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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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) an 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[g,h,i]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
solvent was evaporated for analysis to 30 μL in a K-D concentrator under a gentle N2 stream. Throughout the extraction, cleanup and analysis procedure, the analyses were protected from light by either wrapping the containers with aluminum foil or using amber glassware.

2.4. Analysis

The PAHs were measured with an Agilent 6890 gas chromatograph (GC) coupled with an Agilent 5973 mass spectrometer using electron impact ionization in the SIM mode. The separation was performed on a fused silica capillary column (DB-5MS, 30 m × 0.25 mm i.d., 0.25 μm film thickness). The GC column was programmed as follows: an initial temperature of 100 °C for 2 min, an increase to 160 °C at a rate of 10 °C/min, an increase to 230 °C at a rate of 4 °C/min, and then a 10 °C/min increase to 280 °C, which was maintained for 10 min.

2.5. Quality assurance and quality control (QA/QC)

All analytical procedures were monitored using strict quality assurance and control measures, and the detail methods were same as that reported (Yuan et al., 2014a). PAHs were quantified using an external-standard method (Yuan et al., 2014a). Three quality-control criteria were used to ensure the correct identification of the target compounds (Yuan et al., 2012a). The method detection limits (MDLs) for the PAHs were defined as a signal-to-noise ratio equal to three times the average baseline variation. A matrix spike test was run with six samples to further check for recoveries and standard deviations. The matrix spike recoveries of the standards were between 73 and 101%, with a standard deviation of 9%. The recoveries of the surrogate standards of 2-FBP ranged from 71% to 95% and averaged 79 ± 10% in the soil, and the reported concentrations were not corrected for the surrogate recovery. MDLs (defined as 3 S/N) were in the range of 0.01 – 0.48 ng/g dry weight (Table S2). Any concentrations lower than the MDLs were defined as non-detected (N.D.).

2.6. Analysis of the soil components and statistical methods

The soil minerals and soil organic carbon (SOC) were analyzed, and the detailed methods were described in our previous reports (Yuan et al., 2012a). The results for the SOC contents and soil minerals are shown in the Supplementary Material (Table S3 and Table S4). Pearson correlation analysis was performed using SPSS 19.0 for Windows with a sided-test.

3. Results and discussion

3.1. Concentration and composition

In the 44 soil samples, FLu was the most commonly detected compound with a detection frequency of 95.5%, and followed by PRY and Ph at detection frequencies of 90.9% and 86.4%, respectively (Table S2). The detection frequency of DBA was the lowest at 27.3%, followed by 29.6% for BghiP and 34.1% for BaP. Among 16 PAHs, NA was most abundant compound with an average concentration of 2.03 ng/g, and followed by PHE and PYR with average concentrations of 1.96 and 0.89 ng/g, respectively. The average concentration of IcdP was the lowest at 0.01 ng/g, and followed by 0.02 ng/g for both BaP and DBA.

The total 16 PAHs concentrations (Σ16PAHs) varied from 0.43 to 26.66 ng/g in the 44 samples with a mean value of 9.21 ng/g. The highest concentrations of Σ16PAHs were found to be at the sites of GR44 with 6 km away from Lhasa (Table 1). Tao et al. (2011) reported that the average concentration of PAHs was 58 ng/g in the area of Lhasa. In our case, the soils in the city of Lhasa were not sampled, in which Σ16PAHs should be higher than that observed at GR44. At site GR18 near the second largest town of Shigatse, the concentration of Σ16PAHs was high at 21.19 ng/g. At the GR42 site near Nan Co, Σ16PAHs was 6.37 ng/g, which was close to the previously reported value of 5.54 ng/g (Wang et al., 2014). Σ16PAHs was found to be 6.04 ng/g at the site of GR25 near the Changwengluozha Glacier, which was also close to the reported value of 5.96 ng/g (Yuan et al., 2014a).

As stated above, the samples were collected from four sub-areas (Fig. 1). For 11 soil samples of sub-area I, the concentration of Σ16PAHs ranged from 6.71 and 18.24 ng/g with an average of 13.55 ng/g (Fig. 2). Among the 16 PAHs, NA occupied 50.4% on average, followed by 20.2% for PHE and 10.0% for FL. The percentages of the other PAHs varied from 0 to 3.3%. There were 19 samples in sub-area II, and the levels of Σ16PAHs varied from 0.43 to 26.66 ng/g, with an average of 9.50 ng/g. The three highest values were 26.66, 22.64 and 21.19 ng/g (GR44, GR43 and GR18). The five most abundant PAHs were PRY (16.4%), BBF (16.0%), FLu (13.5%), PHE (13.0%) and CHR (11.8%). Sub-area III included 4 samples, and Σ16PAHs ranged between 2.79 and 5.57 ng/g with an average of 4.0 ng/g. The compounds of ANT and PHE occupied 59.4% and 19.6% respectively. Seven compounds were not detected, including BaA, BBF, BkF, BaP, IcdP, DBA, and BghiP, and the others were all less than 2%. For 10 soil samples in sub-area IV, Σ16PAHs ranged from 0.97 to 10.06 ng/g with an average of 5.96 ng/g. The average proportions of 5 compounds were over 9%, including 43.3% for PHE, 13.2% for FLu, 12.1% for FL, 11.3% for ANT, and 9.6% for PRY. The remaining 11 PAHs had proportions that varied between 0 and 2.8%. Although the concentrations of Σ16PAHs did not varied greatly in the four sub-areas (4.0 – 13.6 ng/g), the compositions between them were significantly different (Fig. 2), which might indicate the differently potential sources of the PAHs in each sub-area.

3.2. Geographical categories

The classification of the sub-areas in Fig. 1 was only based on the sampling areas. Although the composition of the PAHs seems different from one to another sub-area as mentioned above, the reliability of the classification has to be confirmed. Herein, principal component analysis (PCA) was applied for this purpose to look for relationships between the samples (Tao et al., 2011). The concentrations of the individual PAHs in each soil were used as the 16 variables for PCA. The results are shown in Table S6 (Supplementary Materials), and the plot of PC1 against PC2 is shown in Fig. 3. In the plot, the samples were clustered into four groups corresponding to the four sampling sub-areas (Fig. 1). In this case, the samples in the same group would share a similar PAH composition because the 16 variables in PCA were the composition of the 16 individual PAHs in each sample.

In the PCA plot, the 12 samples of sub-area I were all clustered in
group I. In addition to these 12 samples, GR36 was also included in Group I, which suggested that the compositions of GR36 were similar to those in sub-area I. Except for GR 31 and 32, the remaining 16 soil samples in sub-area II were clustered in Group II. Group III included GR24, GR25 and GR27 but not GR26 in sub-area III. Except for GR36 clustered in Group I, the remaining 9 soils in sub-area IV were all clustered into Group IV. Thus, the spatial classification of the sub-areas in Fig. 1 was at least supported by the PCA because most of the soils in the same sub-area were clustered into the same group in Fig. 3. As discussed above, the soils in the same group would share a similar composition of the PAHs, while the soils in a different group might have a different PAH composition. In other words, the PAH compositions were statistically different from one to another sub-area instead of a simple division of sampling area. Therefore, it is also reasonable to discuss the PAH composition and potential sources of the four sub-areas one by one.

3.3. Fractional patterns and potential sources in sub-areas

Although the Indian monsoon may transport PAHs into the south TP by LRAT (Sheng et al., 2013), Tao et al. (2011) suggested that the extensive Himalayan range formed a barrier that restricts the penetration of southwesterly monsoon air masses into the CTP region. In the CTP region, the burning of yak/sheep dung is a common domestic activity. Additionally, the use of liquefied gas has recently increased in large towns, such as Lhasa and Shigatse. Up to 2010, the number of tourists increased to 6.85 million, which is two times the population of Tibet (Yu et al., 2012). Moreover, most of tourism activities were focused in sub-area II (Lhasa to Shigatse to Mount Everest), which should increase the emission of PAHs from traffic. At the same time, the inefficient combustion would enhance the yields of PAHs because the oxygen levels throughout the CTP are relatively low.

Molecular diagnostic ratios are often used for the identification of PAH sources (Katsoyiannis et al., 2011; Tobiszewski and Namiesnik, 2012; Liu et al., 2013). The descriptive statistics of six ratios for the 44 samples are summarized in Table S7 (Supplementary Materials). Some ratios have been used to identify the origin of PAHs as being from combustion sources or petroleum input, fuel or coal/biomass combustion, or traffic-related sources (Katsoyiannis et al., 2011; Yuan et al., 2014c). The ratio of BbF/(BbF + BkF) had even been used to distinguish between coal and biomass combustion (Dickhut et al., 2000). In this case, the values of specific ratio varied greatly in the samples of each sub-area and cannot provide clear information as to the sources of the PAHs (Table S7). Additionally, due to the relatively low detection frequencies for some compounds in this case, the valuable values in the 44 samples are few for some ratios. Moreover, other studies have highlighted the wide range in the emission factors and compound ratios from given source categories (Shen et al., 2012, 2013). Thus, the main sources of PAHs in each sub-area cannot be clearly identified from the molecular diagnostic ratios.
Nevertheless, the proportions of various PAH fractions, including light, intermediate and high molecular weight PAHs, have been applied to identify the possible origins of PAHs. Commonly, the combustion of coal or biomass would dominantly emit light (2 and 3 rings) and intermediate (4 rings) molecular weight PAHs, whereas the high molecular weight PAHs (5 and 6 rings) would mainly be associated with traffic-related emission (Yang et al., 2011; Li et al., 2012; Liu et al., 2012, 2013; Shen et al., 2012). The fractional patterns of the 44 samples are shown in Fig. 4, of which four sub-areas were different from one to another. The difference reversely indicated the existence of local source. If PAHs all be inputted by LRAT from outside, the fractional patterns should be similar or at least related in CTP.

In sub-area I including the third largest town of Ngari, the proportion of (2 + 3)-ring PAHs varied from 74.3% to 98.1% with an average of 91.5%. The proportions of 4- and (5 + 6)-ring PAHs were 6.9% and 1.6%, respectively (Fig. 4). Shen et al. (2012) found that (2 + 3)-ring PAHs were the dominant PAHs emitted from the burning of biomass. In this case, the emission profiles were also similar to that of yak/sheep dung burning (Lu et al., 2006). As mentioned above, the area is far away from the central towns and motorized vehicles are inconvenient. Most of the local emission of PAHs should originate from the domestic burning of yak/sheep dung. The high concentration of \( \Sigma_{16} \) PAHs also suggested that the PAHs mainly originated from local sources, as this concentration was even higher than that of sub-area II where the large towns of Lhasa and Shigatse were included. In sub-area II, the proportion of (5 + 6)-ring PAHs occupied 23.3%, which was higher than 22.6% for (2 + 3)-ring PAHs. 4-ring PAHs were the most abundant at 54.1% (Fig. 4). Because sub-area II included the first and second largest towns and the busiest travel line of Lhasa-Shigatse–Mount Everest, traffic emissions should be massive. Thus, the high proportions of 4- and (5 + 6)-ring PAHs were mainly attributed to traffic-related sources (Yang et al., 2011; Liu et al., 2013).

The location of sub-area III is between sub-areas I and II, and the profiles of PAHs should be influenced by emissions of the former two areas. The fractional proportions for light, intermediate and high molecular weight PAHs were of 85.3%, 14.6% and less than 0.1%, respectively (Fig. 4). Compared with sub-area I, the increase of the intermediate fraction in sub-area III might be due to transport from sub-area II, which will be discussed in following section.

The fractional proportions of PAHs in sub-area IV were 72.0% for light, 26.3% for intermediate and 1.7% for high molecular weight PAHs, respectively (Fig. 4). Compared with sub-area I, the proportion of 4-ring PAHs obviously increased while the proportion of (2 + 3)-ring PAHs decreased. Similar to that in sub-area III, the increase of the 4-ring PAHs might be due to transport from sub-area II, which is discussed in following sections. Different from sub-area I, there is no large town along the sampling transect in sub-area IV. So, local emission should be very limited in the middle of transect.

### 3.4. Transport of local sources from Lhasa

Concentrations of POPs in the environment typically decrease with increasing distance from the source (Gouin et al., 2004; Wania and Westgate, 2008). As discussed above, the PAHs in sub-area IV may be transported from sub-area II. Undoubtedly, Lhasa in sub-area II would be the biggest local source of PAH emission (Fig. 2). In this case, a trajectory was selected to study the transport of PAHs from Lhasa to a great distance away, which included GR44 and GR43 near Lhasa and 9 samples of sub-area IV (Fig. 1). GR36 was excluded from this trajectory because it was not clustered into same group with the other sites of sub-area IV by PCA (Fig. 3). The detailed characteristics of the soils and sampling sites along the trajectory are summarized in Table S8 (Supplementary Materials).

As shown in Fig. 5A, the concentrations of the 16 PAHs logarithmically decreased with increases in the distance from Lhasa. The distance was 86.6 km from the source for half the concentration, 280.2 km for 1/4 concentration, and 504.1 km for 1/8 concentration. Additionally, the concentration of each PAHs fraction also decreased with increase of distances from Lhasa (Table S8). The logarithmic decrease of the PAHs concentrations with increase of the transport distances from the source site was similar to the changing trend modeled for other POPs (Gouin et al., 2004). With increasing distance from Lhasa, the proportion of light molecular
weight PAHs logarithmically increased and those of the intermediate and high molecular weight PAHs logarithmically decreased (Fig. 5B). Furthermore, the concentration ratios of (2 + 3)-rings to (4 + 5+6)-rings were positively correlated with the transport distance from Lhasa (Fig. 5C). Light molecular weight PAHs are more prone to transport from the source site than high molecular weight PAHs (Tao et al., 2011), thus PAHs were gradually fractionated with increase of transport distances from the source site (Gouin et al., 2004). Such results reversely validated that PAHs emitted from south Tibet, especially from Lhasa, have served as local sources and fractional proportions of the PAHs (Table 2). Along the trajectory, latitude and altitude were positively correlated with the transport distance from Lhasa, and longitude was negatively correlated (Fig. 1). Thus, the relationships between the concentration of PAHs and these geographic factors can be reasonably understood.

Along the same trajectory, polybrominated diphenyl ethers (PBDEs) have been found to increase with increasing altitude (Yuan et al., 2012b). PBDEs concentrations in soils were controlled by mountain cold-trapping while inputted from outside of the CTP by LRAT. PAHs, however, mainly originated from local sources, and their concentrations were dominated by the transported distances from source site. Once again, the changes in the concentrations of the PAHs with transported distance from Lhasa validated that the local sources mainly influenced the concentrations of PAHs in soils rather than PAHs transported from outside of the CTP by LRAT. If this was not the case, the concentration of PAHs should increase with increasing altitude along the trajectory or in CTP as the maniers of PBDEs, organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) (Yuan et al., 2012b, 2014b, 2015).

### Table 2

Pearson correlation coefficients (n = 11).

<table>
<thead>
<tr>
<th></th>
<th>Distance (km)</th>
<th>Altitude (m)</th>
<th>Latitude (N)</th>
<th>Longitude (E)</th>
<th>SOC (%)</th>
<th>Clays (%)</th>
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<tr>
<td>( \sum_{i} \text{PAHs} ) (ng/g)</td>
<td>-0.797&lt;sup&gt;a&lt;/sup&gt;</td>
<td>-0.893&lt;sup&gt;a&lt;/sup&gt;</td>
<td>-0.826&lt;sup&gt;a&lt;/sup&gt;</td>
<td>0.669&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.067</td>
<td>-0.122</td>
</tr>
<tr>
<td>( \sum_{2-3} \text{rings} / \sum_{i} \text{PAHs} ) (%)</td>
<td>0.839&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.963&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.893&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.725&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.197</td>
<td>0.197</td>
</tr>
<tr>
<td>( \sum_{4-5} \text{rings} / \sum_{i} \text{PAHs} ) (%)</td>
<td>-0.833&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.905&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.909&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.752&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.211</td>
<td>0.326</td>
</tr>
<tr>
<td>( \sum_{5-6} \text{rings} / \sum_{i} \text{PAHs} ) (%)</td>
<td>-0.706&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.854&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.730&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.581</td>
<td>-0.153</td>
<td>-0.149</td>
</tr>
<tr>
<td>( \sum_{4-5+6} \text{rings} / \sum_{i} \text{PAHs} ) (%)</td>
<td>-0.839&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.963&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.893&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.725&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.197</td>
<td>0.081</td>
</tr>
<tr>
<td>( \sum_{2-3} \text{rings} / \sum_{4-5+6} \text{rings} ) (%)</td>
<td>0.921&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.824&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.961&lt;sup&gt;b&lt;/sup&gt;</td>
<td>-0.850&lt;sup&gt;b&lt;/sup&gt;</td>
<td>0.376</td>
<td>-0.345</td>
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<sup>a</sup> Correlation is significant at the 0.01 level (2-tailed).
<sup>b</sup> Correlation is significant at the 0.05 level (2-tailed).
4. Conclusions

Forty-four soil samples were collected from the CTP, and the concentrations of PAHs ranged from 0.43 to 26.66 ng/g. According to the results of PCA, the samples were divided into four sub-areas, which was consistent with the spatial distribution. The major sources of PAHs were distinguished by the fractional proportions in each sub-area, although molecular diagnostic ratios were not effective for identifying sources. In sub-area I, the PAHs originated mainly from the domestic burning of yak/sheep dung. In sub-area II, PAH sources were dominated by traffic emission and domestic burning. The PAHs in soil samples of sub-area IV were found to be transported from a source in sub-area II. Along a 600 km northward sampling trajectory from Lhasa, the PAH concentrations logarithmically decreased with increasing distances from the source. Meanwhile, the proportion of the light fraction increased logarithmically with increasing distances from the source, while the fractions of the intermediate and heavy fractions decreased logarithmically. The concentrations and fractional proportions of PAHs were also found to be related to the latitude, longitude and altitude since these geographic characteristics were all significantly correlated with the distance of source transport. Based on the study of source identification and transport, it was concluded that PAHs sources transported within CTP and dominated their concentrations in soils, but few of them were transported outside the CTP or TP. Due to the relatively low levels of PAHs, the soils in the CTP can be considered as background soils and serve as a “sink” for PAHs in global cycling.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.envpol.2015.04.002.

References


