Doctoral Thesis

Monitoring and risk assessment of polycyclic aromatic hydrocarbons in the air

(Summary)

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September 2015

Polycyclic aromatic hydrocarbons (PAHs) are recognized as ubiquitously distributed class of potent toxic substances present in the human environment at low concentrations. PAHs are long-lived organic compounds and originate almost entirely from anthropogenic activities (90%) such as traffic and industrial fossil fuel burning, biomass combustion and agricultural activities. Owing to their ubiquitous distribution, potential carcinogenicity, mutagenicity, and teratogenicity, PAHs have been widely investigated since 1970s worldwide. Hazards associated with these compounds are owing to their hydrophobic character, persistence and bioaccumulation properties of several individual PAHs. Hence the primary goal of this thesis is to identify and determine particulate PAHs in the aerosol of Kamihaya, Hiki, and Higashi Hiroshima, Japan. This study was conducted between August 2012 and July 2014. Chapter 1 provides a general overview on PAHs pollution of atmospheric aerosols, the sources of PAHs in air and briefly describes the significance of this study. It also provides the insight of PAHs related human and vegetation health effects.

Chapter 2 deals with the survey conducted to analyze particle-associated atmospheric PAHs at a rural site (Kamihaya) in Tanabe, Wakayama Prefecture, central Japan from 2012–2013. Analysis of particulate matter revealed that total PAHs (Σ17PAH) ranged from 0.036 to 10.16 ng m^{-3} with an average concentration of 3.20 ng m^{-3}. The highest PAHs concentration of 10.16 ng m^{-3} was observed on September 19, 2012. Significant seasonal variations in PAHs concentrations were observed at the site, with higher values during winter/spring and lower values during summer/autumn. This investigation reported high concentration of heavy molecular PAHs compared with low molecular PAHs. 5–6-ring PAHs were dominant followed by 4-ring and least concentration was reported for 2–3-ring PAHs. Trajectories revealed that PAHs in Kamihaya are of domestic origin, mainly from nearby Gobo power generation plant.

Molecular diagnostic ratios and principal component analysis showed that atmospheric PAHs primarily arose from industrial and traffic emissions, while wood combustion appeared to be a minor source. Overall, the results of chapter 2 suggest that atmospheric PAHs in Kamihaya are a potential threat to the underlying vegetation and may cause damage to plants if combined with other air pollutants.

In 2013 – 2014 survey a total of 17 PAHs (16 US-EPA PAH + BeP) were analyzed in aerosol of Kamihaya, Hiki, and Higashi Hiroshima. The mean concentrations of Σ17PAH were in the order of 2.43 > 1.63 > 1.18 ng m^{-3} at Higashi Hiroshima, Kamihaya and Hiki (Chapter 3). The mean Σ17PAH at all three sites varied seasonally with the highest concentrations in winter, whereas summer was characterized with lowest PAHs. T-test found significant seasonal variations of mean Σ17PAH at Higashi Hiroshima (p<0.01) and Hiki (p<0.05), while at Kamihaya variations were non-significant. The concentrations of heavy molecular (5–6-ring) PAHs are dominant at Kamihaya and Higashi Hiroshima, while in Hiki 4-ring PAHs are the highest. The temporal PAHs have been almost constant over the last 15 years in the aerosols of Higashi Hiroshima. However, BaP concentration reported in this study is lower than the previous investigations.

Chapter 3 also discusses the influence of meteorological parameters on particulate PAHs in the aerosols of Kamihaya, Hiki, and Higashi Hiroshima. It was found that ambient air temperature is the most significant meteorological parameter controlling the atmospheric PAHs. Statistically significant high correlation was found between weekly Σ17PAH concentrations and ambient air temperature at Higashi Hiroshima. The correlation between mean Σ17PAH and air temperature was statistically significant and moderate in the aerosols of Kamihaya and Hiki. At Hiki southerly winds were found one of the important meteorological parameters acting as PAHs

regulating force. We could not find significant correlation of solar irradiance, rainfall and humidity with Σ17PAH at all three sampling sites. Trajectories analyses suggested that the PAHs from domestic sources (Gobo power generation) play an important role specifically in Kamihaya, and Hiki in general. Whereas, in Higashi Hiroshima PAHs from other East Asian countries play important role to increase the local pollution level. Based on PCA results it was concluded that PAHs emission is mainly from vehicular emissions and industry related activities. The other emission sources are domestic heating/cooking and biomass combustion. The lifetime lungs cancer risk (ILCR) was estimated at Hiki and Higashi Hiroshima, which was found with in the health-based guidelines of European Union by WHO (10^{-5}), however one order higher than US EPA guidelines (10^{-6}). The PAH at both residential sites pose a moderate lung cancer risk to the residents of these areas. BaP was found the largest contributor to the total risk followed by DiBahA at both sites. 5–6-ring PAHs contributed more than 95% of total risk at both sites and rest was contributed by 4-ring and 2–3-ring PAHs.

Chapter 4 deals with general discussion, concludes the whole study, and suggests the future work. This is the first approach of its kind in Kamihaya (a significant agricultural area) and Hiki to investigate the particulate PAHs. This study deduces that traffic and industry are the significant PAHs emission sources. In Kamihaya and Hiki PAHs are mainly from domestic sources, while in Higashi Hiroshima long-range transport of atmospheric PAHs is important in terms of pollution. This study investigated the importance of meteorological parameters in regulation of atmospheric PAHs in underlying study areas. Ambient air temperature and wind direction were the important meteorological factors in controlling the atmospheric PAHs in study areas. The influence of other meteorological factors such as solar irradiance, rain fall,

humidity was non-significant. Our study will provide the basic information on status of atmospheric PAHs for future detailed studies on PAHs in relation with vegetation and human health. However, it is recommended to develop the atmospheric PAHs inventories for Japan to fully understand the spatial their spatial distribution. This chapter also discusses the atmospheric PAH pollution preventions strategies. It also suggests legislations for PAH emissions from different sources and provision of guidelines of PAHs in different environmental matrices. The investigation of gaseous PAHs is needed to completely understand the status of atmospheric PAHs. A quest for the effects of PAHs on Japanese apricot plant is yet to be explored to understand the mode of PAHs action on tree.