



Human health risks of polycyclic aromatic hydrocarbons in the urban soils of Nanjing, China



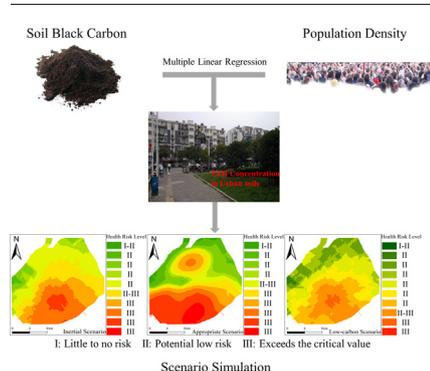
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HIGHLIGHTS

- The risk of PAHs in urban soils was divided into four levels based on human health risk assessment.
- Population density and black carbon content were determined to be the key factors of PAH accumulation in urban soils.
- Multiple linear regression models and the scenarios simulation method were used to predict PAH levels in urban soils.

GRAPHICAL ABSTRACT



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ABSTRACT

Polycyclic aromatic hydrocarbons (PAHs) are a major group of toxic pollutants in urban areas. We calculated the critical concentrations of PAHs in the urban soils of Nanjing, China based on a human health risk assessment. In the study area, the risk was divided into four levels and toxic equivalent values of benzo[a]pyrene (BaP_{eq}) corresponded to $<70 \text{ ng g}^{-1}$, $70\text{--}700 \text{ ng g}^{-1}$, $700\text{--}7000 \text{ ng g}^{-1}$, and $>7000 \text{ ng g}^{-1}$. By this standard, most urban areas in Nanjing fall under level II (potentially low risk), while older urban districts, commercial centers, and transportation centers exceed the critical concentration (level III) at present. Additionally, the correlations between PAH concentrations, factors associated with urbanization, and soil properties were analyzed. Population density and black carbon content were determined to be the key factors involved. Multiple linear regression models and the scenario simulation method were used to predict PAH levels in urban soils through 2030. The results indicated that the future distribution characteristics of soil BaP_{eq} under various scenarios were different than at present, but PAH concentrations remained stable only under the low carbon scenario. Therefore, the consumption of traditional fossil fuels should be controlled and replaced with alternative energy sources. In addition, the growth of traffic land use should be controlled in the southern and southwestern parts of the urban area.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are ubiquitous environmental pollutants that are carcinogenic, mutagenic, and toxic to all living organisms (Tobiszewski and Namieśnik, 2012; Wang et al., 2017). PAHs have been linked to skin, lung, and bladder cancer in humans (Boffetta et al., 1997). The United States Environmental Protection

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Agency (USEPA) has identified 16 PAHs as priority pollutants (USEPA, 2013). PAHs are not readily soluble, readily adsorb to soil particles, and are difficult to degrade. Thus, they tend to accumulate in soils, which are the most important environmental sinks for these contaminants (Wang et al., 2017). In recent years, the problem of PAH soil pollution has gained attention, especially in China, where rapid development over the past several decades has led to massive industrialization and urbanization (Peng et al., 2016; Wang et al., 2017; Wang et al., 2015; Yang et al., 2015).

PAHs primarily originate from anthropogenic sources related to urban development, such as vehicular emissions, fossil fuel combustion, chemical manufacturing, and oil spills (Callén et al., 2013; Orecchio, 2010; Peng et al., 2013; Wang et al., 2015). Liu et al. (2010) found a correlation between PAHs in soil and the development history of Beijing's urban district, inferring an increasing concentration of soil PAHs over time. Residential building age, population density, road density, and distance from the city center can be used as urbanization-level indicators, and significant correlations have been found between these indicators and the concentrations of PAHs in soil (Peng et al., 2013). Jensen et al. (2007) found that PAH concentrations were substantially lower in the less-populated areas of northern Norway than in the more-populated, urbanized areas near Oslo in southern Norway. In general, human activities increase concurrently with urban population, which leads to higher concentrations of PAHs produced in the area (Saltiene et al., 2002).

Moreover, the properties of soil also influence PAH accumulation (Brändli et al., 2008; Bucheli et al., 2004; Zhang et al., 2006). Heywood et al. (2006) indicated that regional and local scale processes including temperature, rainfall, and soil physicochemical properties can affect PAH distribution and fate. Ni et al. (2008) found that the degree of PAH enrichment in different organic matter fractions decreased in order of the light fraction, tightly combined humus, stably combined humus, and loosely combined humus. Li et al. (2010) studied the distribution of PAHs in different size fractions of coke oven plant soil and found that partitioning in organic matter, especially in black carbon (BC), dominates the behavior of PAHs in soil.

Prediction of PAH levels is an important step in preventing the harmful health effects of pollution. Multiple methods have been developed to predict contaminant levels in soils, but most are complex and difficult to apply. Multiple linear regression (MLR) is one modeling technique used to calculate a dependent variable with two or more independent variables. Some models widely used in environmental science have been developed with MLR, including land-use regression and geographical weighted regression (Chen et al., 2016; Song et al., 2014; Whitlock and Montgomery, 2008). Most modeled pollutants are particulate matter with diameters between 2.5 and 10 μm ($\text{PM}_{2.5}$, PM_{10}) and either nitrogen oxides (NO_x) in the air or heavy metals (HMs) in soil (Jedynska et al., 2014; Liu et al., 2013; Zhang et al., 2009). However, few studies have incorporated anthropogenic factors and soil properties in a MLR model to predict the accumulation of organic pollutants, such as PAHs, in soil.

The critical concentration is the acceptable total load of PAHs in soils. Multiple studies on the critical concentration of soil pollutants and their exceedance values exist (Vries et al., 1998; Hettelingh et al., 2007; Wu et al., 2016). However, few focus on the critical concentration of PAHs with respect to human health risk. Health risk assessment is defined as the characterization of potentially adverse health effects from human exposures to environmental hazards (Adgate et al., 2014). At present, the four-step system developed by the USEPA, which incorporates hazard identification, dose-response assessment, exposure assessment, and risk characterization, is the most widely recognized methodology for assessing the risk of environmental contaminants. Exposure assessment is a critical component of estimating the risk of pollutants to human health (Graham et al., 1992). Ingestion, dermal contact, and inhalation are the conventional exposure scenarios in urban soils, and the incremental lifetime cancer risk (ILCR) model is usually adopted to characterize the health risks (Cao et al., 2016; Chen

et al., 2013; Khillare et al., 2014; Peng et al., 2011; Peng et al., 2016). Under most regulatory programs, an ILCR value between 10^{-6} and 10^{-4} indicates potential risk, whereas ILCR values $>10^{-4}$ indicate potential high health risks (Liao and Chiang, 2006). The ILCR value is a probability coefficient and has a large elasticity range, which can impact the management of the soil environment. Therefore, determining the critical concentration of contaminants, based on the human health risk, is an important avenue of investigation.

Accordingly, the aim of this paper was to calculate the critical concentrations for current and future levels of PAHs in urban soil. First, using the ILCR exposure model, we established risk levels based on the concentration of PAHs in soil. Next, we incorporated anthropogenic factors and soil properties (of various PAHs) in a MLR model to predict the future accumulation of PAHs, using the scenario situation method. Finally, we identified high health risk areas within the sampling region.

2. Materials and methods

2.1. Soil sampling

The urban region of Nanjing, China was selected as the research area (Fig. 1). Nanjing has a subtropical monsoon climate, with an average annual temperature of 15.4 °C and an annual rainfall of 1200 mm. As a main port along the Yangtze River, Nanjing is a complex industrial base dominated by electronics, automobiles, and chemicals. The total population of Nanjing exceeds 8 million and >6 million occupy the urban area. At each sampling site, five subsamples (four corner samples and one center sample) were combined to form one composite sample. The sampling campaign was conducted in June of 2014, and common standardized procedures were performed across the study area. A total of 69 topsoil samples (5 cm in depth) were collected. The coordinates of the sampling site locations were recorded using GPS. The study region and sampling sites are shown in Fig. 1.

2.2. Soil analysis

We analyzed levels of Naphthalene, Acenaphthylene, Acenaphthene, Fluorene, Phenanthrene, Anthracene, Fluoranthene, Pyrene, Benz[a]anthracene, Chrysene, Benz[b]fluoranthene, Benz[k]fluoranthene, Benz[a]pyrene, Indeno[123-cd]perylene, Benz[ah]anthracene, and Benz[ghi]perylene. Soil was extracted with hexane/dichloromethane ($v/v = 1:1$) using a Soxhlet extractor, and the extracts were concentrated by rotary vacuum evaporation. Next, the solvent was changed to *n*-hexane and the concentrated extracts were cleaned using silica gel column chromatography. Following this step, the column was eluted with *n*-hexane/dichloromethane ($v/v = 3:2$). The collected PAH fraction was vacuum-evaporated, solvent-exchanged to *n*-hexane, and run on a gas chromatograph fitted with a mass selective detector. The detector was equipped with a fused silica capillary Rtx-5MS column (30 m \times 0.25 mm internal diameter, 0.25 μm film thickness). Ionization was carried out using the electron impact (EI) mode, and data were acquired using the selective ion monitoring (SIM) mode. The external standard method was used to measure the 16 PAHs. The average recoveries, based on matrix-spiked samples (pre-extracted soil was spiked with PAH standards), were 73.8% for Nap and 84.5–104.2% for the remaining 15 PAHs. Each batch of 6 samples included a blank and a matrix blank.

We determined BC content using the method of Lim and Cachier (1996), with slight modification. The procedure was as follows: (1) Acid treatment. Three grams of dried soil was weighed into pre-cleaned polyethylene test tubes and carefully treated with 15 mL of 3 M hydrochloric acid to remove carbonate from the soil matrix. The reaction was left to complete over 24 h. Following repeated centrifugation and rinsing with ultrapure water, 15 mL of 10 M hydrogen fluoride/1 M hydrochloric acid was added to the residue, and the reaction was allowed to continue at room temperature for an additional 24 h. The

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