

Interactive comment on “Aging and hygroscopicity variation of black carbon particles in Beijing measured by a quasi-atmospheric aerosol evolution study (QUALITY) chamber” by Jianfei Peng et al.

Jianfei Peng et al.

pengjianfeipku@gmail.com

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We thank the referee for his/her careful and critical review of our paper. The following are our responses to the referee's comments.

The manuscript investigated the changes in the hygroscopicity of elemental carbon (EC) aerosol during aging in QUALITY chamber where the gaseous species in ambient air constantly diffused into the reaction chamber and products from their photochemical reactions interacted with mono-dispersed EC seed particles. The variations in EC

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particle size, mass, chemical composition, hygroscopicity and CCN properties were monitored throughout the aging processes. This study provides a unique perspective among chamber studies because the EC seed aerosol was exposed to photochemical oxidation products from an environment that closely mimics the actual ambient air. Evaluating the hygroscopicity of EC has been a challenge for the atmospheric sciences research community due to the complex morphology changes. Following the method of using mono-dispersed seed aerosol (Qiu et al. ES&T, 2012 and Khalizov et al. ES&T, 2013), this study provided new insights into this complex research problem. The current manuscript also nicely supplements a recent publication from the same authors (Peng et al. PNAS, 2016) by providing detailed evaluation of the the QUALITY chamber. The characterization of the QUALITY chamber experiments appeared to be thorough and the results appeared to be reliable. The topic is relevant to the scope of the journal of Atmospheric Chemistry and Physics and should be considered for publication. A few comments are provided to facilitate the further improvement of the manuscript: (1) Recent studies showed (for example, Chen et al. Geophys. Res. Lett. 2016, 43, 11080) that the morphology of nascent EC particles can be highly sensitive to even a small change in the organic coating on the primary spheres. As a result, it is prudent to provide experimental data to show that all the EC seed particles started with similar morphology and chemical composition. What was the thickness (or weight percent) of volatile organics in the seed particles at the beginning of each chamber aging experiment?

We thank the reviewer for the suggestion. In our study, the fresh BC particles were highly fractal. For three initial mobility diameters (D_m) of about 100 nm, 150 nm, and 200 nm, the ranges of effective density for fresh BC particles in each experiment are 0.43-0.50 g cm⁻³, 0.34-0.34 g cm⁻³, and 0.24-0.32 g cm⁻³, respectively (Table 2 of the manuscript). Accordingly, the dynamic shape factors (DSF) of 100 nm, 150 nm, and 200 nm BC particle are 2.11-2.30, 2.54-2.55 and 2.44-2.85, respectively. This suggests the highly fractal morphology of the fresh BC particles, and the consistency of fresh BC properties in different experiment. Also, as the BC particles formed from

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the burner were heated to 300 °C before introduced into the chamber, most of the organic coatings were removed. The combined measurement of particle size distribution, density and chemical composition (AMS) shows that organics accounted for less than 10% of fresh BC mass concentration. The single scattering albedo (SSA) of fresh BC particles was only 0.1, further confirming the purity of BC particles. We modified two places in the manuscript to make this clear: Line 132, “The combined measurement of particle size distribution, density and chemical composition exhibited that organics accounted for less than 10% of fresh BC mass concentration. Moreover, the single scattering albedo (SSA) of fresh BC particles was only 0.1, further confirming that few organic coatings existed on fresh BC particles.” Line 238, “For three initial D_m of 100 nm, 150 nm, and 200 nm, the ranges of effective density of fresh BC particles in each experiment were 0.43-0.50 g cm⁻³, 0.34-0.34 g cm⁻³, and 0.24-0.32 g cm⁻³, respectively, indicating highly fractal BC aggregates (Zhang et al., 2008). The small variation for particles with the same D_m also demonstrates the consistency of fresh BC morphology in different experiments.”

(2) Evaluating the hygroscopic changes of EC has been a challenge. It has been proposed (for example, Qiu et al. ES&T, 2012) that the hygroscopicity of the coating materials can be measured or estimated to reflect the real hygroscopic growth of the EC particles. The authors may consider incorporating such approach to convert the apparent HGF into the real HGF. Furthermore, I wonder if similar approach can be developed for the CCN data to estimate the real κ values of aged EC particles.

The reviewer made a very good point here. As discussed in the manuscript, the apparent HGF may underestimate the hygroscopicity of BC particles when particles are not spherical shape. We attribute this the major reason of the difference between κ_{CCN} and κ_{HTDMA} in the beginning of experiment. However, due to the fast growth of BC particles in our study, the morphology of BC particles shifted from fractal to spherical shape within a very short time. Therefore, the apparent HGF at the end of experiments can represent real HGF of particles. Besides, in the CCN data processing, the particle

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mass equivalent diameter was used as the particle dry diameter in the calculation of κ_{CCN} value by Equ (10). Therefore, the κ_{CCN} should reflect the real hygroscopicity of BC particles from this point of view.

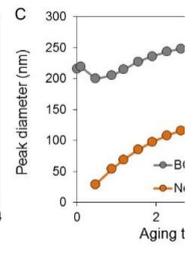
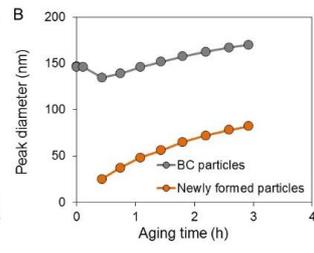
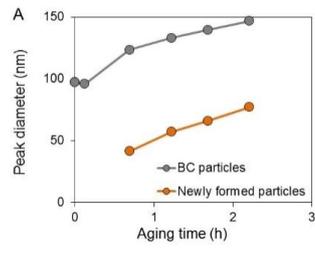
(3) What was the final size of the new particles from nucleation in the chamber? It could be a concern if the nucleation in the chamber was fast and the new particles grew too fast into the size range of the aged soot particles and hence interfere with the property measurements.

Nucleation occurred in the chamber during several experiments of this study. Moreover, the newly formed particles grew along with BC particles inside the chamber. However, as shown in the Figure below, the difference in the peak diameter between BC particles and nucleation particles was always larger than 80 nm for all experiments. Therefore, the influence of nucleation particles to the measurement of BC properties was insignificant.

Figure 1. Peak diameter of BC and nucleation particles inside the chamber during experiments with initial BC diameter of 100 nm (A), 150 nm (B), and 200 nm, respectively.

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