

### General Comments:

Chen et al. conducted ambient measurements in Beijing in summer and winter from which they calculated size-dependent volatility shrinkage factors (VSF) and mixing states of urban aerosols and compared the volatility properties in different seasons. The measurements were conducted with the use of a VTDMA of which size-selected aerosols ranging from 40-nm to 300-nm were heated up to 300°C. The non-volatile particles that remained in the particle phase upon heating up to 300°C were assumed to be black carbon (BC) in the analysis. Although volatility analysis of ambient aerosols has been intensively studied in general, this manuscript presents measurement results in different seasons in north China. The dataset presented in the manuscript is overall comprehensive but could be more thorough when interpreting the results.

### Specific Comments:

1. I have similar concerns about the major assumption of attributing the non-volatile composition in urban aerosols to be BC in your analysis, as already detailed by another referee. Please consider providing more information, such as chemical composition data, or data from other instruments, if available, to support your assumption, which is critical to your analyses and discussions thereafter.
2. Since the manuscript aims to characterize the volatility properties of urban aerosols and tries to link the properties to the source, formation and growth, the authors may consider adding more materials to enrich the discussion, such as air masses origins information and their effect on the aerosols' volatility and mixing states.
3. It is frequently mentioned throughout Section 3 about the impact of new particle formation (NPF) and the growth on the volatility properties of aerosols. Please consider providing more details, such as the number of NPF events in summer and winter, respectively, to give a clearer picture and support to your analysis. Furthermore, although NPF events occurred less frequently in winter, did it have similar impact on aerosols' volatility as that in summer?
4. In line 178 – 181, the authors mention that the distributions of VSF for 150-nm particles were generally unimodal in both summer and winter. However, from Fig. 2(e), it seems that 150-nm particles were generally bi-modal with a non-volatile mode and a high-volatile mode in winter.
5. Section 3.3 compares the diurnal variation of particles volatility between summer and winter based on the mean VSF and VSF probability distribution function (VSF-PDF) as illustrated on Fig. 4, yet I was lost from line 229 to 236 when the number fraction of the low-volatile mode is discussed. Is the discussion still based on Fig. 4 or other figures in the manuscript?

6. Fig. 5(a) presents the time series of the number concentrations of Non-BC, In-BC and Ex-BC 150-nm particles in summer and winter. While this work shall be the same as that presented in their previous publication (Chen et al., 2020), the number concentration of 150-nm particles in this manuscript seems to be different from that on Fig. 5(a) in Chen et al. (2020). The scale also looks different from that of other sizes as shown in Fig. S4 in the Supplement.
7. In line 285 – 288, the authors state: “non-BC concentration / fraction in winter exhibits a daily minimum and nightly maximum”. It is not clear to me whether this observation is supported by Fig. 7. For example, for 40-nm particles, there seems to be a morning peak at 08:00 LT for non-BC concentration in winter. For 150-nm particles, I am not sure whether there was any significant diurnal cycle for non-BC concentration / fraction. Please further elaborate to support your analysis.
8. The analysis method for the ratio of BC diameter discussed in Section 3.6 should be added in Section 2.

Reference:

Chen, L., Zhang, F., Yan, P., Wang, X., Sun, L., Li, Y., Zhang, X., Sun, Y., and Li, Z.: The large proportion of black carbon (BC)-containing aerosols in the urban atmosphere, *Environmental Pollution*, 263, 114507, <https://doi.org/10.1016/j.envpol.2020.114507>, 2020.