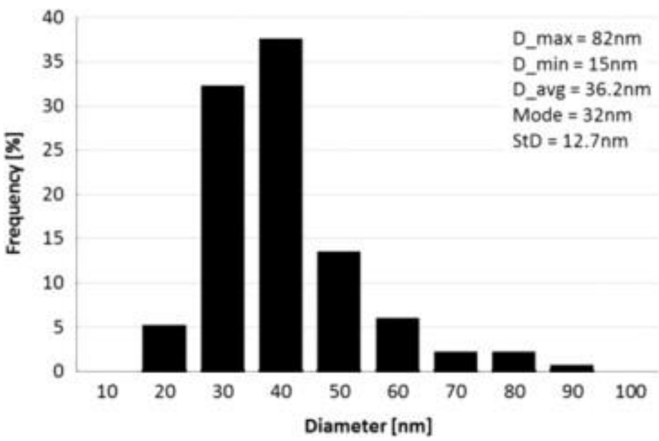
We thank the reviewer for providing insightful comments and helpful suggestions that have substantially improved the manuscript. Below we have included the review comments in black followed by our responses in blue. In the revision of this manuscript, we have highlighted those changes accordingly.

I would like to thank the authors for addressing most of my minor comments. Nevertheless there are still some major comments that have not been adequately considered.

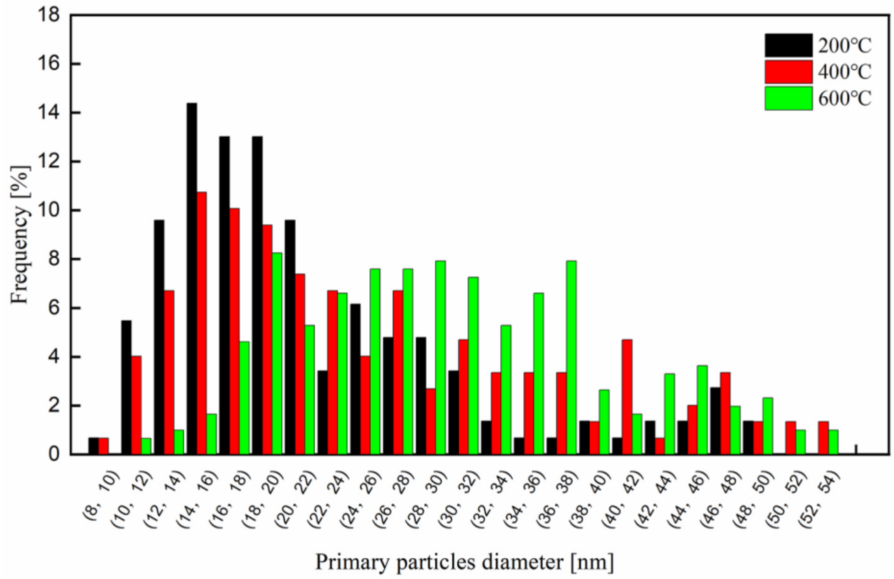
Lines 281-283 read "Figure 4a-b shows that VV-mode fractions in the SF-PDFs of 40-nm and 80-nm particles were higher in warm months than in cold months, indicating that nucleation-mode soot particles were more volatile in warm months"

The figure clearly shows that the fraction of very volatile nucleation mode particles was higher in the warmer months, but the authors should explain how they concluded that the very volatile particles in 40 and 80 nm range contain soot.

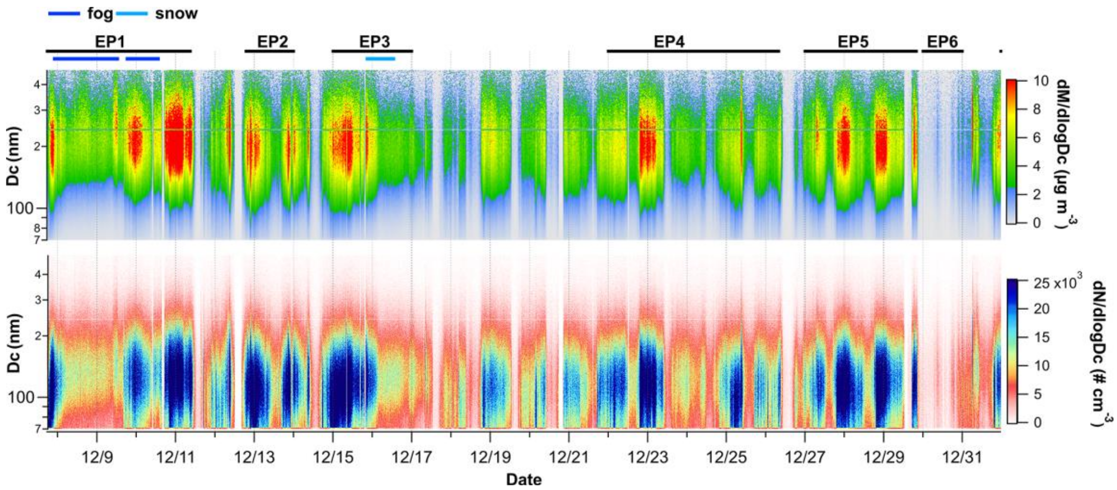
RE: The phenomenon of soot formation can be essentially described in terms of three steps: nucleation, growth and oxidation (Clague et al., 1999). The diameter of initial soot particles is lower than 2 nm. Surface growth, coagulation and aggregation make soot particles grow to tens or hundreds of nm. One method to measure soot particles is using the transmission electron microscopy (TEM). The TEM observations suggest that modern gasoline direct injection (GDI) can emit plentiful soot particles with a diameter smaller than 100 nm (e.g., La Rocca et al., 2015; Amin et al., 2019; Hu et al., 2021). For example, La Rocca et al. (2015) reported that primary soot particles from the emission of light-duty EURO IV GDI engine presented an average diameter of 36 nm with a mode of 32 nm (shown in the first figure below). Hu et al. (2021) sampled particle matter emitted from a China VI vehicle over the Worldwide harmonized Light vehicles Test Cycles (WLTC) and they found that ultrafine soot particles in diameter below 23 nm take up majority of particle emissions (shown in the second figure below). All this reflects that there are many ultrafine soot particles emitted from modern vehicles. Zhang et al. (2021) presented the size distribution of refractory black carbon (*r*BC) above 70 nm measured by SP2 at a site in the NCP (shown in the third figure below). From the figure, it is expected that there are many *r*BC below 70 nm in the ambient although the measurement is limited by the measurement size range of SP2.



The measured soot particles diameter distribution from light-duty EURO IV GDI engine (La Rocca et al., 2015)



The measured soot particle diameter distribution in different oxidation temperature from China VI GDI engine (Hu et al., 2021)



Time series of the mass size distribution and number size distribution of rBC, as measured by the SP2 at the Gucheng site in the NCP (Zhang et al., 2021).

Line 283-287. The authors reported that NPF events are frequent at the site. The growth of new particles to the size range of 40 and 80 nm is controlled by condensation. Nevertheless, condensation on newly formed particles would be in competition with condensation on soot particles (Matsui et al., 2021). The sentence "All this implies that coating by newly formed secondary matter was the possible reason for the high volatility of nucleation-mode soot-containing particles in warm months. " does not seem correct. Again, the authors need to demonstrate that the very volatile nucleation mode particles contain soot before discussing the high volatility of nucleation-mode soot particles.

RE: NPF events occurred frequently in the North China Plain due to the high concentration of gaseous precursors and strong atmospheric oxidation capacity (Wang et al., 2017; Wang et al., 2018). Secondary matters such as sulfate were produced during NPF events. The mixing pathways of soot with other material are various, including condensation, coagulation and so on. The newly formed particles during events are easily captured by soot particles due to their irregular shapes. Li et al.

(2011) indicated that the tiny soot particles embedded in sulfates could promote particle growth during NPF events, which is based on the TEM observations in the NCP.

Finally, particles smaller than 50 nm after removal of condensed material are less likely to be soot, because primary soot particles are generally larger than 50 nm (Harris 2001). On the other hand, smaller particle containing very volatile molecules are likely composed by non-volatile organics (Kalberer et al., 2004; Wehner et al., 2009). The high mass concentration of BC at the site is not enough to demonstrate that the non-volatile material observed at the site is composed by soot.
RE: Harris and Maricq (2001) reported the size distribution for diesel and gasoline engine exhaust particulate matter from different engine classes: diesel, direct injection gasline, and port fuel injection gasoline. Sine then the engine has been greatly upgraded. Hu et al. (2021) indicated that modern gasoline direct injection (GDI) offered higher power output, improved fuel economy and reduced CO_2 emissions compared with port fuel injection (PFI) vehicles. However, the particle number emission of GDI is larger than that of PFI and also higher than diesel vehicles equipped with diesel particle filter (DPF). The GDI produced more ultrafine soot particles shown in the figures above. Kalberer et al. (2004) characterized the volatility of organics based on the measurement at the maximum temperature of 200^{o}C. However, 200^{o}C is too low to vaporize some volatile or semi-volatile material. Thermo-desorption at 250 to 300^{o}C appeared to be the optimum temperature to avoid size dependent effect due to limited residence time in the thermo-desorption unit (Villani et al., 2007). In our measurement, the temperature of VTDMA was set at 300^{o}C. Wehner et al. (2009) indicated that volatile compounds such as sulfates, nitrates, and most of organics species are evaporated at 300^{o}C. Residual particles are either externally or internally mixed BC particles or other nonvolatile material such as sea salt or crustal particles. In heavily polluted areas and for the submicrometer range, the majority of the nonvolatile particle mass can be assumed to be soot. Wehner et al. (2009) also suggested that some nonvolatile material is produced during particle formation and growth in the polluted Beijing region, but usually ~97% of the particle material is volatile at 300^{o}C. On the other hand, 97% of the newly formed particles consists of volatile particle material which is most likely dominated by sulfate but also volatile organic compounds.

Figure 6a and Fig. S4 in our paper suggest that the fractions of non-volatile mode particles (NF_{NV}) increased obviously in the rush hours for ultrafine particles, which can indirectly indicate that a large amount of ultrafine soot particles are from vehicle emissions.

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