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Spatial and temporal distribution of polycyclic aromatic hydrocarbons (PAHs) in the atmosphere of Xiamen, China

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ABSTRACT

An intensive sampling program was conducted from October 2008 to September 2009 at the five different environmental sites in Xiamen, Fujian Province, to study the spatial and temporal characteristics of Polycyclic Aromatic Hydrocarbons (PAHs) in the gaseous and particulate phase, respectively. The PAHs concentrations at different sites were quite distinct during four seasons. The average concentrations of PAHs in winter were about 8.4 times higher than those in spring, and the concentrations of background were 0.56 times lower than those of industrial area. In addition, the higher temperature in summer affected the particle/gas partitioning of PAHs and led to the higher concentrations of gaseous PAHs. Diagnostic ratios of PAHs, which were employed to indicate the primary sources of PAHs in Xiamen, showed that the traffic vehicle exhaust was the largest contributor and the primary source for PAHs in Xiamen, especially in urban area; while the stationary combustion processes, such as petrochemical factories and power plants, were mainly responsible for PAHs sources in the industrial areas. The health risk of PAHs in the particulate phase was higher than those of the gaseous phase at the five sampling sites. The average toxic equivalent (BaP_{eq}) of the benzo[a]pyrene values for PAHs were 0.14, 0.32, 1.38 and 3.59 ng m^{-3} in spring, summer, autumn and winter, respectively. Furthermore, the results of average BaP_{eq} in all four seasons indicated that the health risks of particulate PAHs were higher than those of the gaseous PAHs at different sampling sites.

1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), mainly originated from incomplete combustion of organic matter, are persistent organic pollutants existing in the environment extensively, and have a highlight of air pollution research due to their mutagenic and carcinogenic properties (Dong and Lee, 2009).

Sources of PAHs contaminations include domestic and industrial coal combustion, biomass combustions, and vehicle emissions (Wan et al., 2006; Heywood et al., 2006; Shen et al., 2010; Tian et al., 2009). Stationary combustion emission and automobile exhausts have been recognized as the major PAHs contributors (Li et al., 2003; Wan et al., 2006). For example, the previous studies demonstrated that PAHs originated from the coal-related sources made a 70–87% contribution to the total sources in Dalian (Wan et al., 2006). Furthermore, it was estimated that approximately 10.7% of the PAHs emissions in China in 2004 were from the residential coal combustions (Shen et al., 2010). For the vehicle related sources, it was reported that the motor vehicles accounted for as much as 90% of the particle-phase PAH masses in the air in downtown of Copenhagen and the PAHs levels varied both temporally and spatially, depending on the traffic patterns (Nielsen, 1996). The reports of Harrison et al. (1996) confirmed the similar conclusion that primary vehicular emissions accounted for 44–93% of the concentrations of individual PAHs in the air borne particles in Birmingham (UK). There are many researches regarding particle-phase PAHs. However, the concentrations of PAHs in the gaseous phase and distributions of PAHs between particulate and vapor phases were not considered.

Xiamen, a subtropical and coastal city in the southeastern China, has a warm weather without heating episode and high relative humidity throughout the year (air temperature ranged from 4 to 38 °C with 20.9 °C mean during 2008–2009). The prevailing wind directions in autumn and winter are North and Northeast in Xiamen. This could bring cold and dry air masses from northern China. On the other hand, the warm and wet air masses carried by Southeaster from sea dominate in summer. In 2008, Xiamen has a population of 2.5 million with 1565 km^2 area, being a main economic and culture center of Fujian Province. With the rapid development of economy, like many cities in China, Xiamen is increasingly facing some air pollution problems. Specifically, the rising number of motor vehicles has been regarded as one of main reasons for the deterioration of air quality in the urban area, number of which increases from 190,432 in

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2000 to 508,522 in 2008 as shown in Ding et al. (2001) and Ge et al. (2009). The concentrations of total suspended particles (TSP) in the atmosphere also increase from 2000 to 2009 (Ding et al., 2001; Ge et al., 2009). The high levels of TSP often contain carcinogenic and mutagenic organic matter, including PAHs (Ayrault et al., 2010; Bi et al., 2003). Therefore, the risks associated with human exposure to atmospheric PAHs become higher and higher in the urban areas.

Despite the fact that PAHs in Xiamen had been reported by some literatures (Ye et al., 2006), no intensive research had been conducted on spatial and temporal distributions for the PAHs in the atmosphere of Xiamen, especially, the distributions of PAHs in the gaseous phase. The objectives of this paper are: (1) to measure the concentrations of ambient PAHs in gaseous and particulate phases and study spatial and temporal characteristics of the PAHs in the atmosphere, (2) to identify possible sources of PAHs, and (3) to assess the health risk of PAHs in the atmosphere of this coastal city.

2. Experiment

2.1. Sites and sampling

Air samples were collected at five sites in Xiamen: Tingxi reservoir (TX), Institute of Urban Environment, CAS (IUE), Lulian hotel (LL), Xianyue residential area (XY), Xiamen University (XU) (Fig. 1).

The TX site, as a background site, is located in a forestry reserve with a medium-sized reservoir which is the source of drinking water for Tong'an district of Xiamen. The samplers were placed on the rooftop of the reservoir office, and approximately 6 m above the ground. This site is close to reservoir and far from traffic road. The IUE site, as a suburban sampling site, is located in Jimei District with rapid urbanization and is surrounded by highways, schools, residential buildings and Xinglin Bay. The samplers were installed on the rooftop of the laboratory building, and about 30 m above the ground. The LL site, as an industrial sampling site, is located in Haicang District, which is a main base of petrochemical and power sources in Xiamen. The samplers were mounted on the rooftop of a four-story hotel, and about 15 m above the ground.

The XY and XU sites, as the urban sampling sites, were selected in Siming District to represent commercial and educational area, respectively. The samplers at these two sites were placed on the rooftop of the residential building and classroom building, respectively, and were about 15 m and 18 m above the ground, respectively.

The 24-h air samples were obtained from October 2008 to September 2009, about 20 samples were collected at five sites for every season. The air samples were collected with the middle-volume air samplers (PS-1, Thermo Fisher Scientific, USA), at a flow rate of 160 L min^{-1}. Suspended particles were retained on a Whatman glass microfiber filter (GFF) (grade GF/A, diameter 10.16 cm), and vapor-phase species were absorbed on a polyurethane foam plug (PUF) (length 8.0 cm, diameter 6.25 cm). Considering the regular weather change of seasons in Xiamen and the shortage of samplers, these samples although not collected simultaneously at the five sites, the standard deviations of the three measurements for different samples of compounds are less than 10%, and therefore, we believe

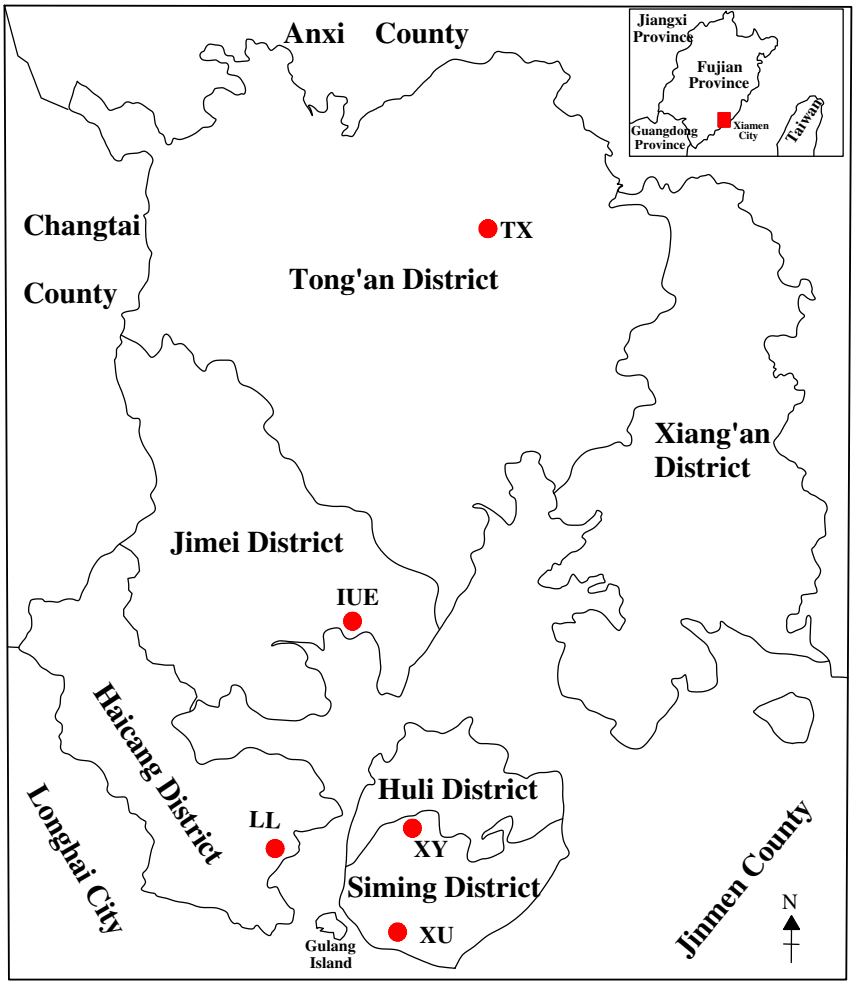

Fig. 1. Location of the five sampling sites in Xiamen shown as large red dots.