

Article

The Transport and Distribution of Polycyclic Aromatic Hydrocarbons (PAHs) Across the Hengduan Mountains, Southwest China

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Abstract

Despite recent advances in polycyclic aromatic hydrocarbon (PAH) research on the Tibetan Plateau (TP), studies investigating the transport potential and accumulation dynamics of these contaminants in the Hengduan Mountains, especially in forest soils which are important sinks for atmospheric PAHs, remain scarce. In the present study, soil and lichen samples (partially located under the forest canopy) were concurrently collected from 62 sampling sites across the Hengduan Mountains to characterize the occurrence, spatial distribution patterns, and underlying controlling factors of PAHs. The total concentrations of the 16 US EPA priority PAHs ($\Sigma_{16}\text{PAHs}$) in soils and lichens ranged from 59.8 to 1163 ng/g and 174 to 3362 ng/g, respectively—values consistently higher than those reported in corresponding matrices from the northern and northwestern TP. Further, concentrations of PAHs in both soil and lichen under the forest canopy are significantly higher than those on the leeward slope without forest. Compositional fractionation of PAHs along the longitudinal and latitudinal gradients of sampling locations indicates significant modulation of PAH distribution by both the Indian monsoon and East Asian monsoon, a pattern further corroborated by air mass backward trajectory analysis. Our results confirm that PAHs can be transported to the southeastern TP slope via long-range atmospheric transport (LRAT). Notably, the combined effects of mountain cold-trapping and forest filtering jointly govern the deposition and spatial distribution of PAHs in this region.

Keywords: polycyclic aromatic hydrocarbons (PAHs); Hengduan Mountains; long-range atmospheric transport (LRAT); mountain cold-trapping effect; forest filtering effect



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1. Introduction

Polycyclic aromatic hydrocarbons (PAHs) are prototypical semi-volatile organic compounds (SVOCs) that constitute a prominent global environmental concern, owing to their strong bioaccumulative potential, inherent toxicity, and potential for long-range atmospheric transport (LRAT) [1]. As a class of carcinogenic organic compounds, PAHs have been designated as priority pollutants by the U.S. Environmental Protection Agency (EPA). Generally, these contaminants are ubiquitous in the environment and strongly correlated

with regional energy structure, energy consumption intensity, and degree of industrialization and urbanization [2]. Notably, the atmosphere serves as the dominant transport pathway driving the global distribution of SVOCs [3,4]. Owing to LRAT, PAHs have been ubiquitously detected across diverse environmental matrices, including remote, historically considered pristine areas like the polar regions [5,6], open oceans [7], and Tibetan Plateau (TP) [8].

Monsoons, characterized as a long-duration, large-scale atmospheric circulation system, play a pivotal role in modulating the fate and transport processes of SVOCs [9]. As reported by Lavin et al. (2013) [10], northwesterly global-scale wind systems drive the transboundary transport of SVOCs originating from background atmospheric reservoirs across Australia and the Southern Hemisphere, with this atmospheric process directly modulating the pesticide profile signatures at high-elevation monitoring sites located in the western region of the Southern Alps. Tian et al. [11] established a clear causal link between the East Asian summer monsoon and the directional transport of hexachlorocyclohexanes (HCHs), demonstrating that this large-scale circulation system acts as a key conduit for HCH migration from southeastern to northeastern China. Multi-year monitoring data further revealed marked seasonal variability in SVOC concentrations across the southeastern TP, with these temporal trends showing a statistically significant association with the recurrent phase shifts in the Indian monsoon [2].

However, the LRAT and deposition of SVOCs in remote mountain ecosystems remain poorly constrained by a range of critical modulating factors: land cover and topographic heterogeneity, local anthropogenic or biogenic emissions, and spatial proximity to potential primary or secondary source regions [12]. Complementarily, findings from our prior investigations demonstrated that HCH and dichlorodiphenyltrichloroethane (DDT) residue concentrations in coniferous needles exhibited a statistically significant positive correlation with elevation across the southeastern TP [13]. Similarly, Zhu et al. [14] demonstrated that forests function as effective filters for SVOCs via comparative analyses of contaminant concentrations between forested stands and adjacent clearings. Against the backdrop of the aforementioned complexity in SVOC LRAT and deposition within remote mountain ecosystems, further targeted research is therefore warranted to elucidate the mechanistic underpinnings of SVOC migration pathways and identify the key biotic and abiotic factors governing the environmental fate of SVOCs in these sensitive high-elevation regions.

Notably, the TP—the largest and highest plateau on the planet—spans an area of 2.5 million square kilometers and boasts an average elevation exceeding 4000 m above sea level (a.s.l.). Its distinctive meteorological and topographic attributes collectively give rise to an extreme cold climate, coupled with sparse human habitation and negligible industrial and agricultural activities, thereby rendering the region a pristine, minimally disturbed alpine system ideal for investigating the LRAT of SVOCs. The Hengduan Mountains (Mts.) constitute a contiguous mountain range in southwest China and are characterized by typical parallel ridge–valley topographies, serving as a critical topographic link to the southeastern margin of the Tibetan Plateau (TP). This region is characterized by a typical monsoonal climate, where moisture transport exhibits distinct seasonal variability. Specifically, summer moisture is predominantly derived from the Bay of Bengal and the South China Sea, whereas autumn moisture mainly originates from the western Pacific Ocean. In contrast, the northern hemisphere westerlies deliver only limited water vapor to the region during winter and spring [9,15,16]. Together, the interplay between the complex topography and the monsoonal circulation makes the Hengduan Mountains the first site where we can decouple elevation-driven cold-trapping from valley-channeled advection of PAHs—a critical missing link for improving LRAT models beyond current polar- and alpine-centric frameworks [17].

While numerous studies have focused on the transport and distribution of PAHs across most regions of the TP, research remains scarce for the Hengduan Mountains—a critical southeastern TP component—where atmospheric circulation and geographical patterns are more complex than other TP regions due to unique topographic heterogeneity and the synergistic monsoon influences discussed earlier, presenting a distinctive opportunity to elucidate how global circulation dynamics regulate organic contaminant transport and address key gaps in regional and global SVOC cycling. To fill this void and advance understanding of PAH environmental behavior in this understudied alpine area, paired soil (as a contaminant sink) [18] and lichen (as a passive atmospheric sampler) [19,20] samples were collected from 62 sites across the Hengduan Mountains. There are three core objectives of this work: (1) characterize PAH concentrations and spatial distribution patterns in these matrices; (2) identify key biotic, abiotic, and meteorological factors governing PAH distribution; (3) delineate potential sources and dominant transport pathways, thereby providing empirical evidence for linkages between global circulation and contaminant accumulation in remote mountain ecosystems, complementing existing TP PAH research and offering novel insights for global environmental monitoring and pollution management.

2. Experimental Section

2.1. Study Area and Sampling

The Hengduan Mts. cover the easternmost section of the Tibet as well as the western Yunnan and Sichuan Provinces, touching upon parts of the southern Qinghai Province. Generally, the Hengduan Mts. are mainly controlled by two climate systems including the southwest monsoon of the Bay of Bengal and Indian Ocean, as well as the southeast monsoon from the western Pacific, which is the main precipitation air mass in summer [9,17]; and the westerly wind of the northern hemisphere in winter, carrying less water vapor. The sampling sites were located in the mountain slopes and stretch over three provincial areas, namely Yunnan, Tibet and Qinghai (Figure 1).

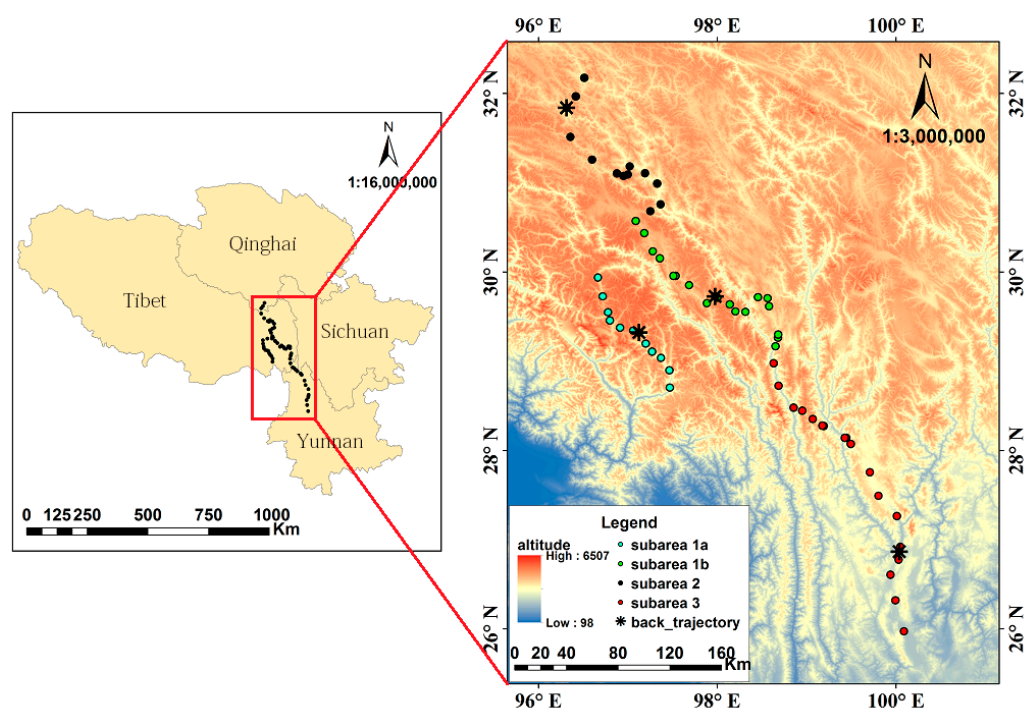


Figure 1. Geographic location and sampling sites. Numerical information is summarized in the Supporting Information, Table S1.

A total of 62 sample locations were selected across the south-to-north Hengduan Mts. at an altitude between 1930 m and 5094 m. Monsoonal circulation in the Hengduan Mts. expands to 40° N at the beginning of August but withdraws to 30° N in October. In order to learn more about the spatial changes in PAH concentrations, the study area was spatially divided into three sub-areas including the “central” (sub-area 1a and sub-area 1b), “northern” (sub-area 2), and “southern” (sub-area 3) Hengduan Mts, based on the meteorological conditions and the specific location (Figure 1). Sub-area 1a and sub-area 1b are located in Tibet. Sub-area 2 extends from Tibet to Qinghai and most of the sampling points are located in Tibet. Sub-area 3 is located in Yunnan and adjacent to urban areas and potential source areas.

Surface soil samples (0–5 cm) were collected using a stainless steel spade in August, 2013. Before collecting the soils, superficial litter and large roots, if present, were removed before the sample was taken. Composite lichen samples were gathered at 3–4 positions close to the soil site. All samples were sealed in zippered plastic bags individually and transported to the laboratory where they were stored at −20 °C until analysis. The geographical locations, elevations and related information are provided in Table S1 in the Supporting Information (SI).

2.2. Chemical Analysis

Sample pretreatment procedures and instrumental analysis are presented in detail in Text S1 of the SI. Briefly, soil and lichen samples were spiked with surrogate standards after being freeze-dried, ground and crushed, and then extracted using mixed solvents *n*-hexane and dichloromethane (1:1, *v/v*) by accelerated solvent extraction (Dionex ASE 350, Thermo Scientific, Waltham, MA, USA). The extracts were purified using multiple columns packed with 6 g of deactivated silica gel (3% of water), 4 g of deactivated aluminum oxide (2% of water) and 5 g anhydrous sodium sulfate from bottom to top. The eluted solvent was 50 mL of mixed hexane with dichloromethane in a ratio of 3:2 (*v/v*). The lichen extracts were further treated with gel permeation chromatography (GPC). The eluents were finally concentrated to 0.2 mL and spiked with injection standards of 2-fluorobiphenyl before instrumental analysis.

Concentrations of PAHs were measured with an Agilent gas chromatograph 7890 coupled with mass spectrometer 5975 (GC-MS, Agilent Technologies, Pimpri-Chinchwad, MA, USA). A DB-5 capillary column (30 m × 0.25 mm i.d. × 0.25 µm film thickness) was used for separation and an electron ionization (EI) source was used for detection. The MS was operated in selective ion monitoring (SIM) mode. Quantification was conducted by the internal standard method. The detailed instrumental analysis is described in Text S1 of the SI.

Total organic carbon (TOC) contents in soils were analyzed using a TOC analyzer (TOC-V, Shimadzu, Kyoto, Japan). The lipid content of lichen was determined gravimetrically.

2.3. Quality Control

A procedural blank was run every set of eight samples. Method detection limits (MDLs) were in the range of 0.04–0.97 ng/g dw, which was derived as 3 times the signal-to-noise value (S/N). The average recoveries of spiked surrogates in all samples were 62.7% ± 11.1%, 73.5% ± 10.1%, 89.9% ± 10.3% and 110.5% ± 16.6% for the deuterated PAHs. All the reported values were blank-corrected and subsequently corrected using the recoveries of the surrogates.

2.4. Air Back-Trajectories

The pathways of air mass to the sampling sites were determined to assess potential pollution source regions using NOAA’s hybrid single-particle Lagrangian integrated tra-

jectory (HYSPLIT) model accessed on 5 January 2012, 15 July 2012, and 24 August 2012 (<http://www.arl.noaa.gov/ready/hysplit4.html>). Five-day air mass back-trajectories with a starting height of 500 m were generated for each sampling period. The geographical coordinates of each representative sampling point for the sub-areas were used to simulate air mass transport (Figure 1).

3. Results and Discussion

3.1. General Remark on Concentrations of PAHs

Statistical dry-weight-based (dw) concentrations of PAHs are shown in Table 1. TOC contents in soil and lipid contents in lichen are listed in Table S1. A comparison of PAH concentrations in this study with those reported in other remote areas is presented in Table S2.

Table 1. Statistical concentrations (min-max, mean, and ng/g dw) of PAHs in soil and lichen.

Compounds		Soil	Lichen
2–3 rings	NAP	19.4–1041 (142)	34.3–1278 (336)
	ACY	0.52–6.94 (1.86)	0.36–13.2 (4.10)
	ACP	0.75–20.8 (4.48)	3.85–105 (40.5)
	FLR	2.40–70.4 (13.5)	2.69–59.8 (18.0)
	PHE	2.07–245 (24.4)	6.22–188 (47.9)
	ANT	0.24–25.1 (2.50)	1.03–112 (39.7)
4 rings	FLT	0.08–69.7 (6.49)	5.12–776 (157)
	PYR	0.35–47.5 (4.43)	0.88–62.9 (11.8)
	BaA	0.31–26.7 (2.35)	5.38–500 (58.4)
	CHR	0.53–44.4 (3.70)	1.56–297 (35.6)
5–6 rings	BbF	0.24–66.5 (4.43)	0.26–206 (13.6)
	BkF	0.08–22.1 (1.67)	14.4–185 (65.9)
	BaP	0.31–36.2 (2.45)	0.35–455 (49.5)
	IcdP	0.03–34.9 (2.32)	1.75–657 (37.5)
	DahA	0.08–6.79 (0.47)	0.64–195 (14.1)
	BghiP	0.25–24.7 (1.98)	0.76–128 (13.3)
Σ_{16} PAH		60–1163 (223)	174–3362 (871)

The concentrations of 16 US EPA priority PAHs (Σ_{16} PAHs) in soil and lichen samples ranged from 59.8 ng/g to 1163.3 ng/g (mean: 222.5 ng/g) and from 174.4 ng/g to 3362.1 ng/g (mean: 871.0 ng/g), respectively. Naphthalene (NAP) was identified as the most abundant congener, with an average value of 143.1 ng/g in soil and 358.3 ng/g in lichen. Low-molecular-weight (LMW) PAHs containing 2–3 aromatic rings dominated the compositional profiles, which accounted for an average of 83.8% and 72.6% of Σ_{16} PAHs in soil and lichen, respectively. In the same sampling sites, PAH concentrations in lichen samples were significantly higher than those in soil samples (Figure 2a), suggesting a pronounced capacity of lichen for PAH bioaccumulation. Even after normalization to TOC content or lichen lipid content, this trend of elevated PAH levels in lichen relative to soil remained consistent (Figure 2b). PAH deposition in both matrices is a combined process involving both particle-associated and vapor phase fractions. Notably, the proportion of high-molecular-weight (HMW) PAHs (i.e., 4-ring and 5–6-ring congeners) in lichen was higher than that in soil (Figure 2c), implying that HMW PAHs are more prone to be taken up and retained by the lichen matrix than by the soil matrix. Lichen, characterized by a large specific surface area and the absence of a protective cuticular layer, can directly sorb atmosphere contaminants via surface interactions [21]. The surface adsorption process may predominate over lipid-mediated PAH accumulation, thereby enabling lichen to scavenge

a greater quantity of particulate-phase PAHs from the atmosphere. This mechanism likely explains the observed higher relative abundance of 4–6-ring PAHs in lichen compared with soil.

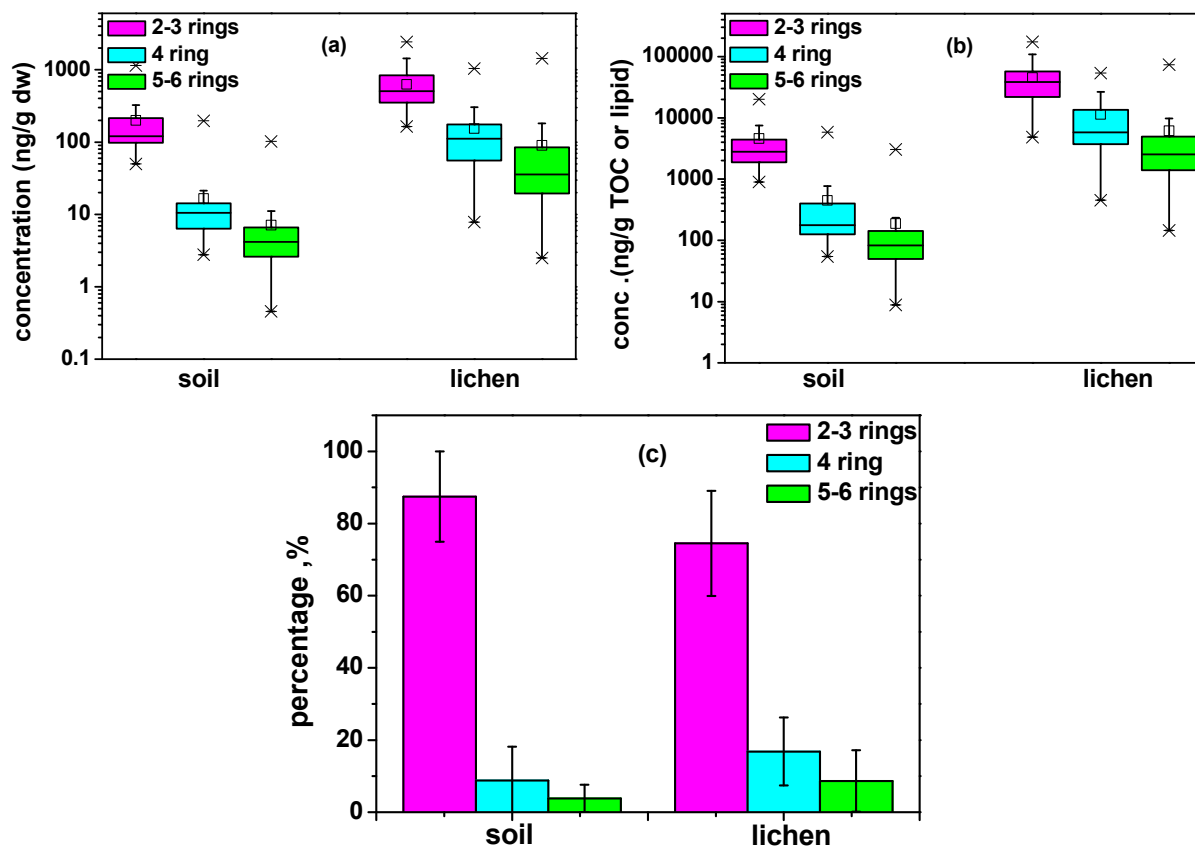


Figure 2. Comparison of PAHs (a–c) in soils and lichen on both a dry-weight and TOC- or lipid-weight basis. The box represents the data between the 25th and 75th percentiles. The small box and the horizontal line inside the box indicate the mean and median data. The whiskers above and below the box indicate the 95th and 5th percentiles, and the symbol “x” above and below them represents the outliers.

PAH concentrations in the soils investigated herein were distinctly lower than those reported in urban regions, such as the north-central part of India [22] and Kathmandu in Nepal [23]. By contrast, these soil PAH concentrations were one to two orders of magnitude higher than the levels documented in the central TP [24] and the west or northwest TP [25], while being comparable to those in soils of the southeast TP [26]. Additionally, the soil PAH concentrations in this study were comparable to the values measured in soils from the Changbai Mountains [27] and the Dawangling alpine ecosystem in China [28], yet substantially higher than the corresponding levels reported in the central Italian Alps [29], Costa Rica [30], and western Canada [31]. For lichen samples, PAH concentrations were significantly higher than those in the western TP [32] and comparable to the levels detected in the Pyrenees mountains in Europe [33] and in the Dolomites, Italy [34]. Furthermore, lichen PAH concentrations were marginally higher than the values in the southeast TP [8] and one to two orders of magnitude higher than those in the remote Antarctic Peninsula [35].

3.2. Spatial Distribution

In general, concentrations of HMW PAHs (4–6 rings) in soil and lichen from the southern segment of the study area (i.e., sub-area 3) were higher than those detected in the central (sub-area 1a and 1b) and northern (sub-area 2) segment. By contrast, LMW PAHs (2–3 rings) exhibited no such latitudinal distribution pattern (Figure 3). Along the south-to-north transect, a marked increase in the relative proportion of volatile LMW PAHs (2–3 rings) with increasing latitude was observed in both soil and lichen samples (Figure S1); conversely, the proportion of HMW PAHs (4 rings and 5–6 rings) decreased progressively with latitude (Figures 3 and S1). These observations imply that compositional fractionation of PAHs occurred during LRAT, driven by the prevailing Indian and East Asian monsoons [2,17,26,36]. Specifically, HMW PAHs tend to be preferentially deposited in proximity to emission sources, whereas LMW PAHs are more susceptible to LRAT. These results confirm that PAHs emitted from source regions in the southern Hengduan Mts. region—including urban areas in Yunnan and pollutants transported from the Indian subcontinent—act as both local contamination sources and precursors for LRAT. Concomitantly, the altitude of sampling sites exhibited a general increasing trend along the south-to-north latitudinal gradient (Figure 1). Furthermore, PAH fractionation during atmospheric transport was characterized by the preferential deposition of LMW PAHs, particularly in cold climatic conditions [35,37–39]. Collectively, particulate scavenging and cold condensation are proposed as the major mechanisms governing the compositional fractionation of PAHs along the studied latitudinal profile.

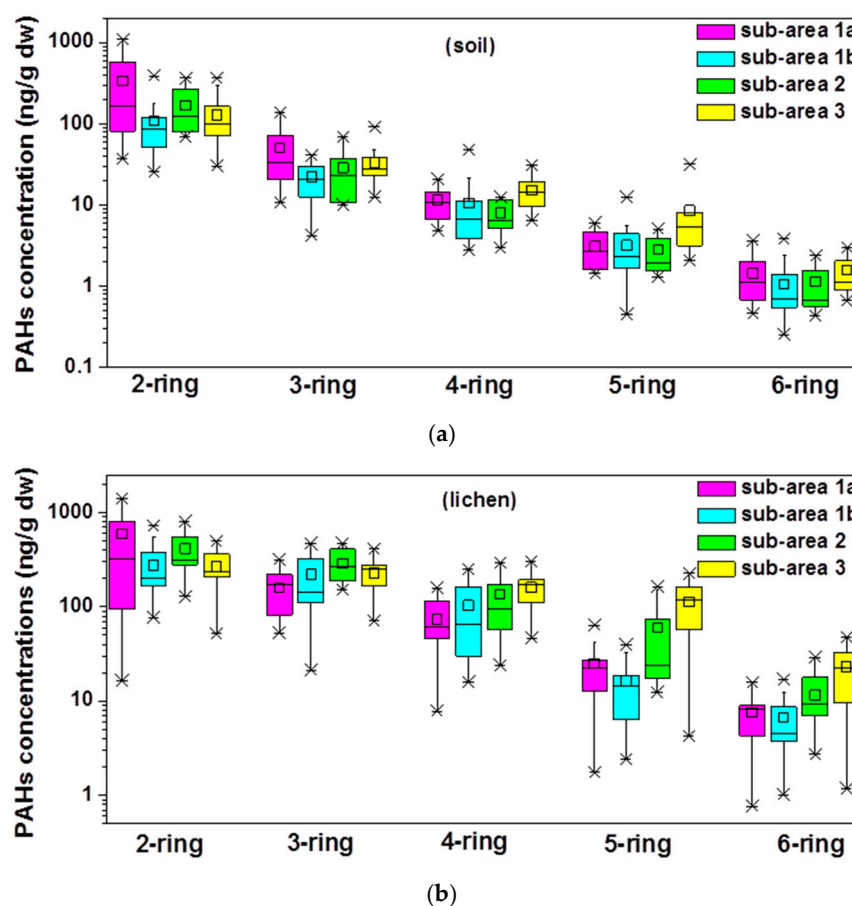


Figure 3. The distribution of PAHs in soils (a) and lichen (b).

Principal component analysis (PCA) was employed to elucidate associations among PAH pollution profiles of sampling sites across different sub-areas. Three distinct clusters were identified from the PCA results (Figure S2). Sub-area 1a and sub-area 1b were grouped into the same cluster, indicating potentially similar PAH sources. In contrast, sub-areas 2 and 3 formed separate clusters, both distinct from the sub-area 1 cluster. This suggests significant differences in PAH distribution patterns and source contributions among the sub-areas, further verifying the rationality of the sub-area division in this study.

Sub-area 1a, located in the central Hengduan Mts., features a mountain-valley topography that functions as a channel for water and vapor transport. Sampling sites in sub-area 1b are roughly parallel to the mountain-valley corridors of sub-area 1a. According to the air back-trajectories in Figure S3, southwest winds and westerlies are the dominant air currents, prevailing in the sub-areas 1a and 1b year-round. Consistent with this, PCA results further corroborated that sub-areas 1a and 1b exhibit highly consistent PAH distribution patterns (Figure S2), likely attributed to their analogous topographic conditions and dominant airflow influences.

Sub-area 3, located in the southern Hengduan Mts, is in close proximity to the urban regions of Yunnan Province—areas with high population density, elevated energy consumption, and consequently enhanced PAH emissions [32]. Air masses originating from Yunnan are likely responsible for the significantly higher HMW PAH concentrations in sub-area 3 (Figure S3c) [40]. For sub-area 2 in the northern Hengduan Mts, the climate is primarily regulated by the Indian monsoon and westerlies, with additional impacts from secondary airflows (Figure S3c). These secondary currents (northwesterly and northeasterly winds) pass through Xinjiang and Qinghai, implying that energy consumption patterns in these regions may also modulate PAH distribution in sub-area 2.

3.3. Identifying Driving Factors on Deposition in Mountain Slopes

3.3.1. Comparison Between Windward and Leeward

Sub-area 1a is situated in a mountain valley, which typically serves as the main channel for warm, humid air currents driven by the Indian monsoon. This valley is composed of a windward mountain slope and a leeward mountain slope. Notably, PAH concentrations in both soil and lichen on the windward slope are significantly higher than those on the leeward slope (Figure 4). This discrepancy is attributable to the windward slope being more strongly influenced by the humid summer monsoon along the valley, compared to the leeward slope [41]. On the one hand, precipitation on the windward side is greater than that on the leeward side [42], contributing to more intensive wet deposition of PAHs on the windward side. In mountainous regions, increasing precipitation at high altitudes can enhance the accumulation of semi-volatile organic pollutants at high altitudes [43]. On the other hand, the land cover types differ substantially between the two slopes: the windward slope is covered by forests, which can scavenge PAHs from the air and transfer them into the soil, whereas the leeward slope features a semi-arid meadow environment due to less precipitation [10]. In addition, the diurnal wind pattern—characterized by upslope winds during the warm daytime and downslope winds at night—also plays a role in regulating PAH deposition on the two slopes. A previous study conducted on south Island of New Zealand similarly documented that diurnal wind patterns may exert a stronger influence on the leeward slope than the windward slope of the mountain [10].

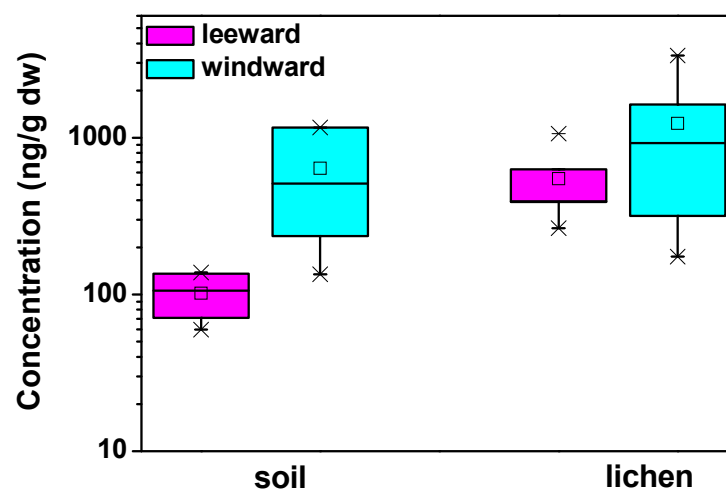


Figure 4. Comparison of PAH concentrations and altitudinal distribution between windward and leeward slopes of the mountain.

3.3.2. The Role Played by the Forest and Altitude

Principal component analysis combined with multiple linear regression analysis (PCA/MLRA) is widely used to quantify and apportion different sources of organic pollutants in the environment, such as source identification of PAHs in the urban atmosphere [44] and in remote Mt. Gongga [45]. In this study, PCA/MLRA was performed using the original data (latitude, longitude, altitude, TOC and Σ_{16} PAHs) of windward slope soil samples in sub-area 1a. Latitude was excluded during stepwise statistical analysis due to its collinearity with longitude and altitude. The first two principal components (PCs) with eigenvalues greater than 1 accounted for 90.6% of the total variance in the dataset (Table S3). PC1, explaining 61.9% of the total variance, was strongly dominated by altitude and Σ_{16} PAHs, indicating that altitude plays a key role in the deposition of PAHs [39,45]. PC2, explaining 28.7% of the total variance, was highly associated with TOC and Σ_{16} PAHs. As SVOCs are lipophilic, soil TOC largely regulates the partitioning of SVOCs between air and soil/vegetation [46–48], highlighting forests' role in transferring atmospheric PAHs to soil [49,50].

Temperature is a key factor regulating the global distribution of SVOCs. Certain POPs exhibit preferential deposition and accumulation in remote regions, including high latitudes and high elevations, with condensation favored in low-temperature environments [14]. This concentration magnification of POPs along altitudinal gradients in mountains has been termed “mountain cold-trapping” [39,51]. Given that temperature decreases with increasing altitude, PC1, strongly dominated by altitude and Σ_{16} PAHs, confirmed mountain cold-trapping was an important factor regulating PAH distribution in high-mountain forest soils [43,51,52].

Furthermore, POPs repeatedly undergo exchange processes between the atmosphere and the terrestrial surface during LRAT, profoundly shaping the fate and distribution of SVOCs on regional and global scales. As extensive vegetation covers, forests have a significant impact on the atmospheric concentration of SVOCs [14,53,54] and enhance air-to-soil fluxes [52] by absorbing SVOCs in the atmosphere and transferring them to soil via fallen leaves. This capacity of forests to filter airborne organic pollutants has been named the “forest filter effect” [49,50], which effectively attenuates the LRAT of SVOCs [55]. Forested soils accumulate pollutants primarily through direct deposition and subsequent accumulation, rather than enhanced thermodynamic partitioning driven by low temperatures, as shown by a study from Liu et al. (2014b) [45]. Considering the facts that fallen leaves carry SVOCs and increase soil organic matter, PC2, highly associated with

TOC and $\Sigma_{16}\text{PAHs}$, indicated the forest filter effect was another important factor regulating PAH distribution in high-mountain forest soils.

From the above analysis, the mountain cold-trapping effect and forest filter effect jointly regulated PAH distribution in high-mountain forest soils. The relative contribution of the mountain cold-trapping effect increased with altitude, exceeding 75% at higher-altitude sites (mean: 41.4%) (Figure 5), indicating its enhanced dominance with increasing elevation. At higher altitudes with low temperatures, the partition coefficients of SVOCs from the atmosphere to condensed phases increase significantly [5]. Additionally, low temperatures reduce microbial degradation rates and re-volatilization rates of SVOCs [12,43,52]. Furthermore, precipitation strongly influences scavenging of SVOCs from the atmosphere [12,53], and at higher altitudes, precipitation more frequently occurs as snow is a more efficient scavenger than rain [53,56–58]. Thus, lower temperatures and more efficient wet deposition (e.g., snow) facilitate enhanced PAH condensation and accumulation with increasing mountain altitude.

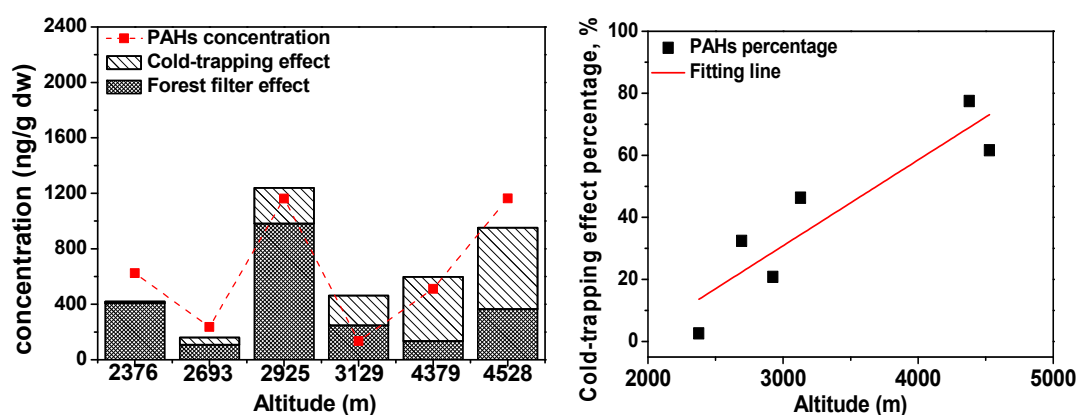


Figure 5. Patterns of PAHs in O-Horizon soils by principal component analysis/multiple linear regression (PCA/MLRA).

The forest filter effect accounted for an average proportion of 58.6% of the contribution to PAH distribution in this study area. Its relative contribution was positively correlated with TOC and, conversely, exhibited a negative linear correlation with altitude. This trend was consistently observed across sampling sites. Vegetation in the TP displays a distinct vertical zonation, with evergreen broad-leaved forest, mixed coniferous–broad-leaved forest, subalpine coniferous forest, and alpine shrub meadows distributed sequentially from low to high elevations [59]. Forest net primary productivity in the sampling region decreases with increasing altitude [60], resulting in greater leaf litter fall and subsequent accumulation in the soil of low-altitude ecosystems. In addition, SVOCs have been documented to deposit more heavily in deciduous forests than in coniferous forests [49]. Collectively, these observations confirm that the forest filter effect weakens progressively with rising altitude.

3.4. Source Assessment

Of the 16 priority PAHs, fluoranthene (FLT) and pyrene (PYR), with a molecular weight (MW) of 202, as well as benzo[ghi]perylene (BghiP) and indeno[1,2,3-cd]pyrene (IcdP), with an MW of 276, exhibit distinct emission profiles from different sources. For this reason, the isomer ratio method has been widely employed for the qualitative source apportionment of PAHs. According to the diagnostic ratio criteria, $\text{FLT}/(\text{FLT}+\text{PYR}) < 0.4$ indicates petroleum-derived sources, $0.4 < \text{FLT}/(\text{FLT}+\text{PYR}) < 0.5$ indicates petroleum combustion sources, and $\text{FLT}/(\text{FLT}+\text{PYR}) > 0.5$ indicates combustion of grass, wood, and coal; similarly, $\text{IcdP}/(\text{IcdP}+\text{BghiP}) < 0.2$ indicates petroleum-derived sources, $0.2 < \text{IcdP}/(\text{IcdP}+\text{BghiP}) < 0.5$ indicates petroleum combustion sources,

and $IcdP/(IcdP+BghiP) > 0.5$ indicates combustion of grass, wood, and coal [61]. Diagnostic ratio results (Figure 6) show that $FLT/(FLT+PYR)$ values in most soil and lichen samples exceeded 0.5, and $IcdP/(IcdP+BghiP)$ values in the majority of these samples were also above 0.5, collectively demonstrating that PAHs in these samples were predominantly derived from the combustion of grass, wood and coal. Notably, $FLT/(FLT+PYR)$ and $IcdP/(IcdP+BghiP)$ values differed distinctly between soil and lichen samples, suggesting disparities in PAH compositions between the two environmental matrices. However, in a very small number of samples, $FLT/(FLT+PYR)$ and $IcdP/(IcdP+BghiP)$ values were lower than 0.4 and 0.2, respectively, implying minor contributions from petroleum-derived and petroleum combustion sources.

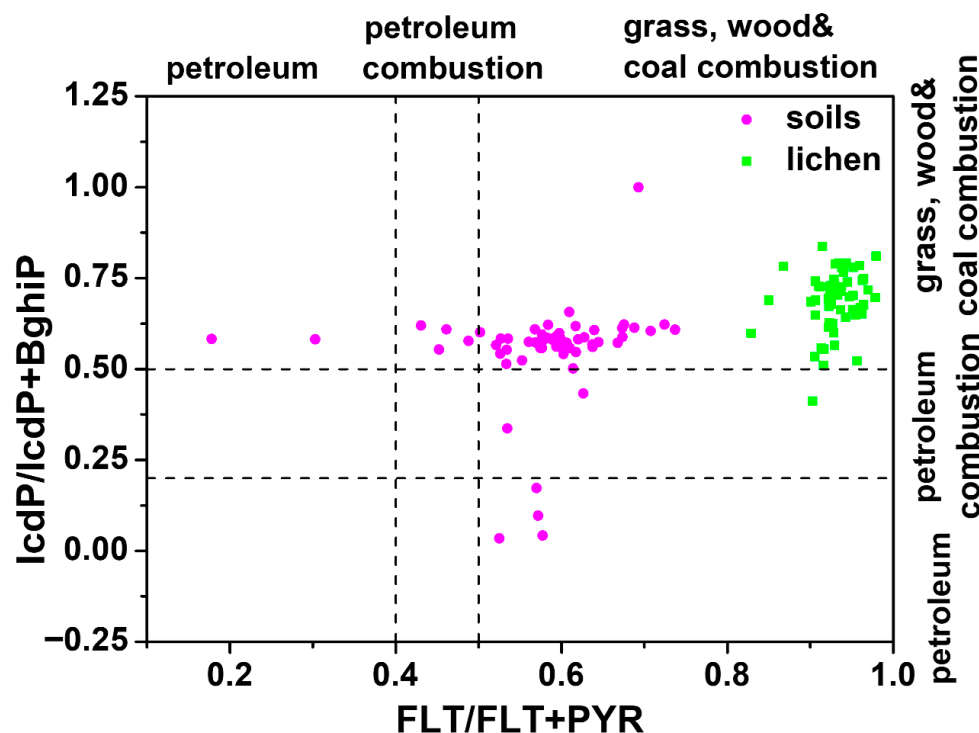


Figure 6. Bivariate plot for diagnostic ratios of $IcdP/(IcdP+BghiP)$ vs. $FLT/(FLT+PYR)$.

4. Conclusions

Studies on the transport and accumulation of SVOCs in the Hengduan Mts., located in the southeast part of the TP, remain very limited. In the present study, PAHs were analyzed in a total of 124 soil and lichen samples across the Hengduan Mts. The concentrations of $\Sigma_{16}PAH$ in soils and lichen ranged from 59.8 to 1163 ng/g and from 174 to 3362 ng/g, respectively. Notably, the PAH concentrations in both soils and lichens were higher than those reported in the northern and northwestern regions of the TP. The compositional fractionation of PAHs along with the latitude (and altitude) of the sampling sites indicated the distribution of PAHs is significantly influenced by LRAT, by both the Indian monsoon and the East Asian monsoon. Furthermore, the mountain cold-trapping effect and forest filtering effect jointly dominated the deposition and spatial distribution of PAHs in the forest areas of the Hengduan Mts., southeast TP.

Supplementary Materials: The following supporting information can be downloaded at: <https://www.mdpi.com/article/10.3390/f17040502/s1>, Figure S1. The distribution of PAHs in soil and lichen from south-to-north transect; Figure S2. The PCA results of PAHs in soils (A) and lichen (B); Figure S3. The air back trajectories of representative points in Summer (A), in Winter (B), and in later

Summer (C); Table S1. The detail information of sampling sites; Text S1: Detailed information of Sample Analysis.

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