

Referee comment to acp-2020-576-RC1

The manuscript by Yu et al. reports one-year concurrent measurement of airborne PAHs at 12 sites across China. Size-segregated PAHs together with typical organic markers are measured to evaluate health risks of PAHs in different size particles and attribute emission sources of PAHs over different regions in China. The finding that toxic PAHs are concentrated in ultrafine particles is particularly interesting. The authors also find that PAH pollution is high in the northern China and nation-widely increases in wintertime, due to the unfavorable meteorological conditions and enhanced emissions of coal combustion and biomass burning. I think this is an important work nowadays in China as well in the global air pollution community. Overall this manuscript is well-organized and well-written and should be accepted after the authors address the minor issues below.

Major comments:

1. The PM samples were collected in 6 regions of China, including urban, sub-urban and remote sites. The authors are suggested to add more comparison of PAH concentrations and compositions among different types of sampling sites.

Reply: Thank you for your suggestion. In the revised manuscript, we add more discussion on \sum_{24} PAHs concentrations, compositions, sources and BaP_{eq} concentration and sources among different types of sampling sites. And Figure 1-4 were added in supporting information file and the revised manuscript as Figure S2, Figure S4, Figure S5 and Figure S6. Figure 5 and Figure 6 was added in the revised manuscript as Figure 7b and Figure 7e. (The line numbers here refers to the 'tracking changes' file)

“The concentrations of \sum_{24} PAHs at urban sites (82.7 ng m⁻³) were significant higher (p<0.05)

than those at sub-urban (48.0 ng m^{-3}) and remote sites (18.0 ng m^{-3}) (Figure 1) (Line 228-229). And BeP_{eq} (Figure 2) and ILCR (Figure 3) were both the highest at urban sites. All these indicated that people in urban regions of China were faced with higher exposure risk of PAHs pollution as compared to those in rural and remote areas. Figure 4 exhibits that 4- and 5-rings PAHs are the majority in $\sum_{24}\text{PAHs}$ at urban, sub-urban and remote sites, which totally accounted 72.2, 63.8 and 66.6% of the total amounts in TSP, respectively. The percentage of 5-rings PAHs dominates at urban sites, and 4-rings PAHs makes the largest proportion at sub-urban and remote sites (Line 244-250). PMF result showed that at urban and sub-urban sites coal combustion was the largest source of $\sum_{24}\text{PAHs}$ (70.4 ng m^{-3} , 85.1% and 30.5 ng m^{-3} , 63.5%), followed by biomass burning (10.1 ng m^{-3} , 12.2% and 16.3 ng m^{-3} , 33.9%) and vehicle emission (2.2 ng m^{-3} , 2.6% and 1.2 ng m^{-3} , 2.5%), while at remote sites the contributions of coal combustion (9.1 ng m^{-3} , 50.6%) and biomass burning (7.8 ng m^{-3} , 43.7%) were comparable and vehicle emission (1.0 ng m^{-3} , 5.7%) had minor contributions (Figure 5) (Line 410-415). Coal combustion was the predominated source of BaP_{eq} , and its contribution at urban sites (8.3 ng m^{-3} and 96.4%) were larger than those at sub-urban (3.3 ng m^{-3} and 90.8%) and remote (1.0 ng m^{-3} and 82.5%) sites. (Figure 6) (Line 426-428)

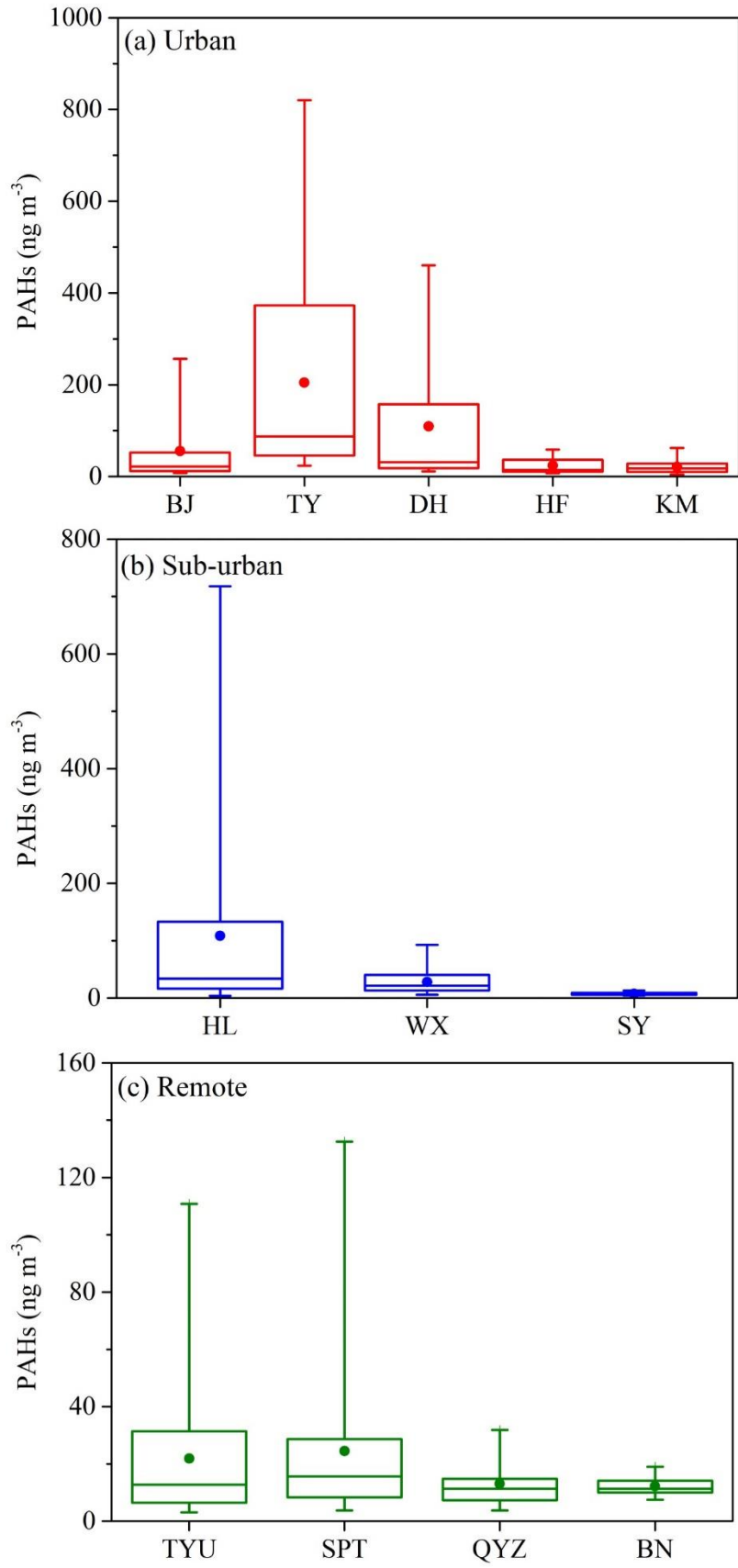


Figure 1 Concentrations of Σ_{24} PAHs at urban, sub-urban and remote sites.

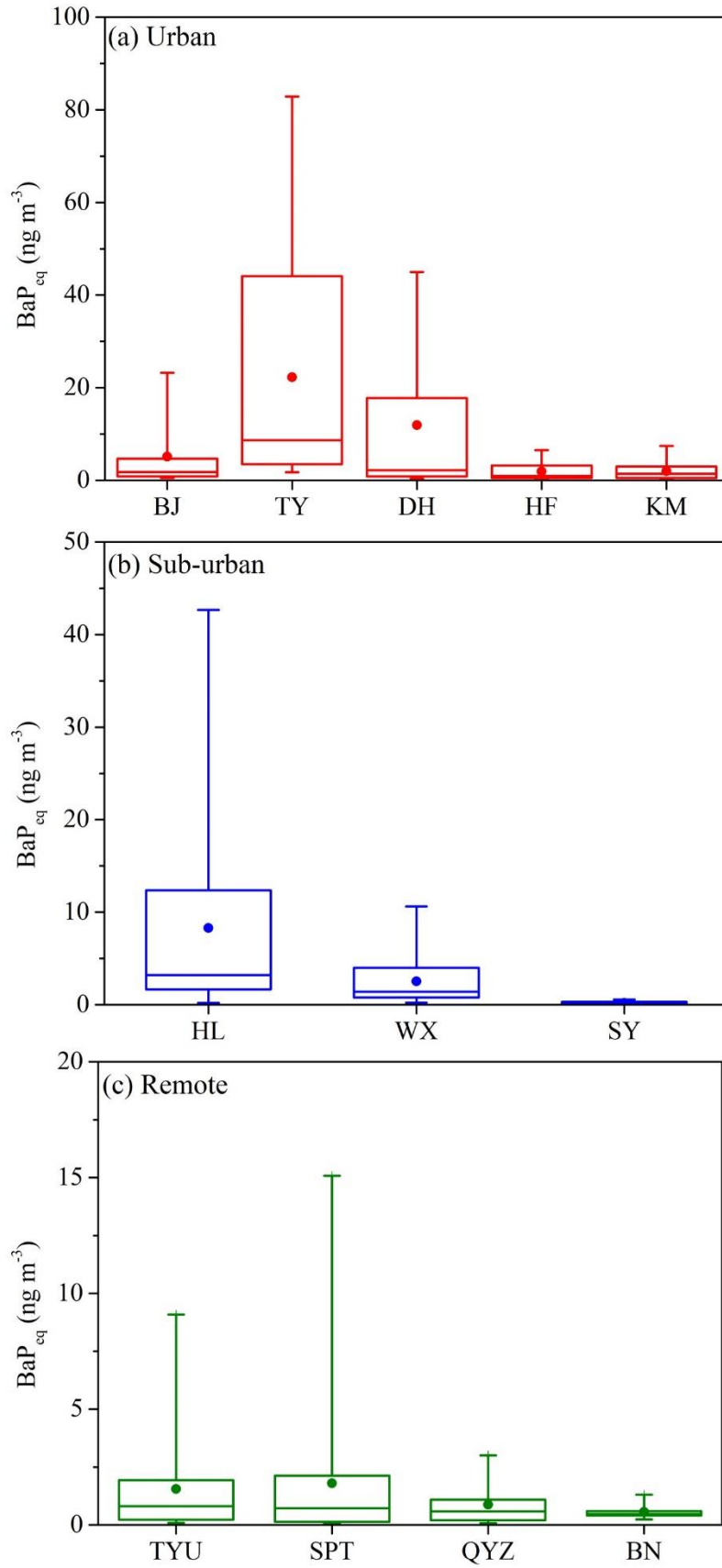


Figure 2 Concentrations of BaP_{eq} at urban, sub-urban and remote sites.

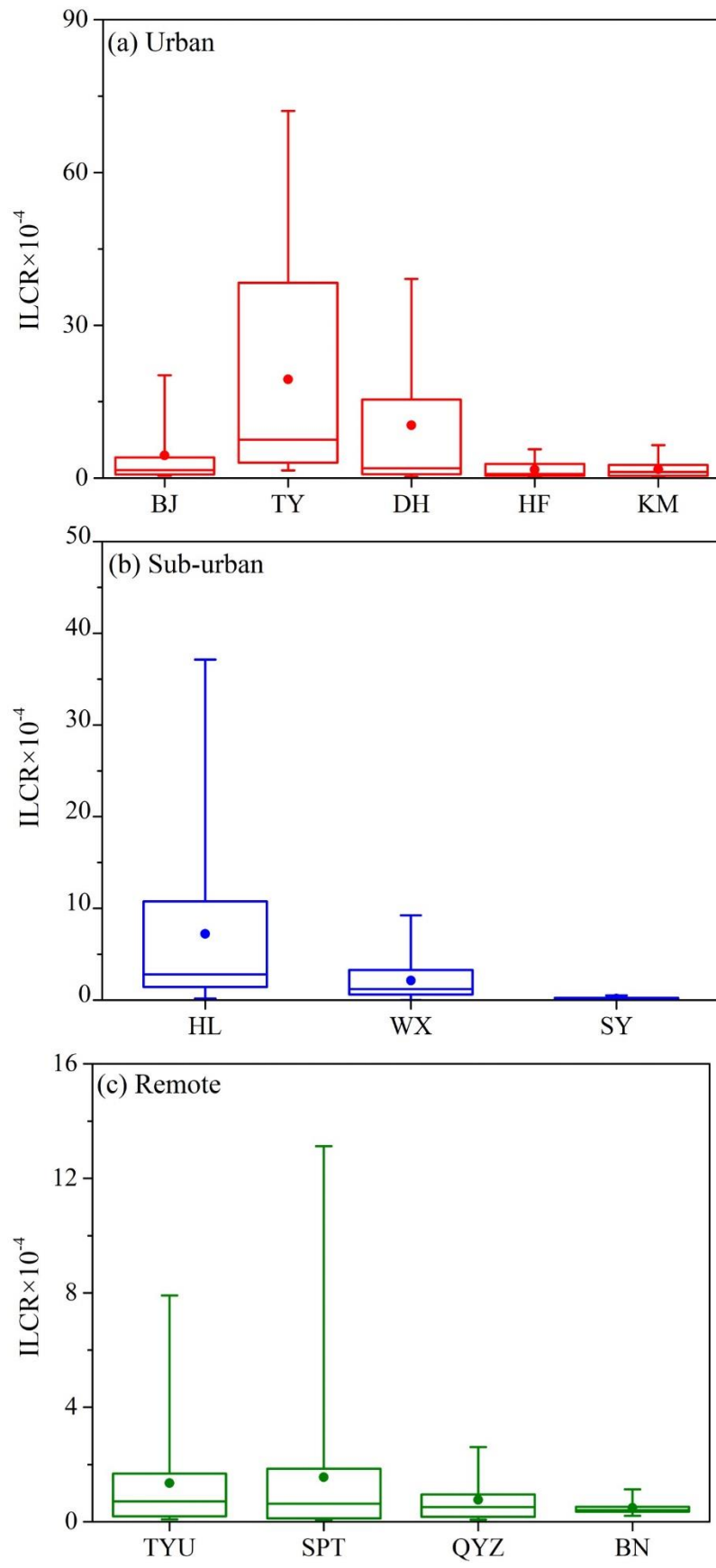


Figure 3 ILCR at urban, sub-urban and remote sites.

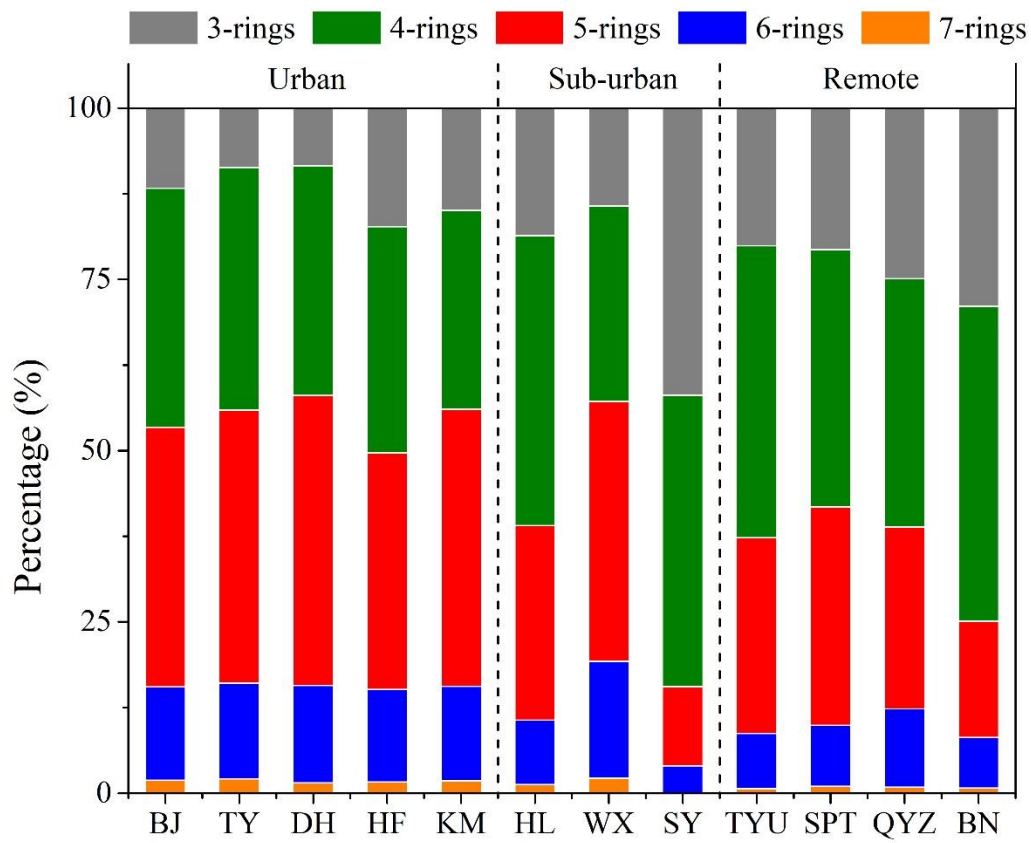


Figure 4 PAHs composition at urban, sub-urban and remote sites.

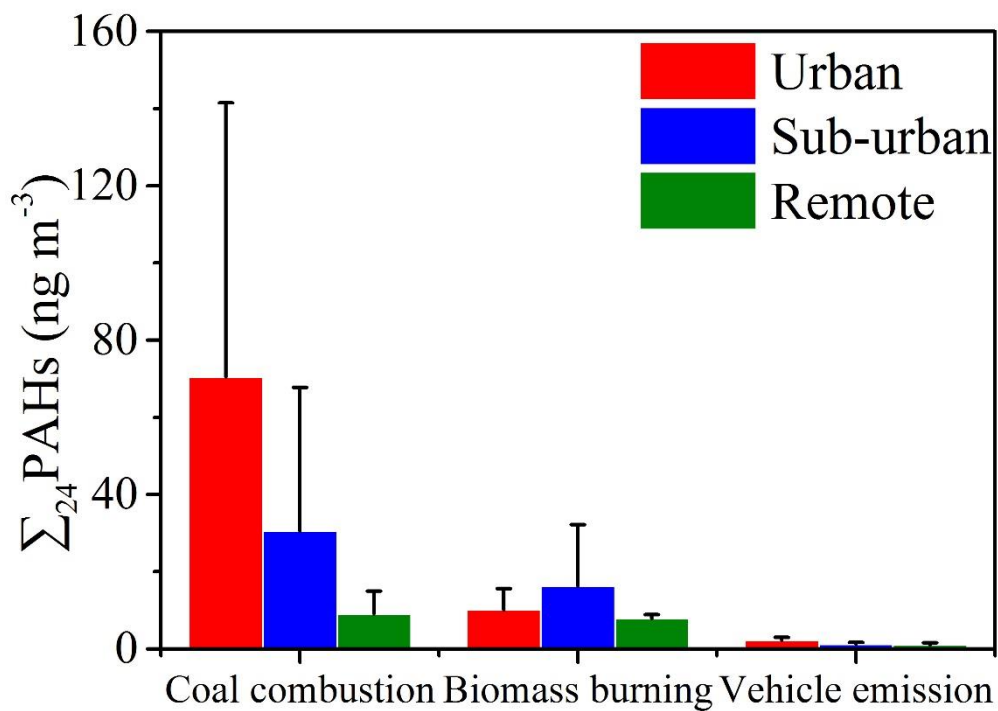


Figure 5 Difference of Σ_{24} PAHs sources at urban, sub-urban and remote sites.

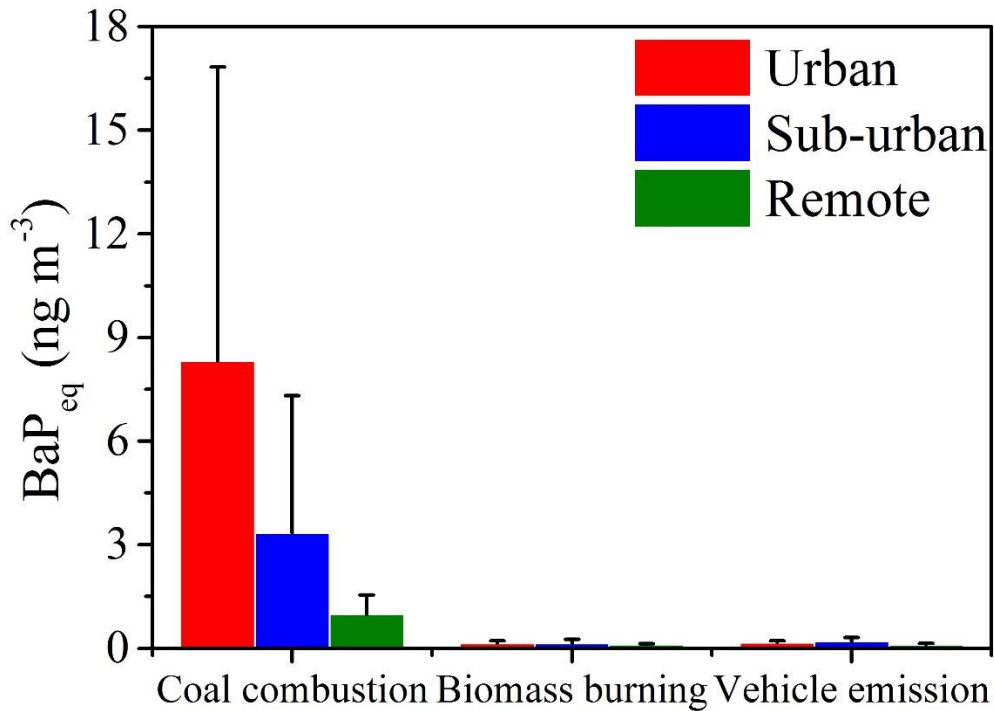


Figure 6 Difference of BaP_{eq} sources at urban, sub-urban and remote sites.

2. As I know, the national standard is not for BaP_{eq} but BaP. The authors should directly compare measured BaP levels with the national standard.

Reply: Yes, the national standard (1.0 ng m⁻³) is for BaP. In the revised manuscript, we directly compare measured BaP levels with the national standard.

“Annual averages of BaP in TSP among the 12 sites were in the range of 0.09 to 11.0 ng m⁻³ with a mean of 2.58 ng m⁻³. The highest level of atmospheric BaP occurred at TY and the lowest existed at SY. The BaP values at five sites (WX, BJ, HL, DH and TY) exceeded the national standard of annual atmospheric BaP (1.0 ng m⁻¹) by factors of 1.2 to 11.0. For BaP_{eq}, annual averages ranged from 0.21 to 22.2 ng m⁻³ with the predominant contribution from 5-rings PAHs (Figure 1b).” (Line 230-235)

Specific comments:

1. Line 52. Replace “associated” to “was associated”.

Reply: Revised as suggested. (Line 57)

2. Line 57. Replace “enriches” to “enrich”.

Reply: Revised as suggested. (Line 62)

3. Line 58. Replace “and” to “which”.

Reply: Revised as suggested. (Line 63)

4. Line 91. Delete “in”.

Reply: Revised as suggested. (Line 96)

5. Line 133. Replace “8h” to “8 h”.

Reply: Revised as suggested. (Line 144)

6. Line 146. Replace “3.3 μm ” to “3.3 μm ”.

Reply: Revised as suggested. (Line 161)

7. Line 190. Replace “site” to “sites”.

Reply: Revised as suggested. (Line 207)

8. Line 214. The unit is misspelling. It should be “ng m⁻³”.

Reply: Revised as suggested.

9. Line 259. Replace “high” to “higher”.

Reply: Revised as suggested. (Line 294)

10. Line 264. The abbreviation of boundary layer height is “BLH”. Please replace “BHL” to “BLH” throughout the manuscript.

Reply: Revised as suggested. (Line 307, Line 309, Line 311, Line 316, Line 446, Line 788)

11. Line 281. Replace “within each northern region” to “within each region in the northern China”.

Reply: Revised as suggested. (Line 330-331)

12. Line 299. Replace “high” to “higher”.

Reply: Revised as suggested. (Line 349)

13. Line 306-308. The sentence “This is also confirmed by the significant correlations of \sum_{24} PAHs with the biomass burning tracer, levoglucosan, the coal combustion tracer, picene, and the vehicle exhaust tracer, hopanes at most sites.” should be re-phrased to “This is also confirmed by the significant correlations of \sum_{24} PAHs with the typical tracers of biomass burning (levoglucosan), coal combustion (picene) and vehicle exhaust (hopanes)”.

Reply: Revised as suggested. (Line 356-357)

14. Line 314. Replace “biomass tracer” to “biomass burning tracer”.

Reply: Revised as suggested. (Line 367)

15. Line 338-340. Provide the full words for the abbreviation “SCE”.

Reply: Revised as suggested. (Line 391-392)

16. Figure 8. Please illustrate in the figure caption that the black dot-line represents the ILCR.

Reply: Revised as suggested.

17. Table S4. Please add a line in the table to distinguish the sites in the northern China and the southern China.

Reply: We revised Table S4 to distinguish the sites in the northern China and the southern China.

18. Figure S11. Please add legend in the figure.

Reply: Revised as suggested.