Referee's comments:

1. Section 2.2.2: Please give more information of reference data used in the köhler theory when performing the CCNC calibration. This is very important because different parameterizations will retrieve different critical supersaturations (Rose et al., 2008;Wang et al., 2017).

Response:

We agree with the referee that different parameterizations will retrieve different critical supersaturations. When performing the CCNc calibration, we include some important reference data for the ammonium sulfate particles that we used in the CCNc calibration. Specifically, the density and molecular weight of ammonium sulfate were assumed to be 1770 kg m^{-3} and 0.132141 kg mol^{-1}, respectively.

Changes in the manuscript:

We have added three sentences on L15-20 on p.10 regarding the CCN calibration, "Previous studied showed that different parameterizations in the Köhler theory can retrieve different critical supersaturations (Rose et al., 2008; Wang et al., 2017). When performing the CCNc calibration, we assumed the density and molecular weight of ammonium sulfate to be 1770 kg m^{-3} and 0.132141 kg mol^{-1}, respectively. We also set the temperature and the pressure to 298.15 K and 1026 hPa, respectively. A temperature gradient ΔT of about 3-8 K in the CCNc column was also used in the calibrations."

2. Section 2.3.4: It should be section 2.3.3. Still, I am confused with the method to perform the CCN prediction based on HTDMA data. I would suggest the authors give an exemplary case either in the text or supplement referring to Lukas et al., (2010).

Response：

We proposed five schemes to predict the CCN activity as shown in Table 4. The prediction of N_{CCN} using activation curve means that the N_{CCN} was calculated based on eq. 6 (L21 on p.14). The activation curve can either be real time (Scheme 1 in Table 4) or average (Scheme 2 in Table 4). The prediction of N_{CCN} using the D_{50} means that the N_{CCN} was calculated based on eq. 7 (L6 on p.15) from either

real time data (Scheme 3 in Table 4) or average data (Scheme 4 in Table 4).The D_{50} was determined from fitting the size-resolved activation ratio by eq. 4 (L14 on p.13). All the above schemes (Schemes 1-4) use an unadjusted surface tension of water ($\sigma_{s/a}$=0.072 N m^{-1}). Scheme 5 predicts N_{CCN} using activation curve from real time data and it uses an adjusted surface tension of water ($\sigma_{s/a}^{*}$=0.058 N m^{-1}). The approach we used here is similar to the one employed in Kammermann et al. (2010) as we described the detailed changes below.

Changes in the manuscript:

We have now modified the relevant paragraphs in section 2.3.3 (previously misspelled 2.3.4) as follows,

L5-7 on p.14: "Figure 1 is the schematic diagram of the four approaches we followed to predict N_{CCN} based on the above two measured datasets. In the first approach (I in Fig. 1), the mixing state and size dependence were taken into account."

L11-14 on p.14: "A particle with a κ value higher than $\kappa_{critical}$(Dp,SS) was considered to be activated as an CCN (Fig. 1a) and the shadow area represented the particles which can be activated as CCN for a known diameter and SS."

L17 on p.14 to L12 p.15: "This approach is similar to the one employed in Kammermann et al. (2010), however, we used the size-resolved activation ratio (AR_{SR}) to calculate the N_{CCN}. The AR_{SR} was determined by fitting the AR(Dp,SS) to the diameter Dp using eq. 4 for the five measured diameters (Fig. 1d). Thus, the calculated N_{CCN} using the activation ratio can be expressed as (Fig. 1e):

$$N_{CCN}(SS) = \int_{0}^{\infty} AR_{SR}(Dp,SS)N_{CN}(Dp)dDp \tag{6}$$

In the second approach (II in Fig.1), the particles were assumed to be internally mixed. The D_{50} was determined by fitting the AR(Dp,SS) to the diameter Dp (Fig. 1d). The N_{CCN} was obtained by integrating the cloud nuclei concentration for particles larger than D_{50} based on the particle size distribution (Fig. 1f), according to the following equation (eq. 7):

$$N_{CCN}(SS) = \int_{D_{50}}^{\infty} N_{CN}(Dp)dDp \tag{7}$$

In the third and fourth approaches (III and IV in Fig.1), the particles were also

assumed to be internally mixed. We then calculated the κ value according to the ZRS rule (eq. 8) based on the AMS measurements.

$$\kappa = \sum_i \varepsilon_i \kappa_i \tag{8}$$

where ε_i is the volume fraction of each component in the particles, κ_i is the κ value of each component."

L6-10 on p.16: "Here instead of being determined from fitting of AR_{SR} to Dp used in the second approach, the D_{50} was calculated from the above κ values using eq. 5. In the third approach, the κ values were size-resolved because the chemical composition of the particles was size dependent (Fig. 1b). In the fourth approach, the particles were assumed to have the same chemical composition and hygroscopicity as those in PM_1 (Fig. 1c). The N_{CCN} was then predicted using eq. 7 (Figs. 1g and 1h)."

Since we moved eq. 9 forward to eq. 7, we hence modified the numbers of the subsequent equations in order, i.e., original eq. 7 to eq.8, 8 to 9, etc. We have also modified Fig. 1 for better clarification.
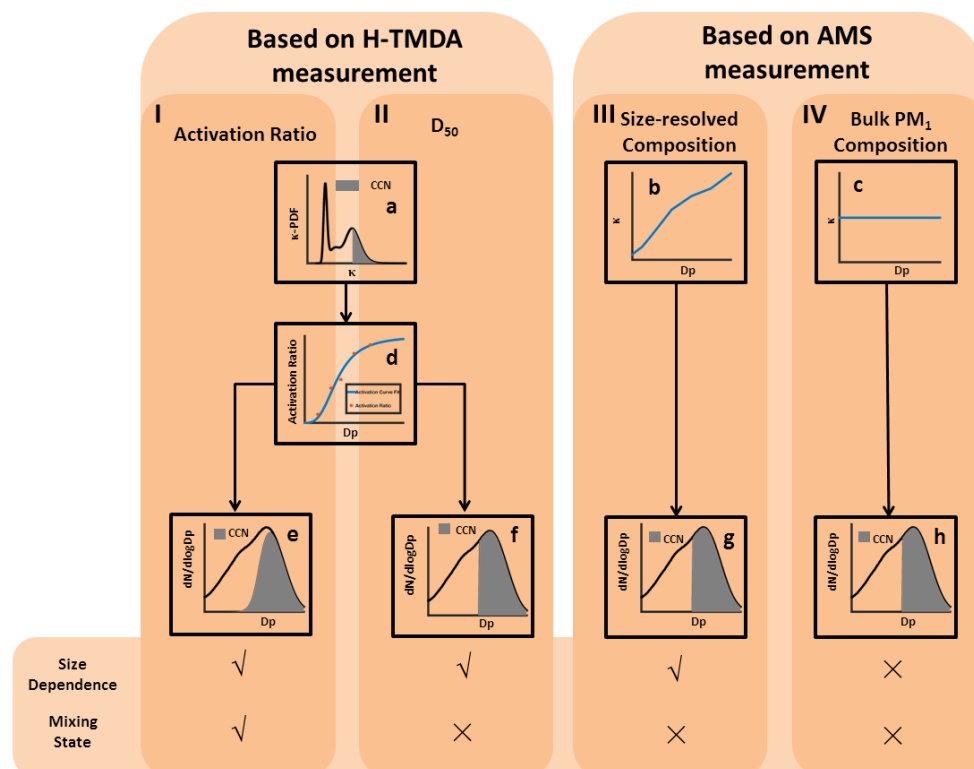

Fig. 1. A schematic representation of N_{CCN} prediction based on the H-TDMA and the AMS measurements. The N_{CCN} can be predicted based on the fitted activation

ratio (approach I) and the D_{50} (approach II) both obtained from the H-TDMA measurement, the size-resolved composition (approach III) and the bulk PM_1 composition (approach IV) both obtained from the AMS measurement. Panel (a) is the representation of calculating the activation ratio for a specific diameter and SS and the shadow area represents the particles which can be activated as CCN; (b) and (c) are the representations of the κ values obtained respectviely from size-resolved chemical composition and bulk chemical composition; (d) is the reprentation of fitting the activation ratio to the particle diameter Dp (red dot); (e), (f), (g), and (h) are the representations of predicting the N_{CCN} using the four approaches respectviely and the shadow area repsents the particles which can be activated as CCN.

3. Table 1: where these data come from, please add reference. I guess these kappa data are retrieved with T of 298.15 K. But in Section 2.3.1, the T you used is 293 K, why?

   We have included the references in Table 1. We have changed the temperature in Section 2.3.1 to 298.15 K for consistency.

Table 1. The κ values of the related species in the study.

Species	κ
NH_4NO_3	0.58[a]
NH_4HSO_4	0.56[a]
H_2SO_4	0.90[a]
$(NH_4)_2SO_4$	0.48[a]
Organics	0.10[b]

[a] The κ of inorganics compounds are derived from ADDEM (Topping et al., 2005)

[b] The κ of organics was taken from Meng et al. (2014)

4. Table 2: Based on the SMCA measurements, you should get size-resolved activation ratio, so I do not understand the max. and min. values of AR here? In principle, it should be 1 and 0. I guess you calculate the overall AR, please clarify

and explain the reason why you put it here

The activation ratio (AR) in Table 2 represents the ratio of total number of cloud condensation nuclei (N_{CCN}) to the total particle number ($N_{CN, tot}$). The total AR was not only affected by the particle hygroscopicity but also the particle number size distribution (PNSD). For example, if the PNSD was unimodal with a peak at about 100 nm, while the D_{50} was about 180 nm at 0.1% SS, resulting in only a minor fraction of particles that were larger than the D_{50}, implying a low value of AR. However, during the pollution events, the PNSD was often boarder and the peak was shifted to a larger size (e.g., 130 nm), leading to a larger AR. To avoid any confusion, we use $N_{CCN}/N_{CN,tot}$ instead of activation ratio to represent the ratio of total N_{CCN} to the total particle number in the manuscript.

5. Page 16, line 8-10: Please explain this sentence, it is not clear.

   Since the sentence did not contribute substantially to the main point in Fig. 2, we have removed it from the text to avoid confusion.

6. Page 17, line3-4: This is not consistence with the statement in the abstract, please revise.

   In Fig. 3, the D_{50} at a specific supersaturation was the fitted parameter from eq. 4 based on the CCN measurements. For clarification, we have changed the sentence " …the corresponding κ_{CCN} at most of the SS…" to " …the corresponding κ_{CCN} at most of the particle sizes…"(L12 on p. 18).

7. Page 17, line 5-7: The difference between kappa-CCN and kappa-HTDMA may also due to the parameterizations used in the CCNC and HTDMA calibration. See Wang et al., (2017). Please consider it and give more information as suggested in comment 1.

   We agree with the referee. We figured out that the difference between κ_{CCN} and $\kappa_{H\text{-}TDMA}$ is statistically insignificant. We have added a sentence in L9-11 on p. 18, "As shown in Fig. 3, the difference between κ_{CCN} and $\kappa_{H\text{-}TDMA}$ is statistically insignificant at all employed diameters, while the one between κ_{AMS} and κ_{CCN} became statistically significant at larger sizes of the particles."

8. Page 17, line 11-12: Any evidence? I guess the larger hygroscopicity is mainly due to the bigger particle size.

Response:

Based on our measurements, we believe that the peak of less-hygroscopic mode for larger size particles was smaller, indicating that the number fraction of less-hygroscopic particles was lower. Our results further suggest that the fraction of the hygroscopic organics became higher or the organics were coated with hygroscopic inorganics matters, leading to the increase of hygroscopicity for larger particles. As suggested by the other referee, we analyzed the size-resolved f44 of the AMS data. Figure S1 showed that the f44 increased with diameter, indicating that the degree of oxidation of the organics was higher for larger particles. It could also relate to the higher hygroscopicity of organic aerosol for larger particles. Note that the f44 for particle diameters smaller than 100 nm was discarded due to the poor data quality for those particles.

Changes in the manuscript:

We have included Fig. S1 in the supplementary material. We have also added several sentences in L20 on p.18 to L6 on p.19, "Previous studies showed that the κ_{org} values of larger particles are lower than those for smaller particles (Lance et al., 2013; Zhao et al. 2015) and hygroscopicity of organics is often found to be related to its chemical composition (f44 or O/C) in both field and laboratory studies (Chang et al. 2010; Massoli et al., 2010; Lambe et al., 2011; Mei et al., 2013, and others reference therein). We showed that the f44 increased with the particle size from the AMS data (Fig. S1). Note that the f44 for particle diameters smaller than 100 nm was discarded due to the data quality. The results indicate that the degree of oxidation of the organics was higher for larger size particles and the hygroscopicity for larger particles is higher (Chang et al., 2010)."
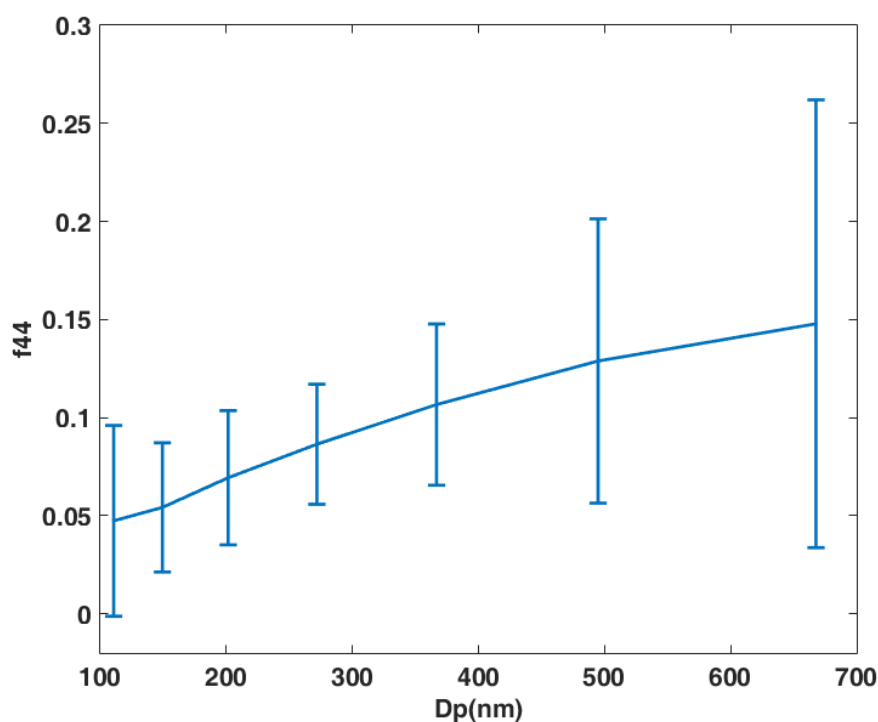
9. Figure 4, have you corrected the double charge effect of DMA?

Response:

The contributions of multiply charged particles were taken into account and we did the multiply charged correction for the SMCA, SMPS and H-TDMA data when the data were inverted. Thus the effects of multiply charged particles in Figs. 5 and 7 have been considered. Figure 4 shows the fitted curve of the activation ratio measured by the SMCA, which has also been corrected for the contribution of doubly charged particles.

Changes in the manuscript:

We have now incorporated the above discussion to L16-18 on p.12, "Note that we include multiply charged correction for the SMCA, SMPS and H-TDMA data when the data were inverted so that the contributions of the multiply charged particles were accounted for all the measured particle data." We have modified the caption of Fig. 4 for clarification, "Fig. 4. The sized resolved activation ratios measured by the SMCA at four different supersaturations. Note that the curves were fitted according to the SMCA measurements."

10. Page 17, line 16-19: This sentence ("the peak in the less-hygroscopic mode declined … while the one in the more-hygroscopic mode climbed…") is not clear. Do you mean the relative fraction of less-hygroscopic compounds decreased and more-hygroscopic compounds increased at larger particle size?

We have added several sentences after "…, indicating that larger particles tend to be internally mixed"(L19 on p.20 to L3 on p.21), "Since less-hygroscopic particles were usually associated with externally mixed black carbon (BC) or fresh organics and more-hygroscopic particles usually represent the inorganics matters or BC coated with inorganics matters (internally mixed). The decrease of peak area of less-hygroscopic mode and the increase of more-hygroscopic mode indicate that the number fraction of less-hygroscopic particles decreased while the more-hygroscopic particles fraction increased. Thus, the particles became more internally mixed."

We believe that we have addressed this issue and we have then deleted the titled sentence for clarification.

11. Figure 7: I am not quite sure that the impacts of organics can fully explain the difference between the calculated and measured AR. The bias is still obvious even the configured surface tension (0.072) is used, indicating the other factors should also be considered. Many studies (Petters et al., 2009;Wex et al., 2009;Hersey et al., 2013;Wu et al., 2013;Hong et al., 2014;Hansen et al., 2015;Mikhailov et al., 2015;Pajunoja et al., 2015;Zhao et al., 2016) have reported the different hygroscopic properties from CCNC and HTDMA measurements. I would suggest more discussions should be added here.

Response:

The referee raised a very good point on the factors which affect the agreement between the calculated AR and the measured AR. The H-TDMA measures the hygroscopicity of the particles regardless of their compositions. For particles containing non-hygroscopic compounds, e.g., the sparingly soluble compounds, a unity of growth factor was measured in the H-TDMA, while for those with hygroscopic compounds, a growth factor greater than unity was measured. Thus the impact of the sparingly soluble compounds in the particles was taken into account when the AR was calculated. However, we agree that factors other than surface tension may also play roles. Previous studies found that the hygroscopicity of the particles measured by the H-TDMA could be lower than that measured by the CCNc (Chan et al., 2008; Pajunoja et al. 2015; Petters et al., 2009; Hansen et

al., 2015; Hong et al., 2014) which might be attributed to low soluble compounds in the particles. However, the contributions of low soluble compounds cannot currently be quantitatively evaluated. Laboratory experiments showed that the CCN activity would increase by adding organics to sulfate ammonium particles (Engelhart et al., 2008). Our study found that the calculated AR values were systematically lower than the measured ones if a value of 0.072 N m^{-1} was used for the surface tension. We hence believe that the surface tension might play a major role in the AR prediction than other factors, i.e. the effect of "sparingly soluble" compounds. Indeed, as we showed in the paper, the R^2 of the fitting between the measured AR and the calculated AR became higher and the δ_{AR} approached to zero by adjusting the surface tension from 0.072 N m^{-1} to 0.058 N m^{-1} (Fig. 8), suggesting that the AR prediction could be greatly improved by simply adjusting the value of the surface tension. Because we use a single $\sigma_{s/a}$ value (i.e., 0.058 N m^{-1}) for all size particles in the AR prediction, it could lead to over- or under-prediction of the AR for a specific particle size. For example, the AR for 150 nm particles at 0.1% SS was overestimated by using a $\sigma_{s/a}$ value of 0.058 N m^{-1}.

Changes in the manuscript:
We have added several sentences to discuss the factors that could cause the difference between the calculated AR and measured AR in L7-14 on p.23,
"Note that the surface tension is not the only factor that determines the AR and other factors such as the sparingly soluble compounds in the particles may contribute to the AR, although they are currently not understood. Previous studies found that the hygroscopicity of the particles measured by the H-TDMA could be lower than that measured by the CCNc (Chan et al., 2008; Pajunoja et al. 2015; Petters et al., 2009; Hansen et al., 2015;Hong et al., 2014) which might be attributed to low soluble compounds in the particles. The deviation of the calculated AR from the measured AR is probably dependent on the degree of dissolution of particles and the oxidative state of the organics in the particles."

12. Page 20, line 20-21: Add reference.

   The reference has been added (L17 on p. 23).

13. Figure 9: please provide R^2.

14. Figure 10 and 11: what dose the black line mean? Is it 1:1 line? then the scale should be checked.

The black lines represent the 1:1 lines. Figure 10 and 11 and their captions have been revised as attached below:
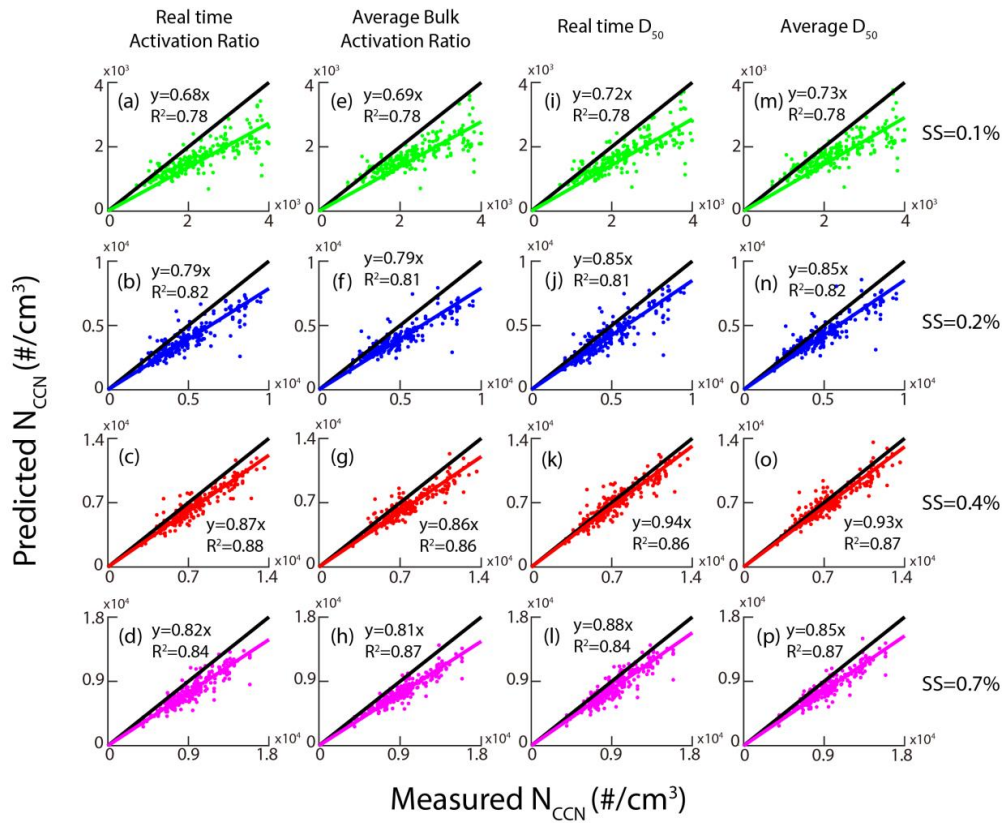

Fig. 10. The relationship between measured N_{CCN} and predicted N_{CCN} based on scheme 1, 2, 3, 4 and 5. The black lines represent 1:1 lines.
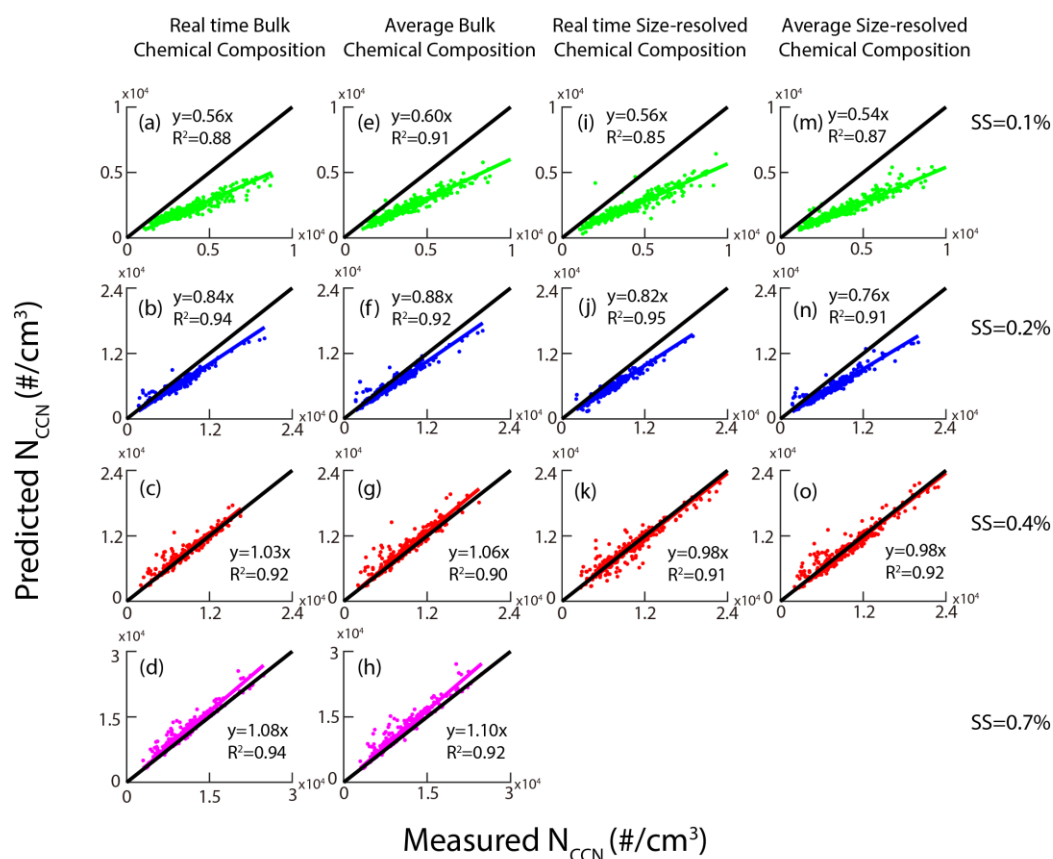
Fig. 11. The relationship between measured N_{CCN} and predicted N_{CCN} based on scheme 6, 7, 8 and 9. The black lines represent 1:1 lines.

15. There are several grammar mistakes in the text, the language and symbols should be checked carefully once more before publication.

We have carefully checked the texts to improve the quality of the manuscript.

References

Chan, M. N., Kreidenweis, S. M., and Chan, C. K.: Measurements of the hygroscopic and deliquescence properties of organic compounds of different solubilities in water and their relationship with cloud condensation nuclei activities, Environ. Sci. Technol., 42, 3602, 2008.

Chang, R. Y. W., Slowik, J. G., Shantz, N. C., Vlasenko, A., Liggio, J., Sjostedt, S. J., Leaitch, W. R., and Abbatt, J. P. D.: The hygroscopicity parameter (κ) of ambient organic aerosol at a field site subject to biogenic and anthropogenic influences: relationship to degree of aerosol oxidation, Atmosp. Chem. Phys., 10, 5047-5064, 2010.

Engelhart, G., Asa-Awuku, A., Nenes, A., and Pandis, S.: CCN activity and droplet growth kinetics of fresh and aged monoterpene secondary organic aerosol, Atmos. Chem. Phys., 8, 3937-3949, 2008.

Farmer, D. K., C. D. Cappa, et al. (2015). "Atmospheric Processes and Their Controlling Influence on Cloud Condensation Nuclei Activity." Chemical Reviews **115**(10): 4199-4217.

Hansen, A. M. K., Hong, J., Raatikainen, T., Kristensen, K., Ylisirniö, A., Virtanen, A., Petäjä, T., Glasius, M., and Prisle, N. L.: Hygroscopic properties and cloud condensation nuclei activation of limonene-derived organosulfates and their mixtures with ammonium sulfate, Atmos Chem Phys, 15, 14071-14089, 10.5194/acp-15-14071-2015, 2015.

Hong, J., Häkkinen, S. A. K., Paramonov, M., Äijälä, M., Hakala, J., Nieminen, T., Mikkilä, J., Prisle, N. L., Kulmala, M., and Riipinen, I.: Hygroscopicity, CCN and volatility properties of submicron atmospheric aerosol in a boreal forest environment during the summer of 2010, Atmos. Chem. Phys., 14, 29097-29136, 2014.

Lambe, A. T., T. B. Onasch, et al. (2011). "Laboratory studies of the chemical composition and cloud condensation nuclei (CCN) activity of secondary organic aerosol (SOA) and oxidized primary organic aerosol (OPOA)." Atmospheric Chemistry and Physics **11**(17): 8913-8928.

Lance, S., T. Raatikainen, et al. (2013). "Aerosol mixing state, hygroscopic growth and cloud activation efficiency during MIRAGE 2006." Atmospheric Chemistry and Physics **13**(9): 5049-5062.

Massoli, P., A. T. Lambe, et al. (2010). "Relationship between aerosol oxidation level and hygroscopic properties of laboratory generated secondary organic aerosol (SOA) particles." Geophysical Research Letters **37**.

Mei, F., A. Setyan, et al. (2013). "CCN activity of organic aerosols observed downwind of urban emissions during CARES." Atmospheric Chemistry and Physics **13**(24): 12155-12169.

Mochida, M., Nishita-Hara, C., Kitamori, Y., Aggarwal, S. G., Kawamura, K., Miura, K., and Takami, A.: Size-segregated measurements of cloud condensation nucleus activity and hygroscopic growth for aerosols at Cape Hedo, Japan, in spring 2008, J. Geophys. Res.: Atmos., 115, 2010.

Koehler, K., Kreidenweis, S., DeMott, P., Prenni, A., Carrico, C., Ervens, B., and Feingold, G.: Water activity and activation diameters from hygroscopicity data-Part II: Application to organic species, Atmos. Chem. Phys., 6, 795-809, 2006.

Pajunoja, A., Lambe, A. T., Hakala, J., Rastak, N., Cummings, M. J., Brogan, J. F., Hao, L., Paramonov, M., Hong, J., and Prisle, N. L.: Adsorptive uptake of water by semisolid secondary organic aerosols, Geophys. Res. Lett. 42, 3063-3068, 2015.

Petters, M., and Kreidenweis, S.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961-1971, 2007.

Petters, M. D., Wex, H., Carrico, C. M., Hallbauer, E., Massling, A., McMeeking, G. R., Poulain, L., Wu, Z., Kreidenweis, S. M., and Stratmann, F.: Towards closing the gap between hygroscopic growth and activation for secondary organic aerosol

– Part 2: Theoretical approaches, Atmos. Chem. Phys., 9, 3999-4009, 10.5194/acp-9-3999-2009, 2009.

Topping, D. O., McFiggans, G. B., and Coe, H.: A curved multi-component aerosol hygroscopicity model framework: Part 1 – Inorganic compounds, Atmos. Chem. Phys., 5, 1205-1222, 10.5194/acp-5-1205-2005, 2005.

Zhang, X. Y., Wang, J. Z., Wang, Y. Q., Liu, H. L., Sun, J. Y., and Zhang, Y. M.: Changes in chemical components of aerosol particles in different haze regions in China from 2006 to 2013 and contribution of meteorological factors, Atmos. Chem. Phys., 15, 12935-12952, 2015.

Zhao, D. F., A. Buchholz, et al. (2015). "Size-dependent hygroscopicity parameter (κ) and chemicalcomposition of secondary organic cloud condensation nuclei." Geophysical Research Letters **42**(24): 10920-10928.