the present study to other coastal studies has been added.

Page 8, line 28. "The larger variations of κ values obtained at CAFÉ station compared to the coastal studies at Hong Kong and Noto Peninsula Japan (Meng et al., 2014, Iwamoto et al., 2016) may attribute to the shorter periods of measurements in these two studies which lasted for nearly 1 month in May and October, respectively, while the

present study lasted for 1 year. Moreover, the κ values reported in these previous Asian studies were derived by size-resolved CCN data which represent the average hygroscopicity of he activated aerosol around the activation diameter (D_a), while the kappa was calculated by D_{cut} in this study which represent the average hygroscopic of the aerosol above the size of D_{cut}. Nevertheless, the aerosol composition at CAFÉ station were frequently influenced by local pollution from urban region and regional pollution associated with winter monsoon through different seasons, as indicated in previous studies (Chou et al., 200, 2010, 2017), hence this explains the larger variations in κ values observed in this study. The influence of different airmass on aerosol hygroscopicity will be discussed in Section 3.2."

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 Iwamoto et al. (2016) measured size resolved CCN properties and Schmale et al. (2018) calculated by using AMS measurements. These findings should also be compared to Irwin et al (2011).

Ans: The difference of kappa calculation between the size-resolved CCN from previous studies, and the integral CCN from this study has been mentioned.

Please refer to comment #4.

6) Page 10 line 25 and Figure 6: The author describes new particle formation events (NPF) in figure 6. I cannot see a significant nucleation mode in Figure 6 in the NPF case. The NPF 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.

Ans: The multiple log-normal fitting for the particle size distribution data in this study was calculated by the DO-FIT method which widely applied in a number of previous NPF studies. There is larger variance of nucleation mode particle in NPF group, and the GMD was within nucleation mode. It is plausible that the concentration of the nucleation mode particles was not significantly high, but the PSD clearly indicated that a NPF process was undertaken at CAFÉ station (see ***Figure 7****). Also, the Aitken mode particle (~100nm) always existed at CAFÉ station, therefore, the particles with diameter near 100nm were both observed in NPF and non-NPF groups.*

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.

Ans: The N30 was the number concentration between 13.6nm <= d <= 30nm, and N30-100 was the number concentration between 30nm < d <= 100nm as well as $N_{100\text{-}736}$ (100nm<d<=736nm). The description about the terms N_{30} , $N_{30\text{-}100}$ and $N_{100\text{-}736}$ has been added in Section 2.

Page 5, line 11. "The nucleation, Aitken and accumulation modes particle number concentrations were represented by N_{30} (13 nm < d <= 30 nm), $N_{30\text{-}100}$ (30 nm < d <= 100 nm) and $N_{100\text{-}736}$ (100 nm < d <= 736 nm)., respectively."

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 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 summery that growth of newly formed particle can explain the increase in CCN.

Ans: We agreed that it is very difficult to measure the nucleation-mode particles of very small size, and in this study our smallest size of measurement is 13nm. However, an significant CCN increased 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. Hence, the discussions relevant to this has been removed and this section has been rewritten.

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

Ans: This section has been rewritten.

10) Overall the discussion and the final finding is very much focusing on the NPF. This is not well described in the method section and the NPF events are not clearly shown. I would recommend 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, the well calibrated detection limit of the instruments and the NPF events need to be presented very clearly and convincing in the manuscript.

Ans: Agreed. We have rewritten this section in the manuscript. Please see previous reply to comment #8.

Minor Comments

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

Ans: The abstract has been revised to include the values of kappa, N_{CCN} and N_{CN}.

Abstract: The chemical composition of fine particulate matters ($PM_{2.5}$), the size distribution and number concentration of aerosol particles (N_{CN}) and the number concentration of cloud condensation nuclei (N_{CCN}) were measured at the northern tipo of Taiwan Island during a continuous measurement from April 2017 to March 2018. The parameters of aerosol hygroscopicity (i.e. activation ratio, activation diameter and kappa) were retrieved from the measurements. Significant variations were found in the hygroscopicity of aerosols (κ: 0.18-0.56, SS: 0.12-0.80%), which were subjected to various local and regional pollution sources, including aged air pollutants originating in the eastern/northern China and transported on the Asian continental outflows, fresh particles emitted from local sources and distributed by land-sea breeze circulations as well as produced by new particle formation (NPF) processes. Cluster analysis was applied to the backward trajectories of air masses to investigate their respective source regions. The results showed that the aerosols associated with Asian continental outflows were characterized with lower N_{CN}, N_{CCN} and higher kappa values (N_{CN}: 2520 cm^{-3}, N_{CCN}: 1110 cm^{-3} and κ: 0.42 at SS: 0.28%), whereas higher N_{CCN} and N_{CN} with slightly lower kappa values (N_{CN}: 4850 cm^{-3}, N_{CCN}: 1460 cm^{-3} and κ: 0.40 at SS: 0.28%) were found for aerosols in local air masses.

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

Ans: A reference for the CAFÉ station has been added in Section 2.
Page 4, line 26. "More detailed information of the site description of CAFÉ station can be referred to Chou et al. 2017."

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 decreases for a given SS, Dss is increasing not decreasing in comparison relative to the Dss of the same kappa.

Ans: We agreed with the comment, but the fact that kappa decrease for a given SS and D_{SS} should be increasing applies if large and small particles have same hygroscopicity. Our observation showed that an increase in SS caused a decrease in k and Dss (Dcut). We suggested that in our case, when SS increases, smaller and less hygroscopic particles present get larger and is considered as the activated particles. As the cut-off diameter (Dcut) was adopted (Rose et al. 2008), a smaller k was computed.

Page 9, line 8: To better clarify, the text has been revised as "It is noteworthy that both the κ and D_{cut} were observed with the increase of SS. This could due to the difference in the chemical composition of the aerosols for smaller and larger particles at the site, with less hygroscopic species in smaller particles and more hygroscopic species in larger particles. As the SS increases, smaller and less hygroscopic particles present get larger and is considered as the activated particles. As the cut-off diameter was adopted (Rose et al., 2008), a smaller κ was computed."

14) Figure 7: The axes of the particle size distribution have only one number this make it impossible to interpret the figure.

Ans: Noted. Figure 7 has been re-plotted.

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.

Ans: We agreed on this. The observed increase CCN could be due to other processes as well, and it is not conclusive to attribute this only to NPF in this study. This could due to the fact that the smallest particle size measured in this study is around 13nm, and hence we cannot conclude on the increase in CCN from the newly formed particles at the early stage of NPF. Nevertheless, a significant increase in CCN has been observed with the growth of particles at the later stage of NPF process (see Figure 7). The discussion has been revised accordingly.

Page 12, line 9. The sentences have been revised as "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 needs to be further studies." Figure 8 has been omitted in the manuscript.

Review Report 2

The manuscript "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 Cheung et al., focuses on the analysis of a 1-year dataset of aerosol and cloud condensation nuclei (CCN) properties measured at the northern tip of Taiwan Island. The article does present a new dataset that assists in characterizing aerosol cloud interaction in east Asia. The backward trajectories analysis is applied to separate the Asian continental outflow from local emissions. Significant difference of aerosol and CCN properties also been found between NPF and non-NPF days and potential contribution of NPF to NCCN has been discussed.

The paper is in general well-structured and presents some interesting results. However from technical aspect, the measurement and method used in this paper may cause large uncertainty, the authors are supposed to be more cautious when implement scientific discussion. Overall, I don't recommend for publish in ACP at current stage. However, the manuscript is in principle publishable if most aspects commented below can be properly addressed.

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) = (D^3 – d^3)/ (D^3 – D_d^3 (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 (D_{cut}), which used to represent average hygroscopic of particle with size above the D_{cut}.

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 D_{cut}. The discussion about the kappa calculated by D_{cut} (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 $PM_{2.5}$ chemical composition, while the kappa values were calculated by the integral CCN data with D_{cut} which mentioned in the reply of previous comments. The presented kappa (~0.5) was obtained at the setting of SS: 0.3% with D: 70nm while the kappa is around 0.2 for the SS: ~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 (~0.2) for smaller D_{cut} 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/NO_x and O_3 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 $PM_{2.5}$ data measured for NPF and non-NPF events have been discussed. Also the hourly data of $PM_{2.5}$ was included in Figure 7 to show the diurnal variation of $PM_{2.5}$ 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 N_{30-100}, and $N_{100-736}$. 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 $PM_{2.5}$ may cause the increase of N_{CN} and N_{CCN}, however, the N_{CCN} does not follows the trend of $PM_{2.5}$ at the early stage of NPF. N_{CCN} increased gradually during the growth process, however, the $PM_{2.5}$ decreased at 10:00 LT (see Figure 7a) while N_{CCN} and kappa keep increasing with N_{30-100}, and $N_{100-736}$. Also, the $PM_{2.5}$ mainly contribute to the mass concentration, rather then number concentration. Therefore, the significant increase of N_{CCN} is not likely due to increase of $PM_{2.5}$ 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 N_{CCN}*.'*

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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