



## Atmospheric transport and accumulation of organochlorine compounds on the southern slopes of the Himalayas, Nepal



Ping Gong<sup>a</sup>, Xiao-ping Wang<sup>a,\*</sup>, Sheng-hai Li<sup>a</sup>, Wu-sheng Yu<sup>a</sup>, Jiu-le Li<sup>a</sup>, Dambaru Ballab Kattel<sup>a,c</sup>, Wei-cai Wang<sup>a</sup>, Lochan Prasad Devkota<sup>b</sup>, Tan-dong Yao<sup>a</sup>, Daniel R. Joswiak<sup>a</sup>

<sup>a</sup> Key Laboratory of Tibetan Environmental Changes and Land Surface Process, Institute of Tibetan Plateau Research, Chinese Academy of Sciences, P.O. Box 2871, Beijing 100101, China

<sup>b</sup> Central Department of Hydrology and Meteorology, Tribhuvan University, Kathmandu 44618, Nepal

<sup>c</sup> Department of Meteorology, COMSATS Institute of Information Technology, Islamabad, Pakistan

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### ABSTRACT

Studies have been devoted to the transport and accumulation of persistent organic pollutants (POPs) in mountain environments. The Himalayas have the widest altitude gradient of any mountain range, but few studies examining the environmental behavior of POPs have been performed in the Himalayas. In this study, air, soil, and leaf samples were collected along a transect on the southern slope of the Himalayas, Nepal (altitude: 135–5100 m). Local emission occurred in the lowlands, and POPs were transported by uplift along the slope. During the atmospheric transport, the HCB proportion increased from the lowlands (20%) to high elevation (>50%), whereas the proportions of DDTs decreased. The largest residue of soil POPs appeared at an altitude of approximately 2500 m, and may be related to absorption by vegetation and precipitation. The net deposition tendencies at the air–soil surface indicated that the Himalayas may be a ‘sink’ for DDTs and PCBs.

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### 1. Introduction

Persistent organic pollutants (POPs), such as organochlorine pesticides (OCPs), hexachlorobenzene (HCB), and polychlorinated biphenyls (PCBs), are a class of organic compounds exhibiting high toxicity, environmental persistence, and a potential for bio-accumulation (Lallas, 2001). There are growing concerns regarding the potential for long-range atmospheric transport (LRAT) of POPs arising from detection of POPs in remote regions (Daly and Wania, 2005; Hung et al., 2010). Of particular interest in this regard are the transport and accumulation of POPs in mountain environments. Mountains have steep environmental gradients on a small spatial scale. Due to the sharp variations in temperature and precipitation, atmospheric POPs are prone to accumulate at high elevations (Daly and Wania, 2005). Mountains, therefore, have been considered ‘traps’ for LRAT of POPs (Vighi, 2006).

With respect to the accumulation in mountain environments, both the variable concentrations and the relative compositions of

POPs have been strongly correlated along altitudinal gradients. Many studies have addressed these patterns along the slopes of high mountains, such as the European Alps (Shen et al., 2009; van Drooge et al., 2004), Rocky Mountains (Daly et al., 2007a), Andes (Estellano et al., 2008; Tremolada et al., 2008), Mount Meru in Tanzania (Parolini et al., 2013), the Himalayas (Wang et al., 2006, 2009; Yang et al., 2008), and Balang Mountain in China (Chen et al., 2008; Liu et al., 2010). A positive correlation between the levels of POPs and altitude may relate to the temperature dependence of precipitation scavenging (Chen et al., 2008; Daly et al., 2007a; Liu et al., 2010; Tremolada et al., 2008). However, when the mountains are close to the emission sources, higher concentrations of POPs occur in the lowlands (Parolini et al., 2013). The relative compositions of POPs also vary with altitude. The more volatile POPs (e.g., HCB, HCHs) are prone to transport to high altitudes, whereas the less volatile POPs (e.g., DDTs) are more likely to accumulate in the lowlands (Chen et al., 2008; Wania and Westgate, 2008). Therefore, the relative compositions of more volatile POPs in air increase with increasing altitude, whereas those of less volatile POPs decrease with increasing altitude. In addition, mountain forests play a role in enhancing the POP deposition on mountains (Nizzetto et al., 2006; Wenzel et al., 2002), which is

\* Corresponding author.

E-mail address: [wangxp@itpcas.ac.cn](mailto:wangxp@itpcas.ac.cn) (X.-p. Wang).

reflected by the higher concentrations of POPs detected in soil under a canopy (Daly et al., 2007a).

The Himalayan range, with an altitude of <100–8844 m, is the highest mountain range in the world. The regions to the south of the Himalayas form the South Asian subcontinent, which has a high population density and high emissions of pollutants (Gupta, 2004; Ramanathan et al., 2005). Organochlorine pesticides are used in agriculture (Abhilash and Singh, 2009), and for the control of malaria and black vectors (Mittal et al., 2004). Additionally, PCBs may be emitted from electronic waste, ship breaking activities, and dumped solid waste in South Asia (Chakraborty et al., 2013). Many studies have reported that POP usage in South Asia is marked by increased concentrations of OCPs and PCBs in the air and soil (Chakraborty et al., 2010, 2013; Devi et al., 2011, 2013; Kumar et al., 2011, 2013; Kumari et al., 2008; Mishra et al., 2012; Syed et al., 2013; Zhang et al., 2008). The climate of South Asia is controlled by the Indian monsoon in the summer (Yao et al., 2012). The Indian monsoon undergoes an orographic uplift (Yao et al., 2012), and affects the transport of POPs to the high-elevation regions of the Tibetan Plateau (Sheng et al., 2013; Wang et al., 2010). For example, OCPs and PCBs can be detected in the high-altitude regions of the Himalayas (Guzzella et al., 2011; Wang et al., 2006). However, there is still no systematic method for observing the transport and accumulation of POPs in Nepal.

This study investigates the spatial distribution of POPs in the air, leaves, and soil along a transect on a southern slope of the Himalayas in Nepal with an altitude of 135–5100 m. The objective of this study was to investigate the atmospheric transport characteristics of POPs from the lowlands of Nepal to the heights of the Himalayas, and to discuss the accumulation of POPs in the soil and the vegetation of the Himalayas.

## 2. Materials and methods

### 2.1. Site description and sample collection

The observed north–south transect spanned the whole of Nepal from the region near the India–Nepal border (altitude: 135 m, Fig. S1 in the supplementary material) to the terminal point of a glacier in the high Himalayas (Nepal–China border, altitude: 5100 m, Fig. S1). The ecosystem and morphology varied with the wide altitude range, and the steep slope of the Himalayas. The timber line was at approximately 3000 m (Negi, 1994). The sampling sites included Simara (an agricultural region), Kathmandu (an urban region), Syabru Besi (a forest region), Richhe (a forest region), Kyanjin Gumba (a grassland), and Yala Peak (a glacier region). Detailed information about the sampling sites is listed in Table S1.

A passive air sampler (PAS) based on XAD-2 resin was used to collect air-phase POPs in this study. The PAS units were deployed at six sites, which are listed in Table S1 in detail. Prior to deployment, the XAD resin was Soxhlet extracted using the solvents methanol, acetonitrile, and dichloromethane (DCM). The XAD resin (60 mL of wet XAD in methanol) was transferred to a pre-cleaned stainless steel mesh cylinder and dried in a clean desiccator (Wania et al., 2003). Dry cylinders were sealed in airtight stainless steel tubes with Teflon lids. Three blank resin columns were carried to the sites, which served as the field blanks. The air samples were collected from May to November in 2012 (Table S1). The harvested XAD cylinders were stored in sealed and solvent rinsed glass jars, and sent to the laboratory of the Institute of Tibetan Plateau Research in Beijing. The samplers were stored at  $-20\text{ }^{\circ}\text{C}$  until extraction.

The soil and leaf samples were collected at the same time that the PAS units were deployed (Table S2). In the regions around the PAS sites within a radius of 200 m, five surface soil samples (0–5 cm) were conducted randomly with a cleaned stainless-steel hand trowel, and the samples were mixed as one sample. Similar to the soil sampling, mixed leaf samples were collected near the PAS sites. The dominant tree species were determined at each site, and the samples were taken at heights  $\sim 2$  m above the ground. The soil and the leaf samples were then wrapped in clean aluminum foil and stored in plastic bags, and immediately mailed to the laboratory. Prior to analysis, the soil samples were freeze-dried and manually ground. The soil samples were then sieved through a 0.2 mm mesh to remove large pieces of debris, roots and other rubble.

### 2.2. Sample processing and analysis

The XAD, soil, and leaf samples were extracted and cleaned as described by Wang et al. (2010, 2012). A description of these methods is given in Text S1. All samples and blanks were analyzed on a gas chromatograph (GC) with an ion-trap

mass spectrometer (MS) (Finnigan Trace GC/PolarisQ). The details on the GC/MS analysis are described in Text S2. All samples were analyzed for the following target compounds: PCB 28, 52, 101, 138, 153 and 180; HCB; *o,p'*-DDE, *p,p'*-DDE, *o,p'*-DDT, and *p,p'*-DDT.

The carbon contents of the soil (total carbon, TC, and total organic carbon, TOC) were measured using a TOC analyzer (Shimadzu 5000-A, Japan). The lipid content of the leaves was measured gravimetrically for each sample (Schrlau et al., 2011).

### 2.3. Quality assurance/quality control (QA/QC)

All analytical procedures were monitored using strict QA/QC measures. Procedural blanks and field blanks were extracted and analyzed in the same way as the samples. For the air sample, the trace target compounds were detected in the field blanks (Table S3). The method detection limits (MDLs) were derived as three times the standard deviation of the field blanks, and the MDLs were 0.01–1.71 ng/sample (Table S3). The recoveries were between 74% and 101% for PCB-30, and between 63% and 78% for Mirex.

For the soil and the leaf samples, procedural blanks (extraction of a thimble filled with  $\text{Na}_2\text{SO}_4$ ) were treated in exactly the same manner as the samples. The MDLs were derived as three times the standard deviation of the blanks. The MDLs based on a 25 g soil sample and 5 g leaf sample were 0.01–7.44 pg/g and 0.05–37.2 pg/g, respectively (Table S4). Recoveries were between 55% and 89% for PCB-30 and 53–99% for Mirex.

All of the reported values were field-blank-corrected (mean blank concentrations were subtracted) but not corrected for the recovery rates.

### 2.4. Statistical method

The hierarchical clustering analysis (HCA) method was used to classify the sampling sites into groups based on the variable relative compositions of POPs in sites (Gong et al., 2013; Liu and Wania, 2014), which were then used to discuss the sources of the atmospheric POPs in Nepal in this study. The relative compositions were defined as the ratio of an individual component concentration to the total concentration of POPs at each site. Then, this ratio was chosen as a variable when conducting the HCA analysis (SPSS 16.0). Ward's method and the Euclidean distance were applied.

## 3. Results and discussion

### 3.1. Levels of organochlorine compounds in air

The POPs amounts in PAS (ng/sample or pg/sample/day) were often reported (Liu et al., 2009; Zhang et al., 2008) if the sampling rates of PAS ( $R$ ) fluctuated slightly. However, in this study, the temperature varied from  $-6.4$  to  $28.4\text{ }^{\circ}\text{C}$ , and the air pressure from 629 to 1145 hPa (Table S5). The variable meteorological parameters caused  $R$  to fluctuate by more than two times (Table S5). Therefore, the atmospheric concentrations in this study were calibrated using the  $R$  values. The details about  $R$  are discussed in Text S3. By using the obtained  $R$  values, the volumetric concentrations of atmospheric POPs were estimated (Table 1). The volumetric air concentrations ( $\text{pg}/\text{m}^3$ ) were compared with the data from active samplers (Table S6), and the amounts in PAS ( $\text{pg}/\text{sample}/\text{day}$ ) were also reported for comparing with the other data based on XAD-PAS (Table S7).

The levels of HCB were 128–416  $\text{pg}/\text{m}^3$  (Table 1), which is one order of magnitude higher than those in remote regions, such as the Bay of Bengal and the Indian Ocean (Gioia et al., 2012) (Table S6). The concentrations of the total DDTs (DDE + DDD + DDT) were 71.1–1613.7  $\text{pg}/\text{m}^3$ . The average concentrations of *o,p'*-DDT, *p,p'*-DDT, and *p,p'*-DDE were 159.7, 125.5, and 154.8  $\text{pg}/\text{m}^3$ . The compounds *o,p'*-DDT, *p,p'*-DDT, and *p,p'*-DDE accounted for  $39.7 \pm 6.7\%$ ,  $26.8 \pm 3.0\%$ , and  $31.3 \pm 6.2\%$  of the DDTs. The concentrations of DDTs in Nepal were compared to those in urban India (Zhang et al., 2008, Table S6). The average concentration of  $\sum_6\text{PCB}$  was  $26.9 \pm 25.6\text{ pg}/\text{m}^3$ , which is less than those reported for urban India (Zhang et al., 2008, Table S6).

The concentrations of atmospheric POPs ( $\text{pg}/\text{sample}/\text{day}$ , Table S7) in Nepal were also compared with those in the Asian cities where the XAD-PAS were deployed. It was shown that the levels of HCB, DDTs, and PCBs in Nepal were higher than or of the same order of magnitude as those in the Asian cities (Baek et al., 2013; Nasir

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