The influence of aerosol physico-chemical characteristics on the fate of PAHs in the atmosphere

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Polycyclic Aromatic Hydrocarbon (PAH) compounds are composed of two or more fused benzene ring, and are ubiquitous in the environment. Anthropogenic sources of PAHs are related to the incomplete combustion of almost any fossil fuel which are emitted in both gaseous and aerosol phases. The fate of PAHs residing in the atmosphere has received enormous attention in recent years due to their mutagenic and carcinogenic risks on human health.

The physicochemical nature of aerosols differs relative to their physical size (i.e. grain size). Aerosols larger than 2um are mainly derived from geologic materials such as mineral grains. The smaller aerosols (less than 2um) are made up of materials that come from gas phase reaction (gas to particle conversion) and soot. Only a few studies have given attention to the compositional differences among aerosol particle sizes that influences the fate of PAHs in the atmosphere. This study has focused on such effects, and further stressed the importance of the aerosols surface characteristics in influencing the fate of PAHs in the atmosphere.

Aerosol samples were collected using a low-volume aerosol sampler (SHIBATA AN-200) for a one year period (4/21/2003 to 4/28/2004) in the suburban area of Kanazawa, Japan. The automated sampler can separate aerosol sizes using filters in 9 stages (i.e. 0.43um to 11um). Filters were collected and replaced once a week throughout the sampling period. The particle size of aerosols are referred here as coarse (2-11um) and fine (less than 1.1um) fraction.

PAHs concentration in the coarse and fine fractions of aerosols were measured based on Hayakawa (1996). The PAHs considered in this study include phenanthrene (Phe), Pyrene (Pyr), Benzo(a)anthracene (BaA), Chrysene (Chr), Benzo(b)fluoranthene (BbF), Benzo(k)fluoranthene (BkF) and Benzo(a)pyrene (BaP).

Based on Pankow (1987), the gas phase concentration of PAH is proportional to the sub-cooled vapor pressure and concentration of PAH in aerosol. Adsorption manner of gas phase PAH to the surface of aerosol has been modeled using a Langmuir-type adsorption isotherm (Pankow, 1987). In this model, there must be a linear relationship between the logarithm of PAH concentration per unit surface area (i.e. aerosol) and logarithm of gas phase PAH concentration. The linearity is evaluated in the coarse and fine fractions of the aerosols by linear coefficient values. It was indicated that the coarse fraction has higher values than the fine fraction throughout the sampling period. Adsorption is suggested as the dominant PAH uptake mechanism in the coarse fraction. The seasonal trend of the liner coefficient values were clearly observed especially in the fine fraction of aerosols which are in accord with the Asian monsoon rise and falls. The linear coefficient value for fine fraction of aerosol increased dramatically from 0.10 in autumn to 0.88 in mid-winter. Relatively high values were maintained until the middle of spring, and decreased from late spring to summer.

Thermodynamic analyses have shown that there must be the linearity between the logarithms of PAH concentrations per unit surface area, and the reciprocal of temperature, which can be used in describing the temperature dependence PAHs concentration in aerosols. The calculated linearity is always higher in the fine fraction than in the coarse fraction. This indicates that the cool temperature tends to partition more of the PAHs into the fine fraction of the aerosols. The high linearity related Lagmuir adsorption model in the fine fraction of aerosols for the Asian monsoon period agrees well with the described linearity. Since the inland areas of the Asian continent are generally cooler than Japan, PAHs tend to partition in the fine fraction of aerosols that could be transported by the Asian monsoon towards Japan.