Supporting Information

Variations of the density of ambient black carbon retrieved by a new method: importance to CCN prediction

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Methods and theory for CCN number concentration prediction

Assumption 1: BC internally mixed

BC particles are assumed to be internally mixed with bulk chemical composition, when using a density of BC with the value of 2.1 g cm^{-3} in the sensitivity test. For this scheme, six species are considered, ie., NH_4HSO_4, $(NH_4)_2SO_4$, NH_4NO_3, POA, SOA and BC. By applying the hygroscopicity parameter κ_{chem} into κ-Köhler relationship (Petters & Kreidenweis, 2007), the critical diameter or activation diameter (D_{cut}) can be obtained at a given supersaturation (S). Thus, the CCN concentration can be predicted by using the critical diameter and particle number size distribution.

The equations used in the estimating N_{CCN} are as follows,

$$CCN_{pre} = \int_{D_{cut}}^{D_{end}} n(\log D_p) d \log D_p \quad (1)$$

where D_{cut} is the critical diameter, D_{end} is the upper size limit of the particle number size distribution (PNSD), n (log D_p) is the function of the aerosol number size distribution.

$$D_{cut} = \sqrt[3]{\frac{4A^3}{27\kappa ln^2 S}}, \qquad A = \frac{4\sigma_{s/a} M_w}{RT\rho_w} \quad (2)$$

Where κ is the hygroscopicity parameter, S is a given supersaturation, M_w is the molecular weight of water, $\sigma_{s/a}$ is the surface tension of pure water, ρ_w is the density of water, R is the gas constant, and T is the absolute temperature.

Assumption 2: BC externally mixed

When using a density of 0.14 g cm^{-3} in the sensitivity test, BC particles are assumed to be externally mixed but other five species are treated as internally mixed, ie., NH_4HSO_4, $(NH_4)_2SO_4$, NH_4NO_3, POA and SOA. The CCN number concentration of the internal mixture is denoted as N_{CCN_In}. The way to retrieve the critical diameter (D_{cut}) is same as the assumption 1. The difference is that the N_{CCN_In} should be multiplied by the volume fraction of the internal mixed particles to get the finally N_{CCN} (Ren et al., 2018). The CCN concentration can be calculated as follows:

$$CCN_{pre\_IN} = \left(\int_{D_{cut}}^{D_{end}} n(logD_p) d\, log\, D_p\right) \cdot VF \tag{3}$$

Where *VF* is the volume fraction of the internally mixed components. The other parameters are same as those presented in Eqs. 1-2.

Assumption 3: aged BC internally mixed but fresh BC externally mixed

In this assumption, the fresh BC and POA are externally mixed but sulfate, nitrate and SOA with the aged BC particles are internally mixed. The mass fraction of internal/aged BC and external/fresh BC are retrieved from 2.2. Similar to the assumption 2, the CCN concentration is calculated by using the critical diameter and the PNSD. And the CCN number concentration also should be multiplied by the volume fraction of five internal species. The equation is the same as Eqs. 3. The other parameters are same as those presented in Eqs. 1-3. By varying the densities of internal and external BC particles, a CCN closure test has been done based on this assumption. Then the optimal density of internal and external BC is obtained when the best estimates of N_{CCN} are achieved.

References

Petters, M. D., & Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmospheric Chemistry and Physics, 7(8), 1961-1971, https://doi.org/10.5194/acp-7-1961-2007, 2007.

Ren, J., Zhang, F., Wang, Y., Collins, D., Fan, X., Jin, X., et al.: Using different assumptions of aerosol mixing state and chemical composition to predict CCN concentrations based on field measurements in urban Beijing, Atmospheric Chemistry and Physics, 18, 6907–6921, https://doi.org/10.5194/acp-18-6907-2018, 2018.

Xie, Y.Y., Ye, X.N., Ma, Z., Tao, Y., Wang, R.Y., Zhang, C., et al.: Insight into winter haze formation mechanisms based on aerosol hygroscopicity and effective density measurements, Atmospheric Chemistry and Physics, 17(11), 7277-7290, https://doi.org/10.5194/acp-17-7277-2017, 2017.

Figures

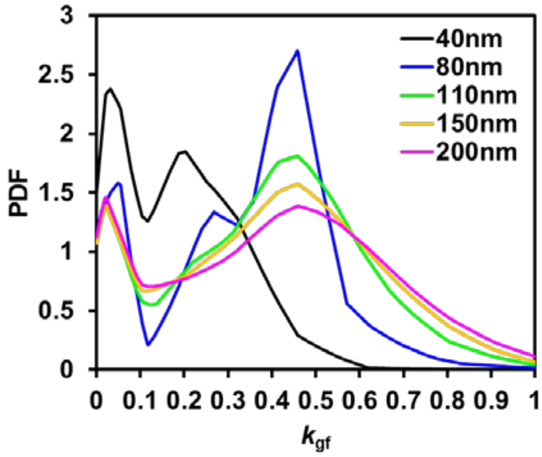

Figure S1. Average *κ*-PDF patterns of particles in different sizes.

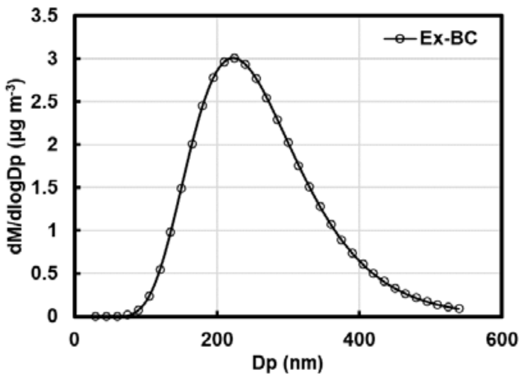

Figure S2. Average mass size distribution of Ex-BC by modeling as a single log-normal distribution.

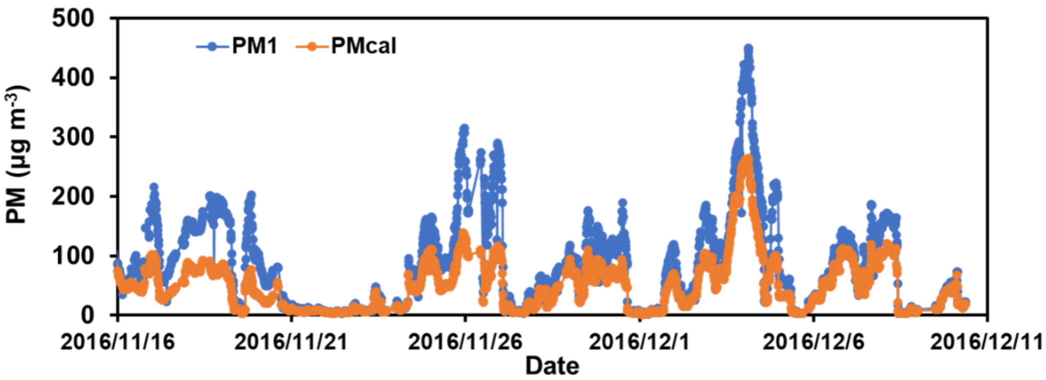

Figure S3. Temporal evolutions of PM_1 concentration (measured with an Aerosol Chemical Speciation Monitor, ACSM and calculated PM_{cal} (measured with a scanning mobility particle sizer, SMPS). The effective density of PM_1 was assumed to be 1.5 g cm^{-3} in the range of 10–550 nm measured (Xie et al., 2017).

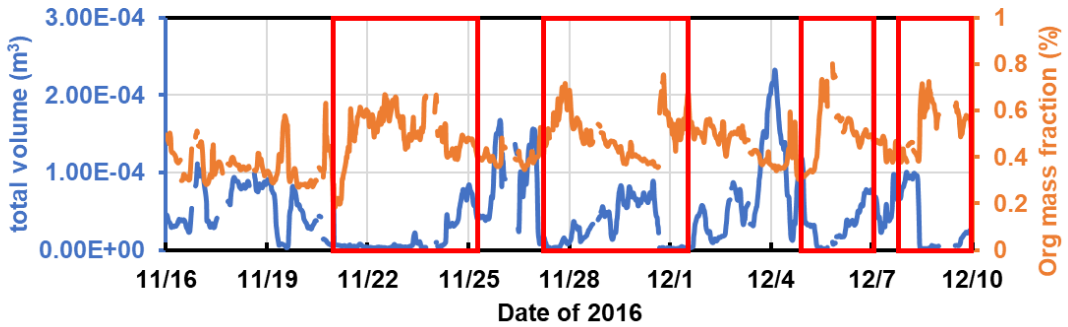

Figure S4. Time series of the calculated total volume of PM_1 and mass fraction of organics.