



The emerging source of polycyclic aromatic hydrocarbons from mining in the Tibetan Plateau: Distributions and contributions in background soils



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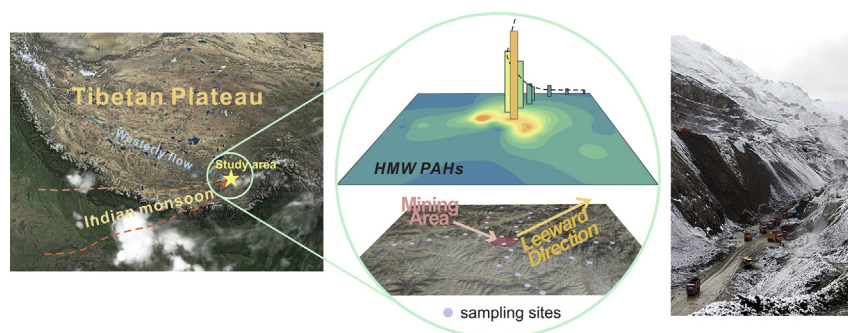
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HIGHLIGHTS

- Four sources of PAHs were quantitatively determined by the PMF method.
- The distribution of HMW PAHs was dominated by transport from mining emissions.
- The distribution of LMW and MMW PAHs was influenced by altitudinal “cold-trapping”.
- The contribution of HMW PAHs to soils from mining activity was estimated for the first time.

GRAPHICAL ABSTRACT



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ABSTRACT

The Tibetan Plateau (TP) serves as the background for persistent organic pollutants around the world. In addition to outside sources, local sources greatly contribute to the environment of the TP in recent decades. Mining activity could serve as an emerging source of polycyclic aromatic hydrocarbons (PAHs), although this issue has been neglected in the TP until now. To investigate the distributions and contributions of PAHs transported from mining activity, forty-one surface soil samples along six sampling directions were collected at a regular distance from the Jima mining area (JMA) of the TP at altitudes between 3726 and 4863 m. The total concentration of 16 PAHs was 52.34 ± 22.58 ng/g. The result of the source identification suggested that mining activity represented the primary source for heavy molecular weight (HMW) PAHs in soils, while light molecular weight (LMW) and middle molecular weight (MMW) PAHs were contributed by multiple sources. For HMW PAHs, the concentrations and proportions decreased logarithmically with transport distance from the JMA. Furthermore, the transport distance of HMW PAHs was found to be affected by the prevailing wind direction. In addition to transport from the source area, the distributions of LMW and MMW PAHs were also influenced by the altitude. In the impact area of Jima mining activity, the soil mass inventory was estimated to be 6.4 ± 0.8 tons for HMW PAHs. In future decades, HMW PAHs emitted from Jima mining activity are projected to exceed 5% of the annual local emission in the TP. Our study evidenced that Tibetan mining activity serves as an important emerging source of PAHs, which would be transported within the TP and threaten the fragile ecosystem of the TP.

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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are persistent organic pollutants (POPs), mainly resulting from combustion. Due to long-range

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atmospheric transport (LRAT) and “mountain cold-trapping”, much attention has been given to the accumulation and effects of PAHs at high latitudes (Friedman et al., 2014; Halsall et al., 1997; Vorkamp et al., 2010) and altitudes (Arellano et al., 2011; Chen et al., 2008; Wang et al., 2008; Wania and Westgate, 2008), such as at the Tibetan Plateau (TP). As “the third pole” of the earth, with an average elevation of higher than 4000 m, the TP covers a vast background area of approximately 2.5 million km² with a mean population density as low as 4.2 people per km² (Tao et al., 2011).

Traditionally, PAHs in the TP were mostly considered to be derived from LRAT (Wang et al., 2007). Their emissions were very limited within the TP during the last century due to the sparse population and minimal energy consumption (Yu et al., 2012). Nevertheless, a study on historical deposition has suggested that the increasing levels of PAHs in recent years are closely associated with Tibetan development (Yuan et al., 2016). Thus, some attention has been given to local sources of PAHs from the domestic burning and traffic emissions in the TP (Tao et al., 2011), which cannot be neglected. Most recently, Yuan et al., (2015) found that PAHs emitted from local sources were transported within the TP and were even dominate in the surface soils of some areas.

In addition to residential emissions, industrial activities have also developed rapidly in the TP, especially mining activity in the last decade (She et al., 2006; Zhang et al., 2010). This may pose a serious threat to the environment of the TP (Huang et al., 2010). For example, Tibetan mining produced 100 million tons of wastewater in 2007 and 18.8 million tons of solid waste in 2009 (Qiu, 2014). Regarding mining activity, most reports have focused on the environmental effects of heavy metals (Huang et al., 2010; Žibret and Šajn, 2008). Nevertheless, it is also reasonable to believe that mining activity, as an “emerging source”, would emit a substantial amount of PAHs and even yield environmental effects in the typical background region of the TP. Previous studies in heavily industrial areas have reported the occurrence of PAHs associated with mining sources (Achten and Hofmann, 2009; Ribeiro et al., 2012). Their analyses were mostly based on variations in the total concentration and diagnostic ratios to indicate the dominant contribution from coal or petroleum combustion related to mining (Bandowe et al., 2010; Liu et al., 2012; Pies et al., 2007; Yang et al., 2008). However, the elevated level of PAHs may not merely point to the contribution of mining activity, since a more significant amount of PAHs could be emitted from nearby plants (Bandowe et al., 2010). Various combinations of PAH compounds generated by agricultural and residential activities could also serve as point sources (Liu et al., 2012; Wang et al., 2013, 2015). Furthermore, although the diagnostic ratio is useful for identifying sources, its validity may be limited due to different reaction rates of compounds during atmospheric transport (Bandowe et al., 2010; Galarnau, 2008). Therefore, it is essential to investigate the distribution of PAHs around the mining area under background conditions and to reveal the possible contribution to PAHs in soils from mining activity by alternative approaches for source discrimination.

On the other hand, Halsall et al., (1997) reported that PAHs were transported several thousand kilometers from Eurasia to the Arctic. Yuan et al., (2015) reported that PAHs derived from domestic burning in Lhasa were transported at least 600 km along a northward trajectory. If massive PAHs were truly emitted from mining activity, they may be transported across the TP. Therefore, it is also significant to investigate the transport distance and the area impacted by PAHs from mining activity, which would help to understand the prospective inventory and their potential to be involved in global cycling.

As one of the largest skarn-porphry polymetallic deposits in China, the Jiama deposit is located in Maizhokunggar County in the TP and has been mined since 2006 (She et al., 2006). In this case, the Jiama mining area (JMA) was selected as a typical example to investigate the potential influence of mining activity as an “emerging source” of PAHs in the environment of the TP. For this purpose, PAHs in soils collected around the JMA were analyzed, since soils are the major reservoir of PAHs in the terrestrial environment (Cabrerizo et al., 2011; Komprda et al., 2013).

Our objectives were (1) to investigate the fractional pattern and the distribution of PAHs in soils from the study area, (2) to determine the contribution of mining activity to PAHs in soils through source identification, (3) to reveal the transport process and influencing factors on the distribution of PAHs, and (4) to estimate the amount of PAHs generated from mining activity in the surrounding soils and their prospective effects on the environment of the TP.

2. Materials and methods

2.1. Study site and sampling

The study area was approximately 4300 km² and centered on the JMA (Fig. 1). The JMA is located in the south of Maizhokunggar County and 100 km away from Lhasa in Tibet. In Maizhokunggar County, the wind time-series data showed that the prevailing wind direction was southwest, which occupied 26% of the total frequency (Fig. S1 in Supplementary material, SM). The wind time-series data were obtained from the observatory station located in the Maizhokunggar area provided by the National Meteorological Administration (<http://data.cma.cn>). This observation agreed with the transport path of air mass ending at the JMA according to a simulation of 10-day backward air mass trajectories (Fig. S2 in SM).

Forty-one surface soil samples (0–5 cm) were collected across the study area between 3726 m and 4863 m above sea level during July 2015. The 41 samples were collected along four cardinal directions and two leeward directions, including the east direction (De1–De7), the west direction (Dw1–Dw7), the north direction (Dn1–Dn7), the south direction (Ds1–Ds7), the northeast direction (Dne1–Dne8) and the southeast direction (Dse1–Dse5) (Fig. 1). In each direction, the samples were successively collected from the JMA with an interval of ca. 5 km. Among the samples, De1, Dw1, Dn1 and Ds1 were collected inside of the JMA, while Dne1 and Dse1 were collected at sites 5 km away from De1. At each site, five sub-samples were collected and mixed thoroughly into a composite sample to reduce random variations. The detailed sampling and storage methods were same as those previously described (Yuan et al., 2014, 2015), and information about the sites is outlined in Table S1 in SM. Details regarding the total organic carbon content, the particle size and the pH value are provided in SM, and the data for each individual site are provided in Table S2.

2.2. Chemicals

Standard solutions of the 16 PAHs were purchased from Cambridge Isotope Laboratories (Andover, MA), 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, the surrogate standards (naphthalene-d₈, acenaphthene-d₁₀, phenanthrene-d₁₀, and chrysene-d₁₂) and the internal standards (2-fluoro-1,1'-biphenyl (2-FBP) and *p*-terphenyl-d₁₄) were from Dr. Ehrenstorfer GmbH (Augsburg, Germany). Information on other reagents and solvents, including the producer, grade, and pre-treating process, was provided previously (Yuan et al., 2014, 2015).

2.3. Extraction and analysis

First, 10 g of the dry samples were freeze-dried and homogenized with anhydrous sodium sulfate. The mixtures were packed into a clean centrifuge tube, and then 20 ml of acetone/n-hexane (1:1, v/v) and the surrogate standard solutions were added. All samples were vortexed for 1 min, and then the mixture was ultra-sonicated for 15 min to extract the total extractable PAH residues. The tube was then centrifuged for 10 min at 2000 rpm. This process was repeated

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