Responses to anonymous referee #1

Comment: Page 3, line 58: "… lead to 40% changes in predicted cloud condensation nuclei (CCN) concentration." The "40% changes" here is confusing, as which is difficult to tell from the sentence whether "a 40% increase or decrease in the NCCN" was resulted from the "50% increase or decrease in κorg". The similar problem exists in the following sentence, which didn't state clearly the corresponding relationship between the average difference in aerosol radiative forcing and change in κorg., e.g.,which scenario (κorg =0.05 or κorg =0.15) corresponds to a higher radiative forcing? A straightforward delivery way is necessary to avoid ambiguity.

Response: Thanks for your comment. Changes are made to those sentences to make them more straightforward. The sentence about CCN is modified to "Liu and Wang (2010) demonstrated that 50% increases in κ of secondary organic aerosol (0.14±0.07) can result in up to 40% increases in predicted cloud condensation nuclei (CCN) concentration.". The sentence regarding aerosol radiative forcing is revised as "Rastak et al. (2017) reported that global average aerosol radiative forcing could decrease about 1 W/m^2 should κ_{OA} increase from 0.05 to 0.15".

Comment: Page 3, line 65: Unlike the variation of κorg itself, I'm afraid I didn't find any connection between the importance of size-dependent κorg and the abovementioned content. Some details and corresponding references are needed to support the importance of the size influence on κorg and related climatic effects. I would recommend the authors to reorganize the context of size influence on κorg, which can be combined with the information provided in the third paragraph (i.e.,contents related to the HTDMA and CCN measurements).

Response: Thanks for your comment. We have deleted "size dependence of"

Comment: Page 4, line 109: How is the 'mobility diameter of 800 nm' obtained? Related information and references are preferred for the conversion here

Response: Thanks for your comment. This part is revised as "particulate matter with aerodynamic diameter less than 1 μm, corresponding to mobility diameter of approximately 760 nm assuming spherical particles and a particle density of 1.7 g/cm^3"

Comment: Page 6, line 151: Based on the introduction of each instrument, only the humidified nephelometer can measure both PM10 and PM1. How can chemical compositions of PM10 be measured with ACSM, which is designed with a PM2.5 aerodynamic lens/impactor as mentioned in Line 148? Similarly, how can SMPS measure the size distribution of PM10?

Response: Thanks for your comment. As introduced in L152, the inlets of instruments of groups 2 and switch every 15 minutes. This setup makes it possible for ACSM, SMPS and the humidified nephelometer to measure both properties of PM1 and PM10 with a time resolution of 30 minutes. However, the ACSM itself has an impactor with a critical diameter of 2.5 μm, which is why it cannot measure the total mass of different components of sampled particles when its upstream inlet is PM10.

Comment: Page 8, line 206: Why is the density for size conversion regarded as 1.7 g/cm3, the same as that of black carbon used in the calculation of κorg? According to the data reported, the organic fraction is always the predominant contributor to the particle mass of PM2.5. This might suggest a smaller density for the ambient particles. Then how to evaluate the uncertainty in the related calculations?

Response: Thanks for your comment. A density of 1.7 g cm3 for the particles larger than 800 nm as a mean density for the coarse mode is a typical value of converting APS aerodynamic diameter to mobility diameter (Wehner et al., 2008). We agree with the reviewer that the aerosol density might change. And the organic fraction is indeed the predominant contributor to the measured particle mass of PM2.5 by ACSM. However, the APS measures the size distribution of coarse particles, and the density of those coarse particles cannot be inferred from or speculated by only using ACSM measurements, because the ACSM cannot measure all components of ambient particles. Some components like dust, which have higher density (Atkinson et al., 2015), cannot be measured by ACSM as discussed in Sec.3.2.

Comment: Page 10, line 243: For the "iterative calculation using the Mie theory", how are the chemical composition and corresponding mixing state of particles considered in the κf(RH) calculation? This would affect the closure/comparison between derived κf(RH) and κchem, thus the interpretation of representativeness of κf(RH)

Response: The iteration calculation procedure is introduced in Kuang et al. (2017) in detail. The mixing state of particles are assumed to be internally mixed.

Comment: Page 11, line 266: I suppose that you were assuming black carbon as hydrophobic, rather than hydrophilic, and κBC is approximately taken as 0. Supporting references would be needed for this point and also for the density assumption of BC in Line 275. A similar typo was found for the description of 'Dust' in Page 12, line 309, which would be hydrophobic instead of hydrophilic.

Response: Thanks for your comment. We have changed hydrophilic to hydrophobic for the description of BC and dust. The reference for density assumption of BC was also added.

Comment: Page 12, line 307: What does the 'by' mean: "… influences of unidentified material by the ACSM …"? Are you suggesting 'not detected by' ACSM?

Response: Yes, it can be understood as "not detected by the ACSM", we think unidentified might be better because those components are indeed sampled in the ACSM but ACSM does not know what they are.

Comment: Page 13, line 313: "Bulk aerosol chemical compositions and aerosol hygroscopicity κf(RH) measurements are available, one would naturally jump to the conclusion of treating κf(RH) as $\kappa chem$ to derive κOrg (both are from bulk aerosol measurements)."

A connection like a conjunction is needed for the whole sentence. "Both" here sounds ambiguous, although I would assume them to be κf(RH) and $\kappa chem$. It's better to specific which two hygroscopicity parameters you were referring to.

Response: This sentence is modified as “one might naturally jump to the conclusion of treating $\kappa_{f(\mathrm{RH})}$ as κ_{chem} to derive κ_{OA} because both $\kappa_{f(\mathrm{RH})}$ and κ_{chem} are from bulk aerosol measurements”

Comment: Page 16, Line 380: Why is the κf(RH) uniform for all the particle sizes? If yes, does it mean that κDp = κf(RH), while κDp itself is size dependent?

Response: Yes, κf(RH) is defined as the uniform value κ that can be used to best fit the observed f(RH). Therefore, κf(RH) being uniform for all the particle sizes is a basic assumption in the κf(RH) retrieval.

Comment: Page 17, Line 387: What does the ‘which’ mean?

Line 388: Is there any consideration of choosing “two extreme cases of size resolved κDp

Response: “which” is changed to “this result of X_c” to make it clearer. The relative difference between κf(RH) and κchem are mostly influenced by shape of size-resolved κDp distribution. Thus, the two extreme cases of size resolved κDp can give the upper range of relative differences of κf(RH) and κchem for PM1.

Comment: Page 18, Figure 4(c): In comparison of the PM1 scenario displayed in Fig.4b, κf(RH) is generally higher than $\kappa chem$, and larger discrepancies exist for the PM10 case. Can you provide some hints for these results?

Line 411: "How much does $\kappa chem$ differ from κf(RH) for PM1 and PM10 samples?" In my understanding, the $\kappa chem$ of PM10 samples is calculated from the corresponding chemical compositions that are actually measured for PM2.5 instead of PM10 (due to the configuration of ACSM with a PM2.5 impactor), when sampling with a PM10 inlet. In this sense, the two hygroscopicity parameters for PM10 samples would correspond to the water uptake by particles of different size ranges.

Response: The chemical component measurements during the field campaign used in this study is not used to discuss differences of $\kappa chem$ and κf(RH) of PM10. The $\kappa chem$ and κf(RH) of PM10 shown in Fig.4c are calculated based on size-resolved κ distribution as shown in Fig.S5. To make this part clearer, the paragraph describing results of Fig.4c is revised as the following: "For PM10, values of κ_{chem} and $\kappa_{f(\mathrm{RH})}$ using κ_{D_p} size distributions derived from ambient measurements (Fig.S5, similar to Fig.4b) were simulated and displayed in Fig.4c. The simulated absolute values of the relative difference between κ_{chem} and $\kappa_{f(\mathrm{RH})}$ ranged from 0.2% to 41% with an average and standard deviation of 16±8 %, with all κ_{chem} lower than $\kappa_{f(\mathrm{RH})}$. This is because, for PM10, super-micron particles typically with low hygroscopicity (Fig.S5) contribute much more to V_{tot} than to σ_{sp} (as shown in Fig.S7). These results indicate that, for PM10, $\kappa_{f(\mathrm{RH})}$ cannot accurately represent κ_{chem}."

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Comment: Page 19, Line 420: It feels like "thus smaller particles play a more significant role in $\kappa chem$" concluded here is a bit too early, as $\kappa chem$ is determined not only by the volume fraction but also by the hygroscopicity of each composition. Small particles with higher κDp normally correspond to much higher κ values for both $\kappa chem$ and κf(RH). Considering the much smaller variation range of $\kappa chem$ or κf(RH) caused by Ångström exponents, influence from κDp of smaller particles would be more significant.

Line 427: in Fig.4a, the variation ranges of $\kappa chem$ and κf(RH) are much smaller than those in Fig.4b of PM1. Can we say that the influence of the particle size distribution (as denoted by the Ångström exponent) is not that important for $\kappa chem$ or κf(RH)?

Response: We think the reviewer has a misunderstanding here. This sentence is to explain why calculated κ_{chem} is smaller than κ_{fRH} for the results shown in Fig.4a. And the variation ranges of $\kappa chem$ and κf(RH) in Fig.4a are much smaller than those in Fig.4b of PM1, because $\kappa chem$ and κf(RH) in Fig.4a are calculated based on fixed size-resolved κDp distribution shown in Fig.3. The $\kappa chem$ and κf(RH) shown in Fig.4b are calculated based on about 23 size-resolved κDp distribution which are derived from measured size-resolved chemical compositions in the NCP region. Based on the results of Fig.4, we can say that the influence of particle size distribution (as denoted by the Ångström exponent) is not that important for $\kappa chem$ or κf(RH).

Comment: Page 20, Line 440: "… with all $\kappa chem$ lower than κf(RH)." For the discrepancies between $\kappa chem$ and κf(RH), is it also because that $\kappa chem$ is only derived from PM2.5 rather than PM10 measurements? How to evaluate the effect of size-cut of ACSM especially for measurements with a PM10 inlet?

Line 449: I guess ‘NR-PM2.5’ and ‘NR-PM1’ is reversely sequenced, similar to the orders of ‘PM10’ and ‘PM1’’ in the following sentence of the same page and in Line 570 of Page 27

Line 456: “During the first period”, is there any predefinition of the first/second or any other period (e.g., the “non-fog periods” in Page 22, Line 478)?

Response: Thanks for your comment. This part is still theoretically discussing the discrepancies between $\kappa chem$ and κf(RH), both kchem and κf(RH) are derived from size-resolved κ distribution of PM10. Discussions here are not relevant to ACSM measurements.

‘NR-PM2.5’ and ‘NR-PM1’ are indeed reversely sequenced, and we have revised these sentences. We have changed “During the first period” to “During the period 1 shown in Fig.5” to make it clearer. The “non-fog periods” are changed to “non-fog periods (periods with RH <100%).”

Comment: Page 26, Line 555: How is BBOA identified from COA, as the diurnal patterns of the two factors seem to be quite similar?

Response:

We thank the reviewer for this comment. Although the diurnal profiles of BBOA and COA have some similarities, the mass spectra and temporal variations of the two factors were different (Figure R1). In particular, the BBOA spectrum was characterized by pronounced *m/z* 60, a tracer *m/z* for biomass burning due to fragmentation of levoglucosan (Cubison et al., 2011), and the spectrum of COA showed much higher *m/z* 55/57 ratio that has been widely used as a diagnostic of cooking emissions (Mohr et al., 2012). Because of the spectral differences between BBOA and COA, these two factors can be well separated by positive matrix factorization (PMF).

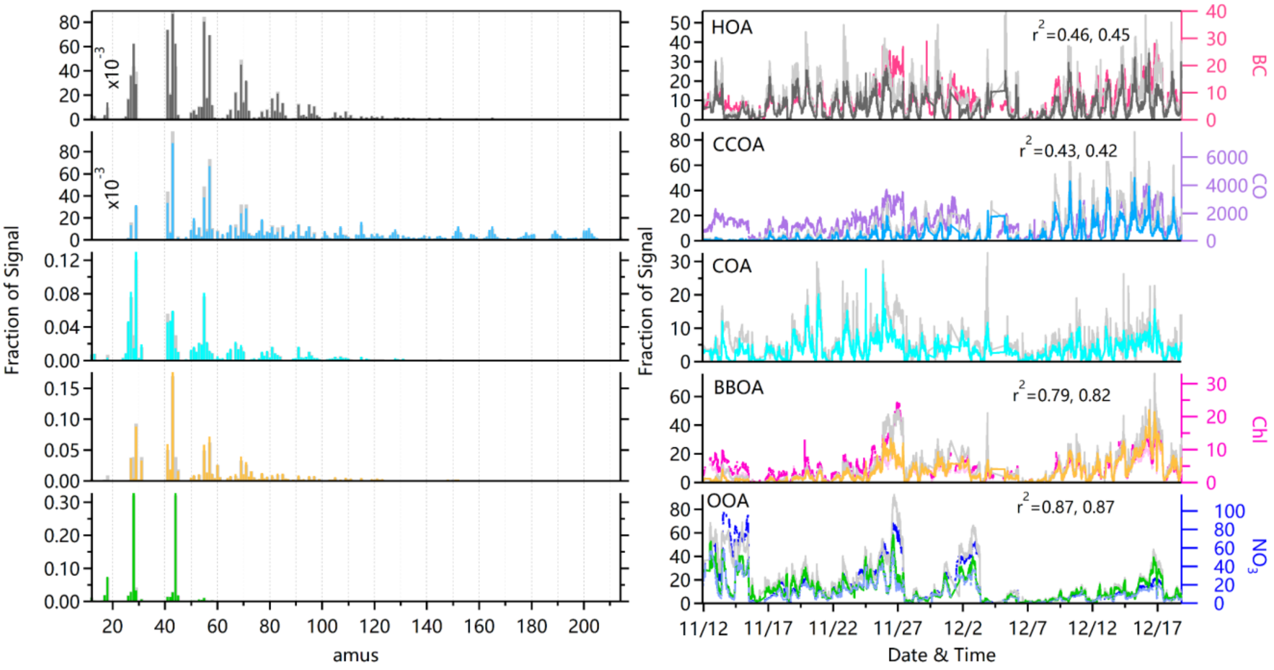

Figure R1. Mass spectra and time series of five OA factors that were identified in this study.

Cubison, M. J., Ortega, A. M., Hayes, P. L., Farmer, D. K., Day, D., Lechner, M. J., Brune, W. H., Apel, E., Diskin, G. S., Fisher, J. A., Fuelberg, H. E., Hecobian, A., Knapp, D. J., Mikoviny, T., Riemer, D., Sachse, G. W., Sessions, W., Weber, R. J., Weinheimer, A. J., Wisthaler, A., and Jimenez, J. L.: Effects of aging on organic aerosol from open biomass burning smoke in aircraft and laboratory studies, Atmos. Chem. Phys., 11, 12049-12064, 10.5194/acp-11-12049-2011, 2011.

Mohr, C., DeCarlo, P. F., Heringa, M. F., Chirico, R., Slowik, J. G., Richter, R., Reche, C., Alastuey, A., Querol, X., Seco, R., Peñuelas, J., Jiménez, J. L., Crippa, M., Zimmermann, R., Baltensperger, U., and Prévôt, A. S. H.: Identification and quantification of organic aerosol from cooking and other sources in Barcelona using aerosol mass spectrometer data, Atmos. Chem. Phys., 12, 1649-1665, 10.5194/acp-12-1649-2012, 2012.

Technical Corrections

Comment: A consistent expression is always recommended in one article, while such inconsistency issues are commonly found in this manuscript. For instance, a subscript format needs to be applied for e.g., (NR-)PM1, (NR-)PM2.5, and PM10. Different symbols like κfRH and κf(RH) are used randomly. OA is defined as the abbreviation for both ‘organic aerosols’ and ‘organic aerosol’. Please check through the whole content and make corrections in all the necessary places.

Some shorten names (such as ACSM, NR-PM) should be defined earlier, i.e., when they appear for the first time.

Response: Corrections have been made according to the suggestions. Subscripts were applied for (NR-)PM$_1$, (NR-)PM$_{2.5}$, and PM$_{10}$. All κfRH were changed to κf(RH), and shortened names like ACSM, NR-PM were defined when they appear for the first time.

Reference:

Atkinson, D.B., Radney, J.G., Lum, J., Kolesar, K.R., Cziczo, D.J., Pekour, M.S., Zhang, Q., Setyan, A., Zelenyuk, A., Cappa, C.D., 2015. Aerosol optical hygroscopicity measurements during the 2010 CARES campaign. Atmospheric Chemistry and Physics 15, 4045-4061.

Kuang, Y., Zhao, C., Tao, J., Bian, Y., Ma, N., Zhao, G., 2017. A novel method for deriving the aerosol hygroscopicity parameter based only on measurements from a humidified nephelometer system. Atmos. Chem. Phys. 17, 6651-6662.

Wehner, B., Birmili, W., Ditas, F., Wu, Z., Hu, M., Liu, X., Mao, J., Sugimoto, N., Wiedensohler, A., 2008. Relationships between submicrometer particulate air pollution and air mass history in Beijing, China, 2004–2006. Atmos. Chem. Phys. 8, 6155-6168.