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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.

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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.

In the manuscript the authors measured several physicochemical properties, including size, density, chemical composition, hygroscopicity, and Cloud Condensation Nuclei (CCN) activity of coated Black Carbon (BC) particles as a function of aging (coating with condensable species from photo-oxidation of VOCs) in a quasi-atmospheric

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aerosol evolution study (QUALITY) chamber using a suite of instruments, such as differential mobility analyzer, aerosol particle mass analyzer, High Resolution – Time of Flight – Aerosol Mass Spectrometer (HR-ToF-AMS), Humidified Tandem Differential Mobility Analyzer (HTDMA), and a CCN Counter (CCNC). Their results show that under ambient condition in Beijing, the BC undergoes rapid growth to 77±33 nm coating thickness with an average growth rate of 26±11 nm h-1. The O/C ratio of the SOA coating is 0.5, lower than ambient level, indicating the lack of aqueous phase oxidation inside QUALITY chamber. The hygroscopic parameter is about 0.035 - 0.040 as measured by HTDMA and CCNC, suggesting that the initial photochemical aging of BC particles does not appreciably alter the particle hygroscopicity in Beijing. This study and the data provided are quite extensive. The most valuable addition of this manuscript is the "close to ambient (Beijing)" aging condition in the QUALITY chamber. The author showed in Figure 3 that the O3, NOx, CO, and SO2 concentrations inside and outside of the chamber is close. However, the VOCs plot inside and outside of the chamber is missing. The authors also mentioned they applied heater, drier, alumina spherules coated with potassium permanganate (KMnO4), and activated charcoal to remove the VOCs, H2S, SO2, NOx, and O3. It would be nice if the authors could also show the VOCs are efficiently removed down to sub-ppb level while injecting the BC particles into the chamber. Did the authors use proton-transfer-reaction mass spectrometer (PTR-MS) to monitor the VOCs inside and outside the chamber? Since the coating is caused by condensable species from photo-oxidation of VOCs and "close to ambient" condition is an important part of the manuscript, I would suggest the authors add the VOC plot.

We thank the reviewer very much for the insight suggestion. We have added another figure (as Figure 3 in current version), which demonstrates the comparison of VOCs concentration between chamber and ambient air before the BC aging experiment. Corresponding discussion is also added in the text as (Line 231): "To investigate the VOCs concentration inside the reaction chamber after the injection of BC particles, both the chamber and ambient air were sampled with VOC canisters just before the BC aging experiment started. These canisters were then analyzed by a gas chromatographymass spectrometer / flame ionization detector (GC-MS/FID, HP inc.) system (Liu et al., 2008). The concentrations of VOCs containing 4 or more carbons are illustrated in Figure 3. Slightly higher concentrations of several VOCs in the QUALITY chamber, e.g., n-butane, n-pentane, toluene, were observed compared with those in the ambient air, duo to the co-injection of a small amount of VOCs together with BC particles into the chamber. Nevertheless, the average increase of the VOC concentrations was only 16% or 0.1 ppb for all focused VOCs species, with the largest increase of 35% or 0.36 ppb, suggesting the insignificant influence of soot burner on VOCs concentrations and SOA formation in the chamber. ”

Besides my major point above, I also have several minor comments. 1) On page 1 in abstract (line 18), “wall loss of primary gaseous pollutants was negligible, : : :”, please add “was negligible compared with the replenish rate by gas exchange”. Because other readers who don’t have gas exchange might also think it is negligible in their chamber, which will cause confusion.

We thank the reviewer for this suggestion. The sentence has been modified as suggested.

2) On page 3 (line 100), is “polytetrafluoroethylene” PTFE? PFA should be “Perfluoroalkoxy alkane”.

We are sorry for such mistake. The “polytetrafluoroethylene” should be “Perfluoroalkoxy alkane”. It has been revised in the manuscript.

3) On page 6 (line 207), “The transmission : : : were measured using a Fourier Transform Infrared Spectroscopy (FTIR) system”. However, in the following context, the authors are discussing about the UV-Vis spectrum of the Teflon film and acrylic shell. Did I miss something? Or does the authors actually mean “UV-Vis” system?

We thank the author for pointing out his mistake. The sentence has been revised as “The transmission efficiencies of each material were measured using an ultravioletvisible (UV-Vis) spectrophotometry (PerkinElmer Inc., model 552).”

4) On page 12 (line 444), Fig. 11a and Fig. 11b should be Figure 10a and Figure 10b. Please correct that.

It has been revised. Thanks.

5) On page 13 (line 473, and equation 3), if some of SOA components are not very soluble in water, the updated formula in reference (Petters and Kreidenweis 2008) (formula 10) should be used. Otherwise, based on the kappa of 0.04, the average molecular weight of the coating material is about 450 g mol-1.

We agree with the reviewer that the updated formula in reference (Petters and Kreidenweis 2008) (formula 10) is more accurate to calculation the kappa value when coating material is not highly hygroscopic. However, the calculation requires the solubility of SOA, which is impossible to be obtained duo to the variable VOC precursors and complex photochemical reactions in the ambient condition. Nevertheless, we added discussion on the uncertainty to use of this mixing rule as “Since the SOA formed inside the chamber was not highly hygroscopic, some of the SOA components might not be able to solve in water droplets (Petters and Kreidenweis, 2008), lead to the underestimation of the κ values of the coating materials in this study.”

6) This is not a comment but a discussion. In the end of “Section 4”, the authors discussed about the relationship between _ and O/C ratio. In the reference (Jimenez et al. 2009), Figure 3 shows at around O/C = 0.5, the _ is about 0.12 for ambient (including Mexico City, Jungfraujoch, Hyytilala SV-OOA, Hyytiala LV-OOA) and _-pinene SOA, but about 0.16 for Trimethylbenzene (TMB) SOA. All the cases have larger _ values than reported here (0.04). However, in another reference (Massoli et al. 2010), Figure 2 shows the _-pinene and m-xylene SOAs have _ value about 0.14 at O/C=0.5, but TMB SOA has _ value of only 0.04, consistent with the value (0.04) reported here. If we believe the latter reference is correct, does that mean the VOC source in Beijing is more TMB like? O/C could not be used as the only parameter for predicting _ as

discussed. This could also suggest the VOC source in Beijing is different from other places, such as Mexico City.

The reviewer made a very interesting point to examine the application of this study. However, we don't have enough evidence to draw such conclusion yet. First, "The O/C ratio of the coating SOA is 0.5 in our experiment, corresponding to the O/C ratio of approximately 0.4 in Jimenez et al. (2009) and Massoli et al. (2010) duo the utilize of updated AMS calibration method in this study." The κ value at O/C=0.4 will be lower than that provided above by the reviewer. Second, the κ_HGF of α-pinene and α-pinene/xylene mixture is consistent with our κ_HGF at same O/C ratio. Third, in the measurements at other sites around the world, e.g., Mexico City, Jungfraujoch, Hyytilala, the measured κ value of organics were for the total OM in the atmosphere, including both primary and secondary OM with different aging degree (from hours to days) through both photochemical and heterogenous pathways. In this study, however, the SOA is mainly from photochemical reactions with aging time scale of only a few hours. Therefore, we can't conclude that the SOA hygroscopiciy in Beijing is different from other cities at the moment. But we believe that this is an interesting aspect and we would like to focus this in our future studies. âĂČ

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