EMISSION, DISPERSION AND EXPOSURE OF POLYCYCLIC AROMATIC HYDROCARBONS IN TIANJIN, CHINA

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Abstract
Emission inventory of polycyclic aromatic hydrocarbons (PAHs) in Tianjin was developed. The annual emission of 16 PAHs was 192 tons in 2003 with an emission density of 16.1 kg/km^2. The dominant emission sources were domestic coal combustion, coking industry and biomass burning. The spatial distribution of PAHs in atmospheric in Tianjin was predicted using a dispersion model (ISCLT3). It was found that the urban districts with high population density were the most heavily contaminated. The relative contribution of various sources to the total emission, the total PAH concentration, and the total banzo(a)pyrene equivalent (BaPeq) concentration at the receptors were significantly different. For instance, domestic coal combustion contributed 43.3% to the total emission, 56.0% to the total PAH concentration at receptors, and 80.8% to the total BaPeq at the receptors. Along with information on PAH concentrations of other environmental media and food items, population exposure to PAHs was assessed using a multi-pathway model with spatial resolution. The calculated chronic daily intake for children, youth and adult in Tianjin were 2.92, 2.33 and 1.90 µg/(kg·d), respectively. Based on these intakes, a lifetime daily exposure in Tianjin was estimated to be 2.06 µg/(kg·d), a level significantly higher than those reported in countries.

Introduction

Tianjin is one of the fastest growing areas in China. Like many other places in the country, this Tianjin is severely contaminated by polycyclic aromatic hydrocarbons (PAHs). Exposure to PAHs of its near-10 million population is a major concern in terms of health impact of environmental pollution. Similar to other northern cities in China, emissions of PAH in Tianjin are mainly from domestic coal combustion, biofuel burning and coke production. In fact, annual consumption of coal in Tianjin is about 23 million tons[1]. In addition, straw burning as energy is common in rural Tianjin[2]. It was estimated that the total emission of 16 USEPA priority PAHs was approximately 175 tons in 2003 with an emission density of 15.4 kg/km^2·y[3].

To estimate spatial distribution pattern of pollutants, dispersion models are often used. One of the frequently used dispersion models is the Industrial Source Complex Long-term (ISCLT3) developed by USEPA[4,5]. The spatial distribution pattern of atmospheric pollutants predicted by this model can provide detailed information necessary for exposure modeling and risk assessment. Based on the provided information, population exposure to certain contaminants can be assessed using a multi-pathway model that takes into account various exposure pathways[6]. For instance, a survey was conducted to assess population exposure to 46 pollutants in the United States[7].

The aims of this study were to develop a PAH emission inventory, to evaluate the spatial distribution pattern of the pollutants in the atmosphere using ISCLT3, and to assess the population level exposure with spatial resolution based on both the results of the dispersion modeling (using ISCLT3) and measured concentrations of PAHs in various food items from the area.

Methodology

Emissions of PAHs from various sources were derived by multiplying the consumed quantities and emission factors. Major PAH emission sources taken into consideration for development of emission inventory were coal used for power generation, gasification, central heating, coke production and other purposes, petroleum consumed in industry and transportation, natural gas combustion, and biomass burning. The locations, emission heights and fuel consumptions of all point sources were tallied individually for each use. Emissions from motor vehicles on highways and major roads were considered as linear sources described by road network and traffic flow patterns. Transportation emission from local roads in urban area was treated as an area source. Other area sources included emissions from biomass burning and domestic coal combustion. Coal used for residential heating was allocated based on the population density and the commercially used coal was allocated to urban districts and various towns based on tertiary productions[2].

The population exposure to PAHs in Tianjin was assessed using a total exposure model taking into consideration of multiple exposure pathways including diet, respiration, and dermal contact. In addition to an average exposure dose for entire population of Tianjin, spatial resolved exposure was also modeled based on the same grid system as the dispersion modeling. The calculated PAH concentrations in air were directly used in inhalation and dermal contact calculation for each grid, while the PAH concentrations in major food items were either directly measured in our laboratory or collected from the literature. Concentrations without spatial variation were used for all food items except vegetable. For vegetable, it was assumed that rural population consumes locally produced vegetables while urban population eats “the average” vegetables from the entire area. The concentrations of PAHs in vegetables were predicted based on the modeled concentrations in the air[8]. Monte Carlo simulation was conducted for a probabilistic exposure assessment.

Results and Discussion

The total emission of PAHs in Tianjin was 192 tons in 2003 with an emission density of 16.1 kg/km^2 which was around 6 times of the national average. The emission densities of domestic coal combustion and biomass burning sources are presented in Figure 1 (left and middle). The relative contributions of various sources were also shown in the pie chart (Figure 1, right). The dominant sources were domestic coal combustion (43.3%), coking industry (22.6%) and biomass burning (21.3%).

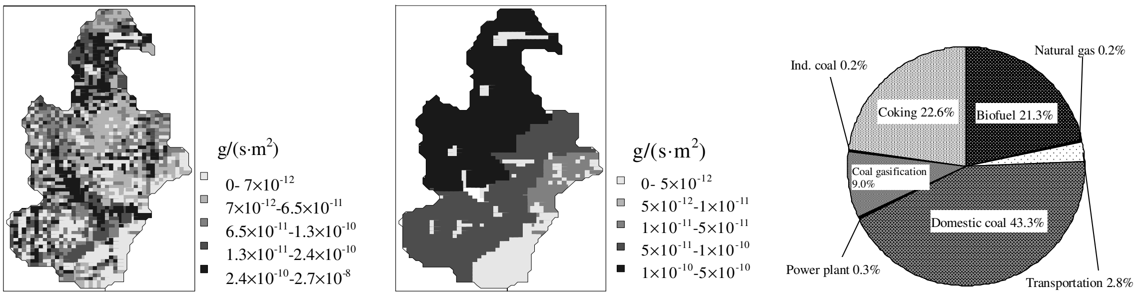

Figure 1. PAH emission densities from sources domestic coal combustion (left) and biomass burrning (middle) in Tianjin and relative contributions of various sources (right).

Concentrations of the 16 individual PAH compounds at low elevation (2 m above ground) were modeled using ISCLT3 at steady-state using a grid of 3113 receptor sectors of 2x2 km^2 covering the entire area of Tianjin. The modeling was performed separately for heating and non-heating season of Tianjin due to extraordinarily different emission rates in the two seasons. The annual weighted average concentrations were then derived. Several examples are illustrated in Figure 2. The predicted results were validated for gaseous and particulate phases separately using the observed data at two monitoring sites during both heating and non-heating seasons. Since the dispersion was modeled for each category of emission sources and the total air concentrations were derived by summing up those from all sources, the relative contributions of individual sources to the PAH levels at a given locations in the area were able to be calculated. It was found that the emission source pattern was remarkably different from the receiving pattern. For example, for domestic coal combustion, the contribution increased from 43.3% at the sources to 56.0% at the receptors. The second important PAH emission source in Tianjin was coke production and its share in total emission was 22.6% but the contribution to the receptor decreased to 6.6%, simply because the emission heights of the sources were much higher than the other sources.

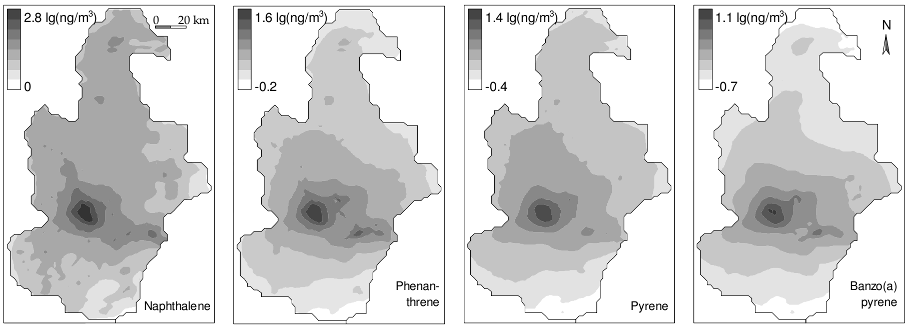

Figure 2. The model predicted spatial variations of annual mean concentrations of naphthalend, phenanthrene, pyrene and banzo(a)pyrene. The weighted averages are mapped in log-scale.

The above mentioned relative contributions of various sources were calculated with respect to the total atmospheric concentrations of 16 PAHs. If, however, banzo(a)pyrene equivalent concentrations, B(a)Peq, are calculated for exposure risk assessment, it was found that the contributions of various sources are totally different from that total concentration. As shown in Figure 3, the domestic coal combustion now contributed to over 80% of the total B(a)Peq in the area while all other sources shared less than 20% of the total.

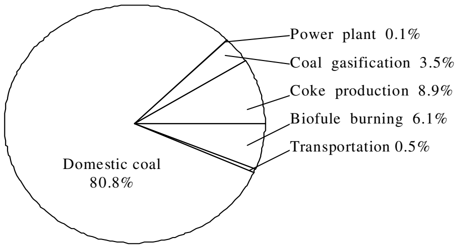

Figure 3 Contributions of various sources to BaPeq in Tianjin

Because of the high emission density of PAHs, various environmental media in the area are expected to be heavily contaminated. An extensive survey conducted several years ago collected 188 surface soil samples and indeed revealed relatively high levels of PAHs contamination. Contaminations were reported for air, water, dustfall, vegetables, grain and various food items. Although relatively high levels of PAHs were found in general, the concentrations vary spatially. For instance, a visual comparison of the PAH concentrations between air and surface soil revealed similarity in spatial distribution pattern with high concentrations in urban districts.

Based on the information on PAH concentrations in various environmental media and food items, the chronic daily intake (CDI) of PAHs were calculated for children, youth and adults in Tianjin. It was reveled that the CDI were 2.92、2.33 and 1.90 μg/(kg·d) for children, youth and adults with a lifetime daily exposure of 2.06 μg/(kg·d) in Tianjin in 2003. As shown in Figure 4, the major exposure pathways in terms of lifetime exposure in Tianjin are grain, meat, vegetable, oil, inhalation and fish. These pathways contribute 93.1% of the total exposure.

As the dominant pathway, the diet exposure of PAHs in Tianjin was much higher than those reported in the other countries in the literature. For instance, the total exposure of 16 PAHs in Tianjin (94 μg/d) was more than one order of magnitude higher than that in Spain in 2003 (7.4 μg/d); the exposure of 9 higher molecular weight compounds in Tianjin (17.9 μg/d) was more than 5 times higher than that in UK in 1983 (3.5 μg/d)[9]. Based on the spatially resolved Monte Carlo simulation, probabilistic distribution of CDI in Tianjian area was derived and presented in Figure 5 (left). It was shown again that the geographical distribution was similar to the emission and the population in the urbanized districts exposed to much higher PAHs than the rural population (Unfortunately, more than 50% of the local population lives in the urban area). Based on the Monte Carlo simulation on the grid system, the exposure probability is presented in Figure 5 as the CDI of BaPeq in Tianjin. It appears that rather high risk existed in the area.

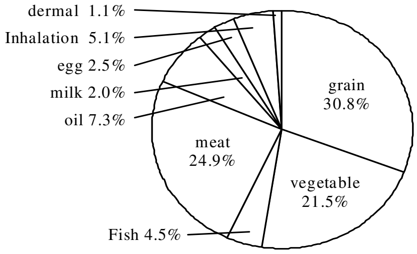

Figure 4 Contributions of various pathways to the lifetime PAH exposure in Tianjin

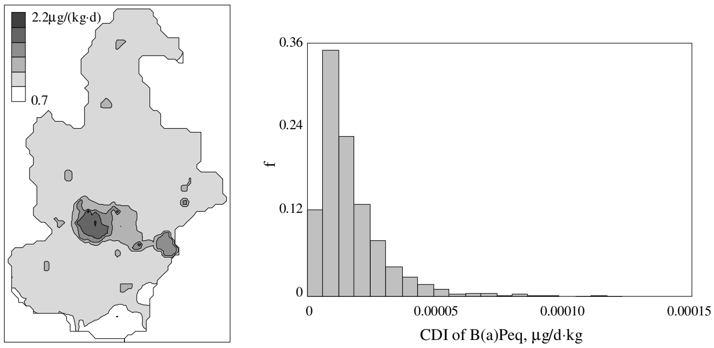

Figure 5 Geographical and probabilistic distributions of Chronic Daily Exposure to PAHs in Tianjin (left) and probabilistic distribution of CDI of BaPeq in Tianjin (right)

Acknowledgements

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