

Referee comment to acp-2020-576-RC2

This work conducted comprehensive field measurements of PAHs in fine particles at 12 sites in China for one year to investigate the chemical compositions, size-distributions, spatiotemporal variations, as well as the public health risk. In addition, diagnostic ratios and PMF model were applied to quantify the contributions from different sources to PAHs in northern China and southern China, highlighting the significant impacts from coal combustion and biomass burning, especially in winter in northern China. The manuscript is generally well written with clear logic, fluent language, abundant data, and deep analyses. There are some minor comments and suggestions below which are required to address before being accepted.

Specific comments:

1. Figure 1, keep the longitude and latitude of the map in same scale. If possible, try to use the coordinate of latitude and longitude instead of Cartesian coordinate when drawing the whole map of China.

Reply: Thank you for your suggestion. We add the coordinate of latitude and longitude in Figure

1.

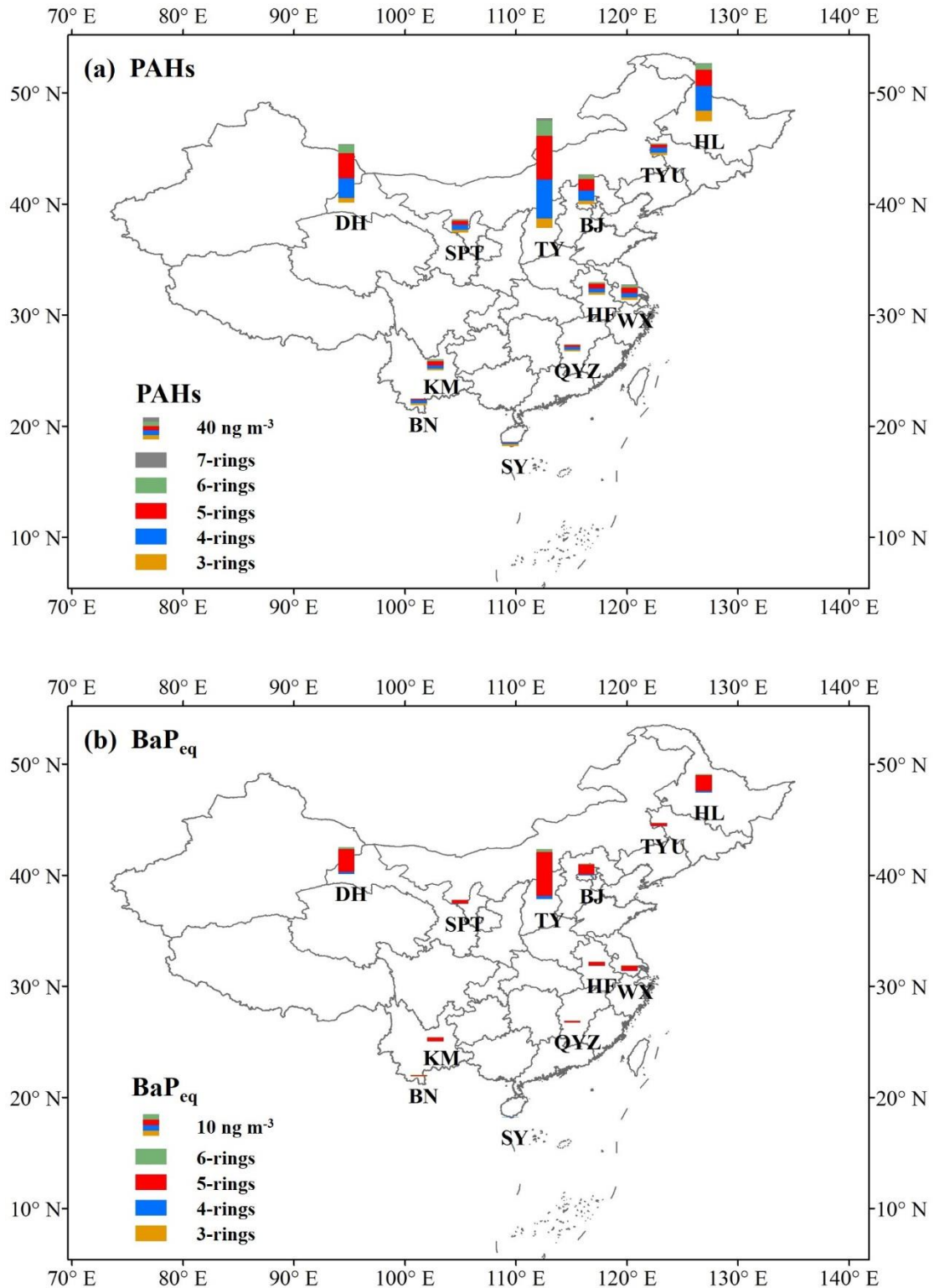


Figure 1 Annual averages of Σ_{24} PAHs (a) and BaP_{eq} (b) at 12 sites in China.

2. Line 91, delete the extra word “in”.

Reply: Revised as suggested. (Line 96) (The line numbers here refers to the ‘tracking changes’)

file).

3. Line 134-135 and Figure 8, state the basis of season division. Why four months are included in summer but only two in autumn?

Reply: Season division is based on consistent annual changes in the weather. According to the meteorological definition, each season lasts three months that spring runs from March to May, summer runs from June to August, fall (autumn) runs from September to November, and winter runs from December to February. In the revised manuscript, we state the basis of season division in the caption and revise the figure. Figure 2 here was Figure 8 in the revised manuscript.

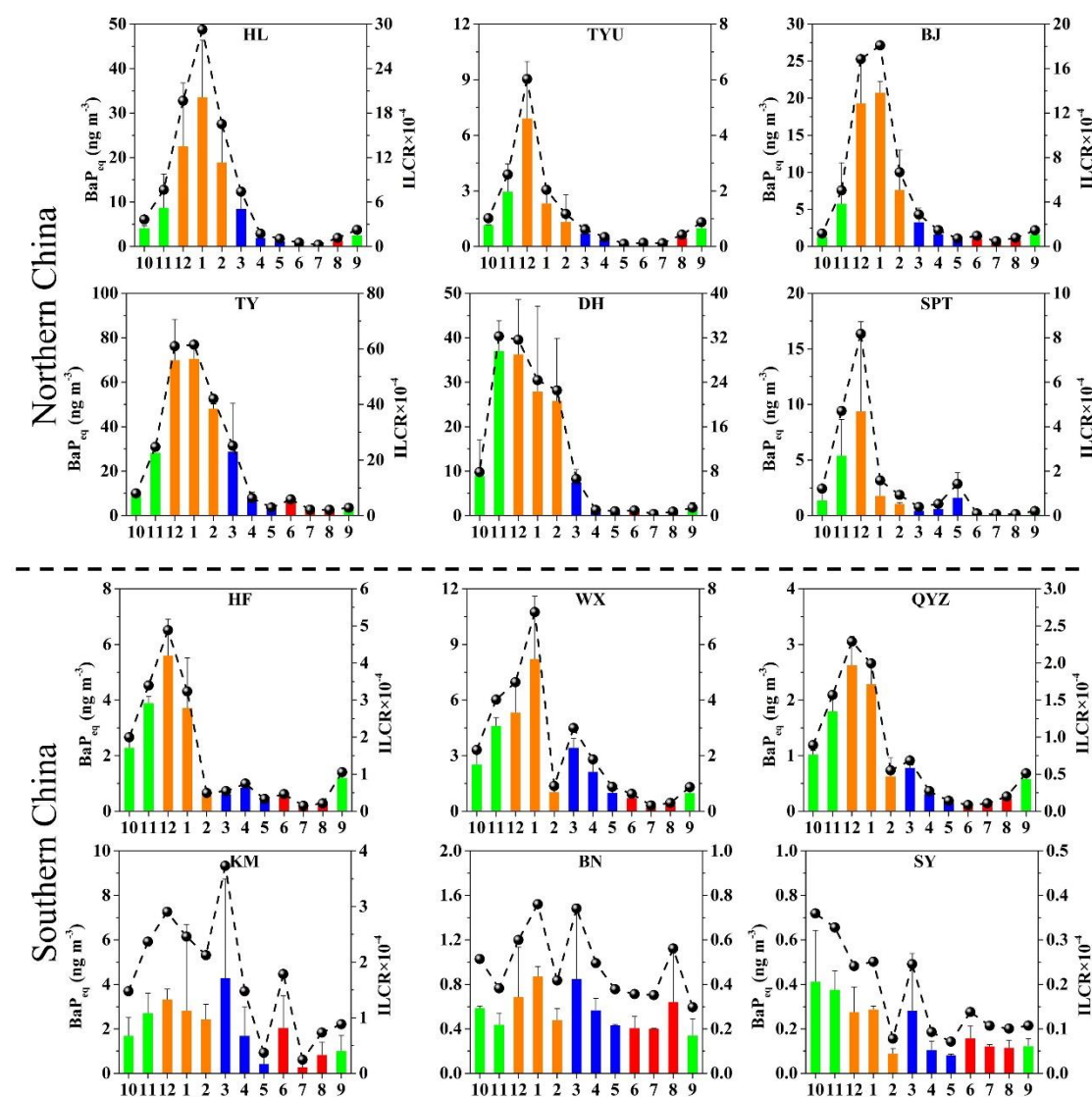


Figure 2 Monthly variations of BaP_{eq} and ILCR at sites in the northern China and the southern

China. The green, yellow, blue and red bars represent BaP_{eq} in fall (October-November, 2012 and September, 2013), winter (December 2012-February 2013), spring (March-May, 2013), and summer (June-August, 2013), respectively. The black dot represented the ILCR.

4. Line 149-153, point out the amount of the added internal standards and the specific extraction method.

Reply: Thank you for your suggestion. In this study, we added 400 µL of internal standards into each sample. The extraction method is ultrasonic solvent extraction.

“Before ultrasonic solvent extraction, 400 µL of isotope-labeled mixture compounds (tetracosane-d50, naphthalene-d8, acenaphthene-d10, phenanthrene-d10, chrysene-d12, perylene-d12 and levoglucosan-¹³C6) were spiked into the samples as internal standards.” (Line 161-164)

5. Line 233-235 and 239-241, why most of the PAHs existed in ultrafine particles and the fractions in ultrafine particles varied with seasons? Is this related to the emission sources?

Reply: Yes, it should be related to the emission sources of PAHs. Atmospheric PAHs are mainly derived from combustion sources. As Shen et al. (2013) reported (see Figure 3 below), PAHs emitted from biomass burning and coal combustion enriched in ultrafine particles (<1.1 µm). Moreover, coal combustion witnessed more enrichment of PAHs in ultrafine particles than biomass burning.

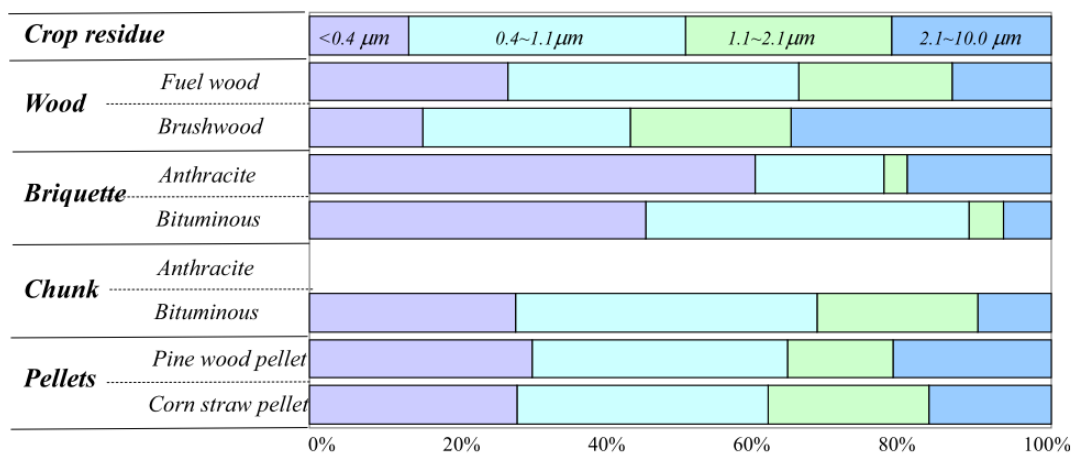


Figure 3 Size distribution of particle-phase PAHs in emissions for different fuels (Shen et al., 2013)

Our PMF results showed an apparently seasonal trend of PAHs sources. For instant, at the DH site, the contributions of coal combustion kept decreasing from winter to summer, while biomass burning kept increasing (Figure 4a). Such a change in PAH sources indeed resulted in the seasonal variations of PAH fractions in ultrafine particles that the mass fractions of Σ_{24} PAHs in $PM_{1.1}$ were the highest during fall to winter and the lowest during summer (Figure 4b).

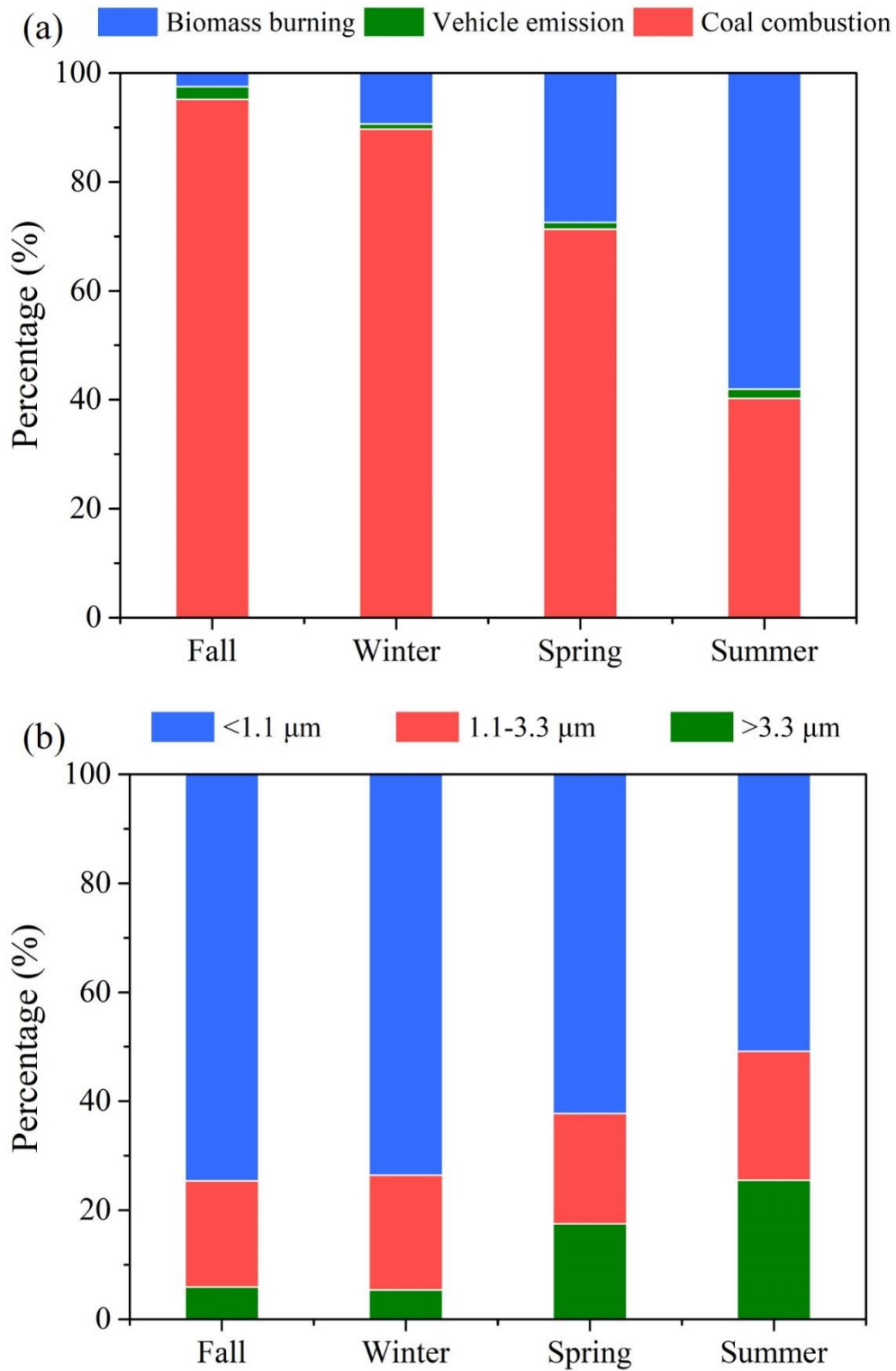


Figure 4 Seasonal variations of PAHs source contributions at DH (a); size distribution of Σ_{24} PAHs in different season at DH.

Shen, G.F., Tao, S., Chen, Y.C., Zhang, Y.Y., Wei, S.Y., Xue, M., Wang, B., Wang, R., Lu, Y., Li, W., Shen, H.Z., Huang, Y., Chen, H., 2013. Emission characteristics for polycyclic aromatic hydrocarbons from solid fuels burned in domestic stoves in rural China. Environ. Sci. Technol. 47, 14485-14494.

6. Section 3.3, is there any difference in the sources and contributions among urban, sub-urban and rural sites?

Reply: Figure 5 and Figure 6 show the difference of $\sum_{24}\text{PAHs}$ and BaP_{eq} sources at urban, sub-urban and rural sites, respectively. At urban and sub-urban sites, coal combustion was the largest source of $\sum_{24}\text{PAHs}$, followed by biomass burning and vehicle emission, while at remote sites the contributions of coal combustion and biomass burning were comparable and vehicle emission had minor contributions. Coal combustion was the predominated source of BaP_{eq} , and its contribution at urban sites were larger than those at sub-urban and remote sites.

In the revised manuscript, we add more discussion on the difference in the $\sum_{24}\text{PAH}$ and BaP_{eq} sources among urban, sub-urban and rural sites in [Line 410-415](#) and [Line 426-428](#). Figure 5 and Figure 6 was added in the revised manuscript as Figure 7b and Figure 7e.

“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)

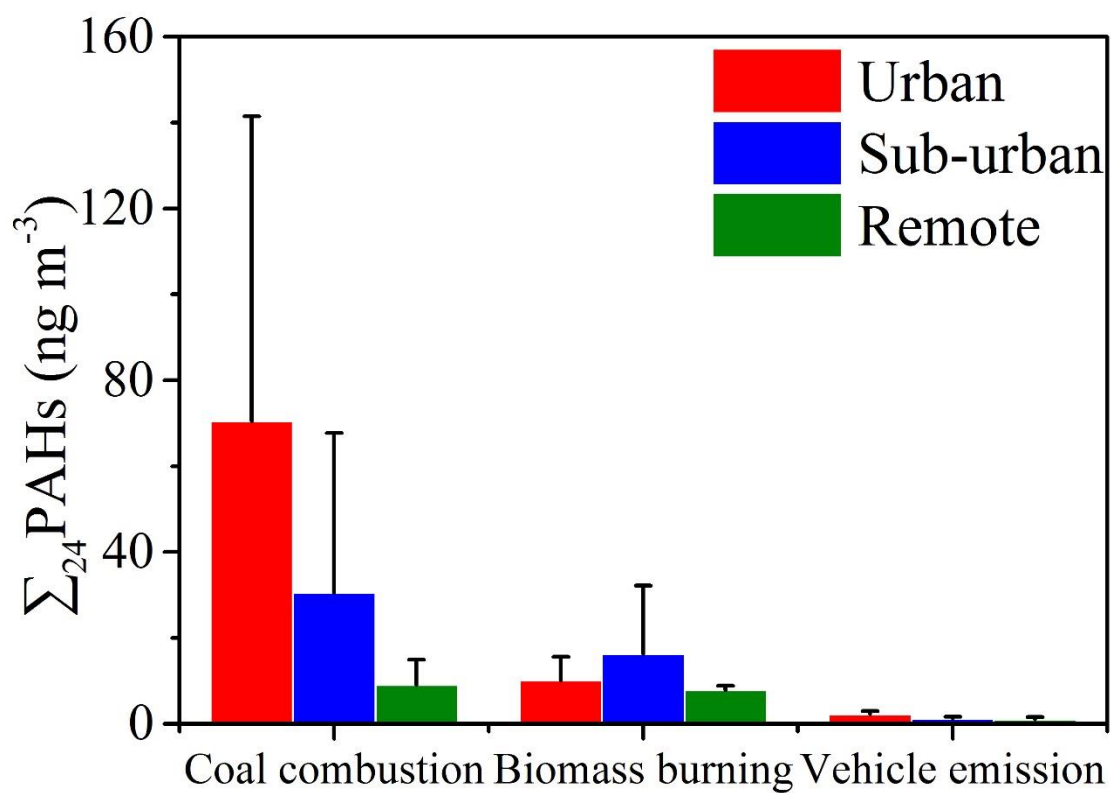


Figure 5 Difference of Σ₂₄PAHs sources at urban, sub-urban and remote sites.

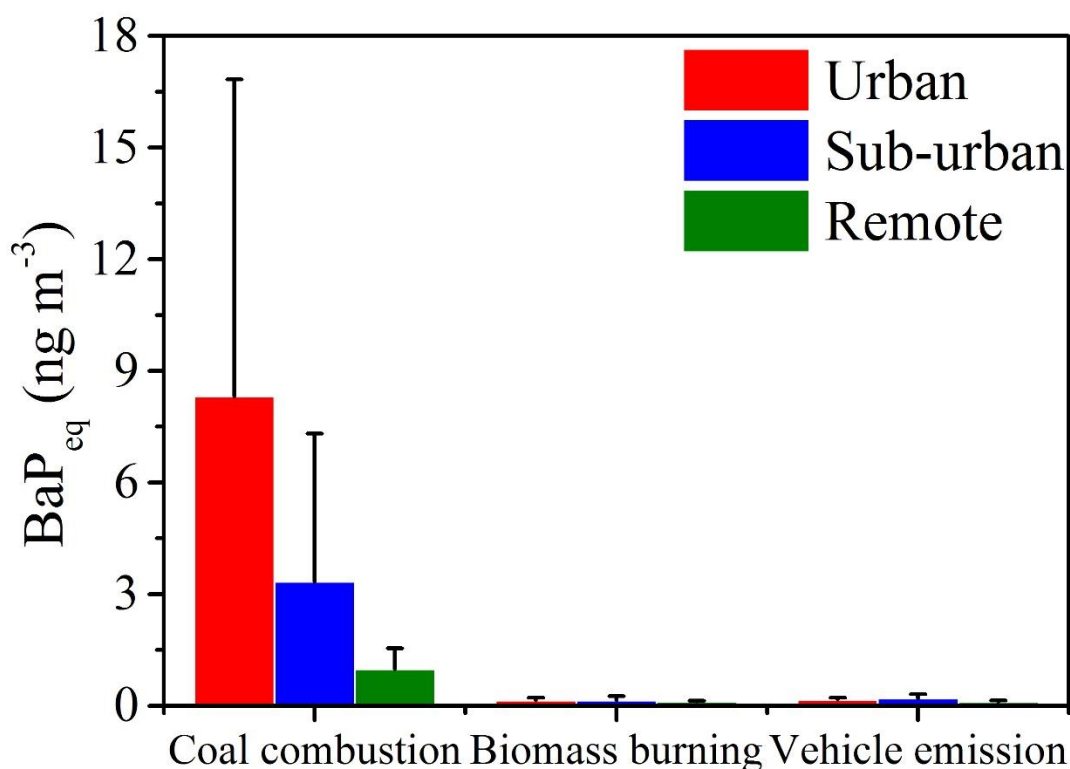


Figure 6 Difference of BaP_{eq} sources at urban, sub-urban and remote sites.

7. Line 336-340, the energy consumption data in 2008 from the Statistical Yearbook are not suitable for comparison. The data in 2013 can be used here.

Reply: Thank you for your suggestion. The data in 2013 from the Statistical Yearbook was used in the revised manuscript. (see below)

“As China statistics yearbook recorded (<http://www.stats.gov.cn/english/Statisticaldata/AnnualData/>), coal was the dominant fuel in China, accounting for 70.6% (24.1×10^8 tons of Standard Coal Equivalent, SCE) of total primary energy consumption (34.1×10^8 tons of SCE) in 2012, followed by crude oil 19.9% (6.7×10^8 tons of SCE) and other types of energy 9.5%, including biofuel, natural gas, hydro power, nuclear power and other power (3.2×10^8 tons of SCE).” (Line 390-395)

8. Figure 8, it's better to remove the repeated ordinate title of the middle graphs.

Reply: Revised as suggested.