



Polycyclic aromatic hydrocarbons in soils of the central Tibetan Plateau, China: Distribution, sources, transport and contribution in global cycling



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ABSTRACT

Forty-four soil samples were collected across the central Tibetan Plateau (CTP) at altitudes between 3711 m and 5352 m, and their polycyclic aromatic hydrocarbons (PAHs) contents were measured to be from 0.43 to 26.66 ng/g. The main sources of PAHs were identified for each of four sub-areas, and their concentrations in soils were determined to be mainly influenced by local sources. Along a 600 km sampling trajectory from Lhasa, which served as the biggest local source, the concentrations of PAHs decreased logarithmically with increasing distances from the source. Meanwhile, the fractional proportions of PAHs were observed to change logarithmically according to the transport distances. Conclusively, PAHs from local sources were transported within the CTP and dominated PAHs concentrations in the soils, but few of them were transported outside the CTP. In global cycling, the soils in the CTP mainly serve as background and a “sink” for PAHs.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs), which are toxic compounds produced by the incomplete combustion of organic materials, can travel long distances in the atmosphere. Due to their long-range atmospheric transport (LRAT), they can reach and affect remote ecosystems at high latitudes (Friedman et al., 2014) and altitudes (Chen et al., 2008; Wania and Westgate, 2008, 2013).

Soils are the major reservoir of persistent organic pollutants (POPs) in the terrestrial environment, including PAHs (Kurt-Karakus et al., 2005; Wilcke, 2007). Furthermore, background soils are key environmental components for considering the relevance of secondary sources in global cycling (Cabrerizo et al., 2011a, 2011b; Komprda et al., 2013). Therefore, it is significant to understand the soil concentration and distribution of PAHs in a background region, such as the Tibetan Plateau at high altitudes (Tao et al., 2011; Wang et al., 2013).

The Tibetan Plateau (TP) has an area of over 2.5 million km², an average elevation of higher than 4000 m and is also called “the

third pole” of the earth. Even though the environment in the TP is less contaminated, PAHs in the air and soil have been detected in some locations (Qi et al., 2003; Wang et al., 2007; Tao et al., 2011; Yuan et al., 2014a). Most recently, Wang et al. (2014) reported the results of air-monitoring and the soil levels of PAHs in the TP. As introduced by the authors (Wang et al., 2014), the soils were almost all sampled from the populated south TP, a sub-alpine region dominated by the Indian monsoon. The central Tibetan Plateau (CTP), a typical alpine region, covers 60–70% of the area of Tibet (Wang et al., 2014). The soil samples from the CTP were few because the environment was too harsh to collect samples (Tao et al., 2011; Yuan et al., 2014a). Therefore, it is important to investigate the soil concentration, distribution, and even possible sources of PAHs in the CTP.

If PAHs were primarily inputted by LRAT from outside of CTP instead of local sources, this area could serve as another sink area, in addition to the Arctic, Greenland and other remote areas, for studying the global cycle of PAHs. Although PAHs levels were very low in CTP, the reported concentrations in various media varied greatly, implying the possibility of contributions from different sources (Qi et al., 2003; Tao et al., 2011; Yuan et al., 2014a). Tao et al. (2011) found that the elevated soil concentrations of PAHs in Lhasa, the largest town in Tibet, mainly originated from local sources. If so,

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it is important to reveal how the local PAHs would influence the concentration of PAHs in soils. On the other hand, it was reported that PAHs detected in the high Arctic atmosphere at Alert were originally from Eurasia (Halsall et al., 1997). If local sources do exist, it is also significant to investigate whether they would be transported outside the CTP and contribute to global cycling.

To elucidate these queries, soil samples were collected from CTP, including from two largest town of Lhasa and Shigatse, a popular travel route (Lhasa-Shigatse-Mount Everest) and the area north of this route. In this case, our goals were as follows: (1) to investigate the distribution and composition of PAHs in the CTP; (2) to determine the possible local sources of PAHs in the CTP; and (3) to reveal the transporting mechanism of local sources in the CTP and their potential contributions to global cycling.

2. Materials and methods

2.1. Sampling

All of the samples were collected during August 2010. Forty-four surface soil samples (from GR1 to GR44) across the CTP were collected at altitudes between 3711 m and 5352 m above sea level, and fallen into mountain shrubby steppe soils and alpine steppe soils. At each site, multiple natural soil samples were collected at a depth of 0–5 cm and then mixed, which were at least 500 m far from the main road to avoid direct vehicle impact. The detailed sampling and storage methods have been previously described (Yuan et al., 2014b), and the information of sampling sites is provided in Table S1 (Supplementary Materials). The 44 sampling sites were collected from four sub-areas (Fig. 1): (I) an area in Ngari Prefecture with a central town of Ngari, which is the third largest town in the northwestern area of the CTP that is less developed than Lhasa and Shigatse; (II) an area along the busiest travel route from Lhasa to Mount Everest including the two largest towns of Lhasa and Shigatse in the CTP, where the majority of the population in the CTP live along this route; (III) a small area between areas (I)

and (II); (IV) a narrow area along a 600 km northward sampling trajectory from Lhasa.

2.2. Chemicals

The 16 US EPA priority PAH standard solutions (1000 µg/mL) were products of Sigma–Aldrich, St. Louis, MO (USA), including naphthalene (NA), acenaphthylene (ACY), acenaphthene (ACE), fluorene (FL), phenanthrene (PHE), anthracene (ANT), fluoranthene (FLu), pyrene (PYR), benzo[a]anthracene (BaA), chrysene (CHR), benzo[b]fluoranthene (BbF), benzo[k]fluoranthene (BkF), benzo[a]pyrene (BaP), indeno[1,2,3-cd]pyrene (IcdP), dibenzo[a, h]anthracene (DBA), and benzo[ghi]perylene (BghiP). In addition, 2-fluorobiphenyl (2-FBP) was used as surrogate standard for the PAHs. Silica gel (100–200 mesh) (Qingdao Haiyang Chemical Co., Qindao, China) was activated in a drying oven at 550 °C for 6 h. Anhydrous sodium sulfate (Beijing Chemical Factory, China) was heated at 600 °C for 12 h to eliminate the remained organic contamination. The solvents were all of analytical or pesticide grade.

2.3. Extraction and cleanup

Ten grams of each soil sample was ground with anhydrous sodium sulfate into a free-flowing powder. Each sample was extracted with 30 mL of hexane/dichloromethane (1:1, vol/vol) by ultrasonication for 4 min and then centrifuged. This process was repeated three times, and the extracts were combined. The concentrated extracts were evaporated to 1 mL in a Kuderna-Danish concentrator under a gentle N₂ stream. Before extraction, 2-FBP was added as a surrogate standard. Extracts were cleaned by using a chromatography column (30 cm × 10 mm i.d.) containing 4 g of silica gel and 2 g of anhydrous sodium sulfate. The column was pre-eluted with 40 ml of hexane/dichloromethane (4:1, v/v) before loading the sample. The fraction containing the 16 PAHs was eluted using 60 ml of hexane/dichloromethane (4:1, v/v). The

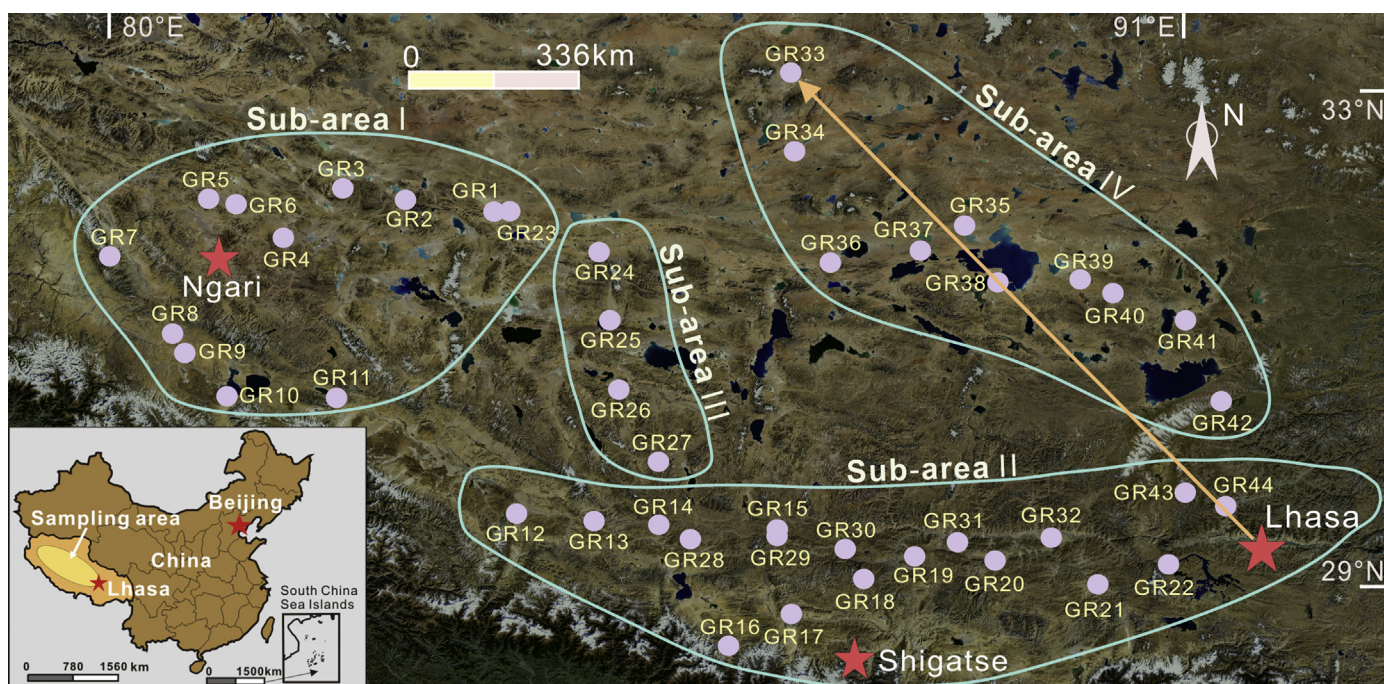


Fig. 1. Map showing the sampling site locations.

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