This contribution presents size-dependent volatility properties of urban aerosols in Beijing during summer and winter time using VTDMA. This work tries to calculate and compare the number concentration of In-BC, Ex-BC and Non-BC particles from the particle volatility distribution after heating up to 300 °C for aerosols under the studied two seasons. In addition, the extent of aging of BC particles was characterized based on current datasets. Although the comprehensive dataset presented in the manuscript is interesting and may carry certain values for the scientific community, the major conclusion separating In-BC, Ex-BC and Non-BC seems to be poorly supported by current analysis. The mixing state of BC particles derived from VTDMA data is highly uncertain or even not correct, and the uncertainties were not carefully analyzed in the manuscript. I therefore do not recommend publication in ACP.

Major comments:

The major results of current manuscript were based on the assumption that in urban areas refractory component in sub micrometer aerosols consists mainly or solely of BC. This assumption might not be true that there are some other important non-volatile aerosol compounds in submicron aerosols, for instance, some extremely low-volatility organic material that does not evaporate even at 300 °C . Nowadays, too many studies (Cappa & Jimenez, 2010; Häkkinen et al., 2012; Poulain et al.,2014; Wang et al., 2017; Zhang et al., 2016) confirmed that besides BC, low-volatility oxygenated organic aerosols also existed in the non-volatile fractions in particles. Specifically, Xu et al. (2016) measured the chemical composition of PM1 after heating at a rural site (Detling, Kent) and found at 250 °C, OA has the largest contribution (~ 40 %) to the residual mass. Hence, based on aerosol volatility properties, you may not be able to characterize the mixing state of BC particles.

Then, it comes with my second comment regarding the method this manuscript used to category In-BC, Ex-BC and Non-BC particles as given in line 146-149. Even though this method has been used in previous studies as in Wehner et al. (2009) and Cheng et al. (2012), it is actually out of date and not recommended to be employed to classify the mixing state of BC-particles. With VSF > 0.82, ambient particles could be composed of only BC, but also In-BC coating with non-volatile organics. Therefore, using current classification, significant uncertainties may be introduced into the estimation of the number fraction of different BC-containing particles. We now rely on some other techniques, for instance, SP2, VTDMA-SP2, SP-AMS to study the mixing state of BC-containing particles.

Moreover, in line 161-162, you concluded your results were reliable for deriving the mixing state of BC by comparing with SPAMS results. However, Fig.2 in your previous publication (Chen et al., 2020) only compared the total BC-containing particles; separation of Ex-BC particles from In-BC particles was not performed. I did not see any other evidence that could support your conclusion here. If you have, please specify.

Large uncertainties using Eq. 1 in your manuscript to calculate completely volatile particles may arise. Transportation efficiency as I understand was determined by volatility measurements of NaCl particles or other non-volatile particles at 300 °C in your study. However, ambient aerosols with different volatility could evaporate differently, resulting in different particle size after heating. Therefore, you have to at least consider the number size distribution of particles after heating into the determination of the transportation efficiency. In other words, number concentration of completely volatile particles should be dependent on the volatility of your ambient particles, thus your Eq.1 should be revised as a function of your VSF or your volatility distribution after heating.

In section 3.3, you studied the effect of the formation and growth of particles on their volatility. However, your discussion and analysis is not thorough. How frequent of NPF events occurred during your studied seasons stated in your manuscript? Did you select the data of the days of NPF events to drive your conclusion, otherwise, how could the influence of other atmospheric processes be neglected? What are the sources of 40 and 150 nm particles? Your 40 nm particles could be newly formed particles or a mixture of newly formed particles after growth with pre-exsiting particles. Your analysis should guide the reader to a clear process level, otherwise your conclusion will be quite difficult for us to sink into. Moreover, are the plots in Fig. 4a-h the average or median values of the whole campaign or of the NPF days? I think Fig. 4g or Fig. 4h in your manuscript present a certain NPF day, how could you compare general patterns with a case event? How could we reader withdraw general information or conclusions by case studies?

In section 3.6, you characterized coating thickness by D_P/D_C ratio. However, I did not find any description or definition about Dp and Dc. You obtained a volatility distribution of particles after heating. How did you get Dc from this distribution? The determination of Dc was quite difficult, even with a SP2, which could accurately measure the mass of BC. Zhang et al. (2016) gave a thorough discussion of the uncertainties associated with determining Dc from SP2 measurements due to the morphology effect. Similar discussion of the uncertainties in determining Dc should be given in the manuscript.

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