Supplementry Material (S1)

Profile of atmospheric PAHs in Rawalpindi, Lahore and Gujranwala districts of Punjab province (Pakistan)

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1. Models/parameters used in the carcinogenic risk assessment

The concentration of benzo(a)pyrene equivalent (BaP. Eqv) were determined using the TEF as reported by Nisbet and LaGoy (1992); USEPA (Nisbet and LaGoy 1992; US EPA 1989; 2009, 2010) and Malcom and Dobson (1994). The inhalation risk of 12 priority parent PAHs (based on available relative potency factors) measured in air samples using the BaP equivalent (BaP_{eq}) concentration was calculated by multiplying 12 individual PAHs with corresponding relative potency factors (RPFs) (U.S. EPA 2010). The risk based on the USEPA point-estimated approach. The risk from the inhalation exposure to PAHs was a product of BaP_{eq} and unit risk of BaP (UR_{BaP}) exposure, there are 2 commonly used UR_{BaP} values, which represent 1.1 case of cancer per 1000,000 individuals over the period of 70 years (i.e. 1.1×10^{-6} determined in a rodent study OEHHA 1993; 2005) and 8.7 cases per 100,000 individuals over the period of 70 years (8.7×10^{-5} per ng m^{-3}); based on a study of the coke-oven worker reported by world health organization (WHO) 2000.

According to OEHHA, the inhalation unit risk represent the upper (95%) limit possibility of contacting cancer when exposed to 1 ng/m^3 BaP concentration for 70 years lifetime period (OEHHA 1993, 2005).

The CR was calculation using the following formula:

$$\text{Lifetime Lung Carcinogenic Risk (CR)} = \sum_{(i=0)}^{n} (C_{PAHi} \times RPF_i) \times UR_{BaP} \quad (1)$$

Whereas n = number of individual PAHs; C_{PAHi} = number of *i*th PAHs measure in air; RPF*i*= the relative potency factor of *i*th PAH; UR-BaP is inhalation unit risk of BaP,

The probabilistic incremental lifetime cancer risk approach (ILCR) was further employed to evaluate the potential risks from exposure to atmospheric PAHs. ILCR is estimated increase in the lifetime cancer risk due to exposure to a carcinogen during an average (70 years of) life time (Meiners and Yandle 1995). According to the WHO guideline (1987), the ILCR of 8.7×10^{-5} can be calculated for inhalation of 1 ng m^{-3} BaP via contaminate air over 70 years of a lifetime. It is important to note that the risk is also contributed by other congeners of PAHs (Masiol et al. 2012).

The ILCR for adults was calculated from the inhalation exposure to PAHs using the following generic formulae:

$$\text{ILCR (Inhalation)} = \frac{\sum BaP_{eq} \times Ti \times \mathit{Inh}\,R \times SF \times ED \times EF \times CF}{BW \times AT} \quad (2)$$

Whereas AT = average life time; (in days); BW = recommended body weight of the exposed person; CF = conversion factor; CSF = carcinogenic slope factor (based on the cancer-causing ability of benzo(a)pyrene); Conc. Dust = concentration of the chemical of interest in dust; ED = exposure duration; EF = exposure frequency; *Inh*R = inhalation rate; ΣTEQ = sum of BaP equivalent concentrations of PAHs. The values and parameters are based on the risk assessment guidance of U.S. EPA and related publications.

1.1. Excessive lung carcinogenic risk

We used point-estimated approach to evaluate excessive lung carcinogenic risk (ECR) assuming the additive approach. This approach is widely accepted for the assessment of carcinogenic risk posed by the inhalation exposure to PAHs (OEHHA 2003). The ECR is a product of sum total of BaP_{eq}-PAHs and the UR_{BaP}. BaP is carcinogenic and mutagenic PAH congener; due to its confirmed carcinogenic properties, the European Union (Directive 2004/107/CE) has established target atmosphere BaP value of $1 ng/m^3$ (San Jose et al. 2014).

The ECR in this study, is representative of 12 priority PAHs (Ant, BaA, BbF, BkF, BghiP, Chry, DBA, Fla, IP, Phe, Pyr, BaP) excluding other congeners of PAHs due to lack of availability of relative potency factors (RPF); however, the concentration of those PAHs was also detected in the study areas, and shows that they could pose additive effects and thus can have significant contribution to ECR.

According to the, UR values proposed by the WHO; the excessive cancer risk from the inhalation of the airborne mixture of PAHs was relatively high in Lahore and Wazirabad, hence the average 56.6 cases per million (ranging from 31.3 to 74.9 per million) and average 51.6 case per million (ranging between 51.1 and 57.7 respectively) in these areas could be attributed to the PAHs exposure. Besides the lowest ESR values observed in Sohdra (9.3 cases per million), the ECR was almost similar in Islamabad, and Gujranwala cities. The ESR values evaluated according to the UR values specified by California Environmental Protection Agency (Cal. EPA) (OEHHAe. 2003) revealed almost a similar trend; however the risk in terms of the number of subjects per millions was low (Figure S5). The higher ECR values in the Lahore and Wazirabad reflected the use of lesser clean fuels, pollution from petrochemical sources, and exhaust emission from the road traffic.

Considering the fact that the risk among different segments of population may vary due to the behavioral, occupational, or susceptibility factors (Jia et al. 2011). We, therefore, also calculated the ILCR for adults (representing general population in residential areas), children (representing the most vulnerable population) and outdoor workers (representing occupationally exposed cohorts) exposed to air-born PAHs in the atmosphere of study areas. The occupational exposure is an important aspect for risk assessment in cancer studies, especially in the lung cancer studies (Boffeta et al. 1997). The U.S. EPA model used for the calculation of ILCR showed no serious risk; in general, the risk remained between 10^{-6} and 10^{-7} for all three exposure groups. However, risk was comparatively high for children, and for the population of Wazirabad and Lahore as compared to other study areas. Lowest risk was seen in Sohdra and Islamabad cities; in conclusion, the risk from inhalation exposure to airborne PAHs was lower (in these cities) than the limits specified by the U.S. EPA (Figure S5).

References:

Please find all the references in the main document

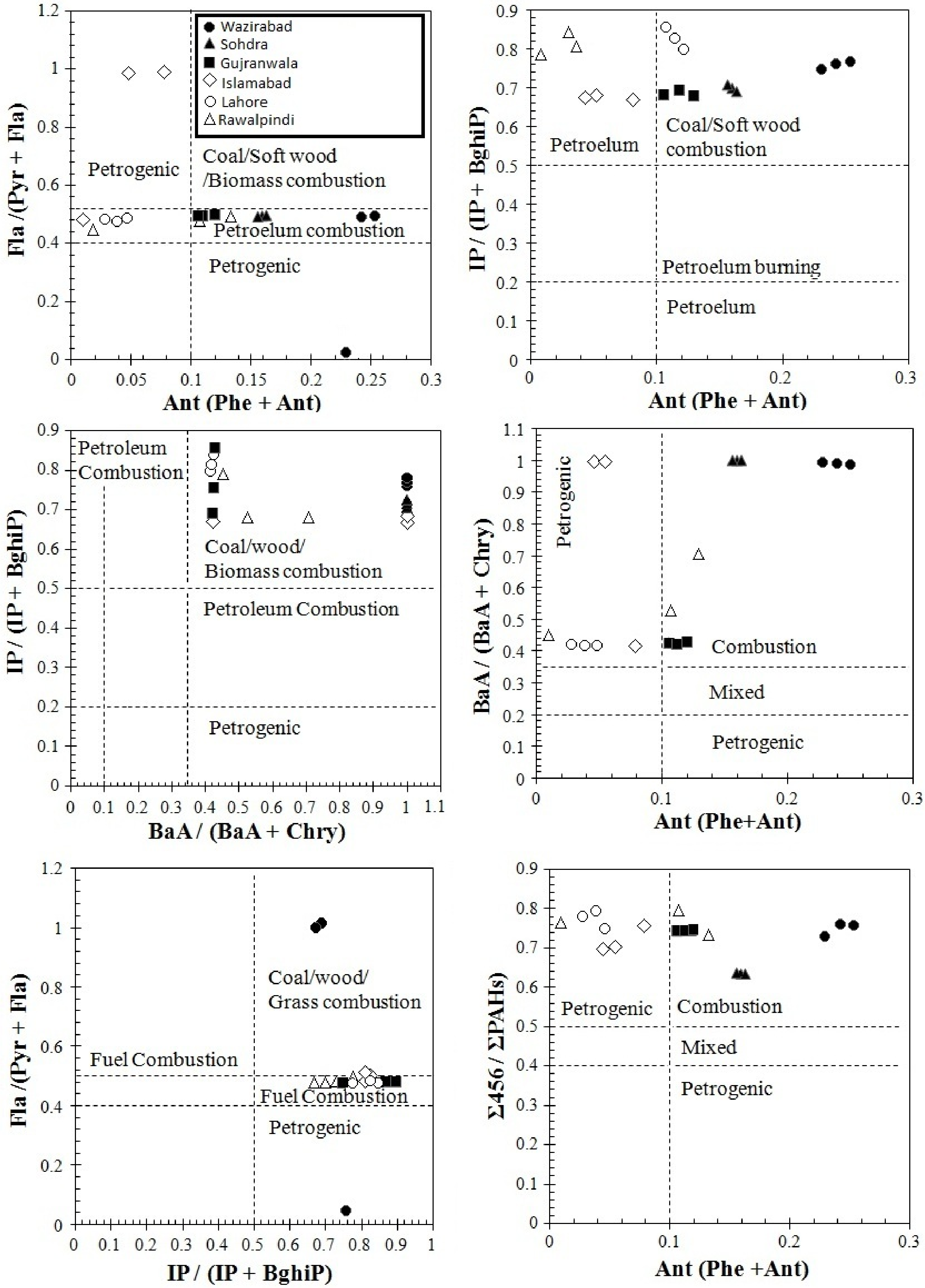

Figure S 1: Source apportionment using diagnostic ratios

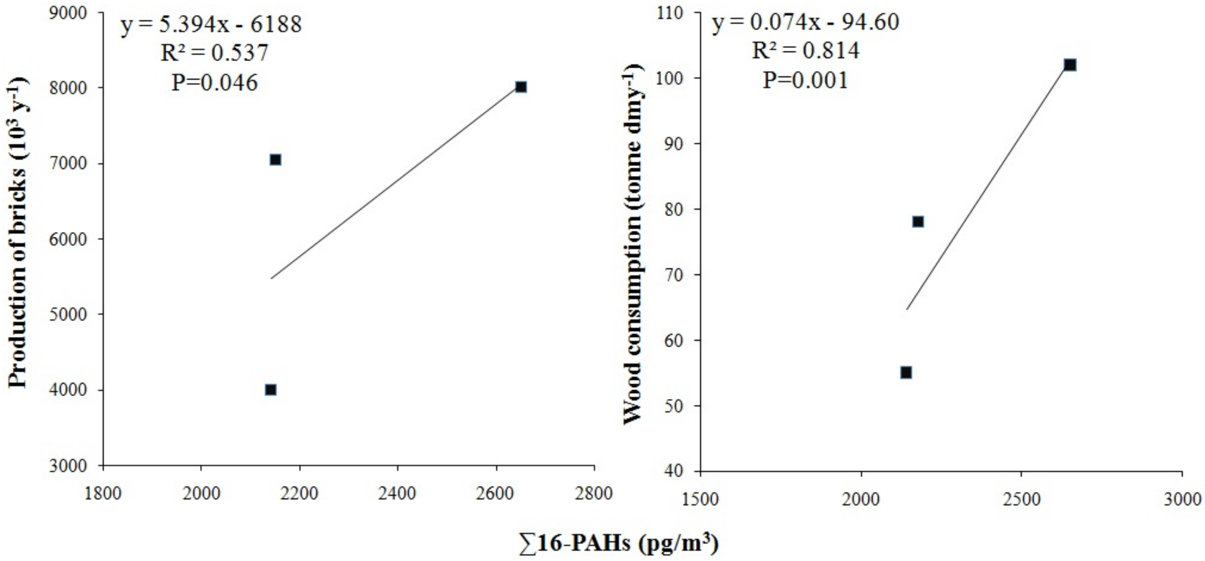

Figure S 2: Association between production of Bricks / wood consumption with measured (total PAHs) concentrations in three currently studied districts

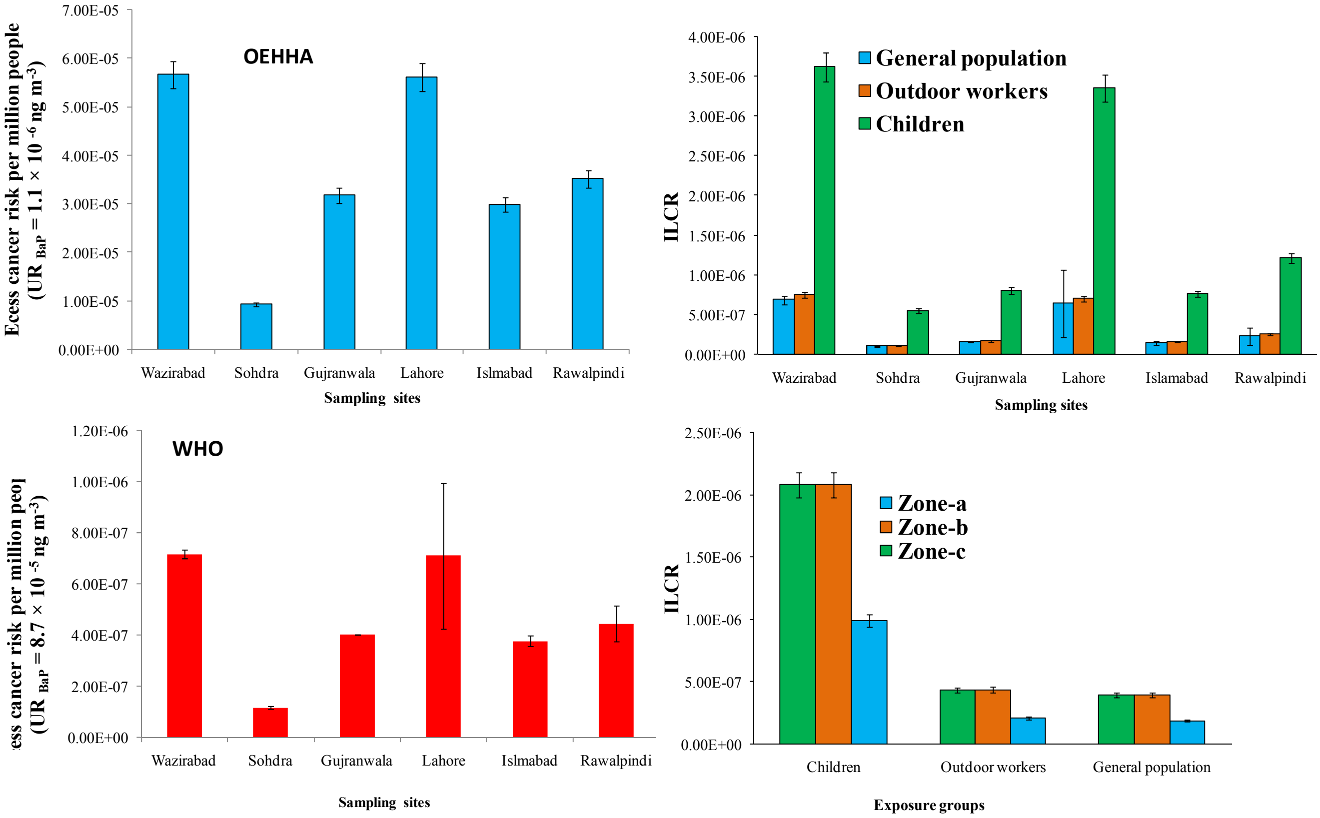

Figure S 3: Excessive carcinogenic risk per person due to inhalation exposure to PAHs in the sampling sites based on UR_{BaP} values specified by OEHHA, and WHO and ILCR value of different exposure groups, cities and zones

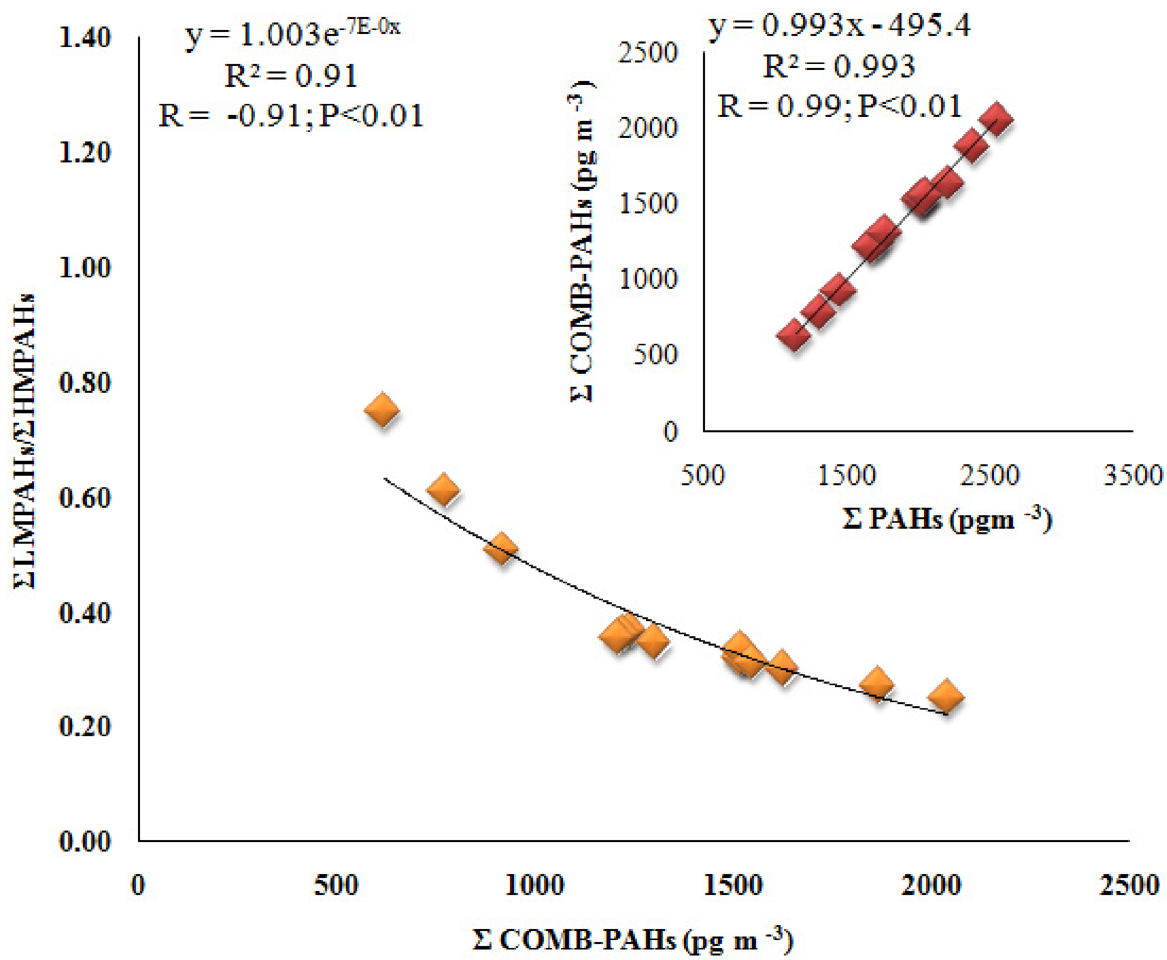

Figure S4: Association between low molecular weight and combustion origin PAHs