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Polycyclic aromatic hydrocarbons in ambient air, surface soil and wheat grain near a large steel-smelting manufacturer in northern China

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ABSTRACT

The total concentrations and component profiles of polycyclic aromatic hydrocarbons (PAHs) in ambient air, surface soil and wheat grain collected from wheat fields near a large steel-smelting manufacturer in Northern China were determined. Based on the specific isomeric ratios of paired species in ambient air, principle component analysis and multivariate linear regression, the main emission source of local PAHs was identified as a mixture of industrial and domestic coal combustion, biomass burning and traffic exhaust. The total organic carbon (TOC) fraction was considerably correlated with the total and individual PAH concentrations in surface soil. The total concentrations of PAHs in wheat grain were relatively low, with dominant low molecular weight constituents, and the compositional profile was more similar to that in ambient air than in topsoil. Combined with more significant results from partial correlation and linear regression models, the contribution from air PAHs to grain PAHs may be greater than that from soil PAHs.

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Introduction

Polycyclic aromatic hydrocarbons (PAHs) are worldwide persistent organic pollutants (POPs) that usually enter plants through: (1) root adsorption, absorption and upward transport and (2) foliage stomata and waxy cuticle after dry and wet deposition of fine particle or aerosol adhered PAHs (CCME, 2008). To date, limited absorption and transport of PAHs via plant roots has been confirmed in the literature, especially for high molecular weight (HMW) species, although a few papers reported a different phenomenon (Fismes et al., 2002). For example, some studies observed that PAHs in different parts of crops were considerably correlated with gaseous and particulate PAHs in air, and thus, ambient air was likely an

important source of the PAHs accumulated by crops (Lin et al., 2007; Tao et al., 2006). Many factors may affect the partitioning processes of PAHs between air and plants, such as the octanol-air partitioning coefficient (K_{OA}) and physiological properties of foliage (Kipopoulou et al., 1999; Lin et al., 2007).

In China, pollution characteristics and crop types are quite variable, and hazardous accidents of agricultural products (e.g., cereals and vegetables) frequently occur and have grown worse in recent years (Si et al., 2015; Xing et al., 2016). In China, systematic monitoring data in representative areas accounting for relationships between environmental pollution and crop safety are insufficient and mainly focused on heavy metals, but rarely on POPs (L. Shen et al., 2013). Handan in Hebei Province, located in the western watershed of the Bohai

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Sea, is one of the main producing areas of spring wheat (*Triticum aestivum*) in North China (approximately 2 million tons/year). Meanwhile, Handan is an important base of raw coal (over 20 million tons/year) in the coking industry (approximately 5 million tons/year) and steel products (over 13 million tons/year); as a result, Handan has suffered severe air and soil pollution. For instance, a published report estimated that the local annual amount of emitted PAHs was up to 878.48 tons, with an emission density at 72.68 kg/km², 27 times higher than the national average (Zhang et al., 2007). The severity of the problem has generated a great deal attention from the local government and the public, particularly for resident morbidity from lung cancer (Zhang et al., 2009) and crop safety. However, corresponding studies on the spatial distribution, emission sources, and potential dietary exposure to PAHs via the local staple crops are fairly inadequate.

The objectives of the current study are to ascertain the following aspects: (1) the total concentration and component profile of PAHs in ambient air, surface soil and wheat grain in the vicinity of the local largest steelwork (Handan Steel-Smelting Manufacturer); and (2) the source apportionment of PAHs emitted and a comparison of the contribution to PAHs in wheat grain between ambient air and surface soil. Briefly, based on actual measurements from multimedia samples (ambient air, surface soil and wheat grain), the main emission sources of PAHs were identified by the specific isomeric ratios of paired components in ambient air as well as by principal component analysis (PCA) and multivariate linear regression (MLR). Further, the contribution to individual PAH species in wheat grain was compared between PAHs in ambient air and in surface soil using partial correlation analyses and multivariate linear regression models.

1. Materials and methods

1.1. Multimedia sampling

Based on a preliminary survey of surface soils and the local dominant wind direction, we selected 8 villages (assigned as S1–S8, respectively) with wide wheat fields in the vicinity of the large scale Handan Steel-Smelting Manufacturer as sampling areas near the local harvest time of spring wheat. The locations of sampling sites in 8 villages can be found in Fig. S1 of the Supplementary materials. Two neighboring wheat fields (S8-1 and S8-2, which are separated by a distance of 1 km) located the farthest distance from Handan Steelworks were considered to represent the local background field. All of the sampling fields chosen were far from the main road to avoid the possible influence of local traffic.

A total of 239 surface (0–5 cm in depth) soil samples were collected from wheat fields in 8 villages. The same number of wheat grain samples were simultaneously gathered, sealed, and quickly stored in a freezer at –15°C. Two improved passive air samplers (Tao et al., 2009) equipped with polyurethane foam (PUF) plugs and glass fiber filter (GFF) were deployed in each sampling village (on a roof or in open space near the wheat fields) to sample gas and particulate phase

PAHs for approximately 33 to 34 days. Before setup, PUF disks were extracted by a Soxhlet device containing 100 mL of acetone, 100 mL of dichloromethane and 100 mL of *n*-hexane over 8 hr, respectively. GFFs were preconditioned by roasting in a muffle furnace at 450°C over 6 hr. After air sampling, all of the sampling media (PUF and GFF) were removed, sealed and stored at –18°C. GFFs were equilibrated in desiccators (25°C) for 24 hr and weighed before and after sampling.

Meanwhile, active air samplers with a small flow volume (XQC-15E, Tianyue Instrument Ltd. Company, Yancheng, China) were installed in the selected 4 villages to calibrate the aforementioned passive sampling. In each sampler, the pretreated PUF tube (22-mm diameter × 76 mm length, Supelco) and GFF mounted inside the tube were employed to collect both gas and particulate PAHs. The sampling rate was set at 1.5 L/min, and the process continued for 24 hr. Due to local limitations and unexpected reasons, a total of 15 passive air samples and 4 active air samples were finally obtained.

1.2. Sample pretreatment and determination

1.2.1. Ambient air samples

Using the Soxhlet device, PUF disks were extracted with a 100-mL mixture of *n*-hexane and acetone (V:V = 1:1) at 70°C for 8 hr. The extract was then concentrated to a final volume of 1 mL using a vacuum rotary evaporator (R-201, Shensheng Sci. & Technol. Ltd., Shanghai, China) in a water bath at 37°C. GFFs were subjected to microwave extraction (MARS2Xpress, CEM, USA) with a 20-mL mixture of *n*-hexane and acetone (V:V = 1:1). The microwave tubes were heated to 100°C at a rate of 10°C/min, held for 10 min, and finally cooled for 30 min. After filter pressing, the extract was concentrated using a procedure similar to that of the PUF extraction and were then transferred into an alumina–silica gel column for purification. The eluent was first concentrated in a water bath at 37°C and then diluted with *n*-hexane up to 1.0 mL. The samples were then sealed in vials and stored at –4°C.

1.2.2. Surface soil samples

After the removal of detritus and plant residues, each soil sample was air-dried at room temperature and ground to pass through a 2-mm sieve. A subsample of 2.0 g of soil was further ground to pass a 70-mesh sieve and then transferred into a tube with a 20-mL mixture of *n*-hexane and acetone (V:V = 1:1) for microwave extraction like that for GFFs. Based on filter pressing, the extract was concentrated using the vacuum rotary evaporator to a volume of approximately 1.0 mL for clean-up in the aforementioned alumina–silica gel column.

1.2.3. Wheat grain samples

The wheat grains were air-dried at room temperature for over 7 days, ground to remove husks, and then stored at –18°C. A 10-g portion of the ground sample was transferred to a tube for microwave extraction using 20 mL of acetonitrile. After extraction and filter pressing, the extract was transferred into a 250-mL separating funnel, combined with 100 mL of deionized water containing 4% sodium sulfate, and then extracted twice using 30 mL of *n*-hexane. The extracted organic phase was shifted to a flask and then concentrated by rotary evaporation to a volume of 1.0 mL for the subsequent column purification step.

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