Atmospheric organochlorine pesticides and polychlorinated biphenyls in urban areas of Nepal: spatial variation, sources, temporal trends and long range transport potential

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Abstract

The study of persistent organic pollutants (POPs) in low latitude tropical and subtropical urban cities is necessary to assess their local and global impacts on ecosystems and human health. Despite studies on levels of POPs in water, soils, and sediments, analysis of the distribution patterns, seasonality and sources of POPs in urban regions of Nepal remain limited. Polyurethane foam (PUF)-based passive air samplers were deployed in three major cities in Nepal: Kathmandu (the capital city), Pokhara and Hetauda (agricultural cities). Dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) were the dominant organochlorine pesticides in the atmosphere at all sites. The average concentrations of POPs were: ∑DDTs, $8.7–1.0\times10^3$ pg/m^3; ∑HCHs, $5.3–3.3\times10^3$ pg/m^3; HCB, $5.8–3.4\times10^2$ pg/m^3; ∑endosulfan, BDL–51 pg/m^3; and $\sum_6$PCBs, 1.4–47 pg/m^3. Isomer and metabolite ratio analyses suggested that the concentrations present were from both new and historical applications of the POPs. Vegetable production sites and their market places appeared to be the major DDT and HCH source areas. Higher atmospheric concentrations of DDT and HCH occurred during the pre-monsoon and monsoon seasons, and winter, respectively, closely associated with their local application for soil preparation and vegetable spraying. The estimated travel distances of the POPs (HCB, α-HCH, γ-HCH, and p,p′-DDT) under the Nepalese tropical climate were all above 1000 km, suggesting that high precipitation levels in the tropical climate were not enough to scavenge the POPs and that Nepal could be an important source region for POPs. Due to their close proximity and cold trapping (driven by low temperatures), the high Himalayas and the Tibetan Plateau are likely the key receptors of POPs emitted in Nepal. These results add to the information available on POPs from tropical developing countries.

Introduction

Persistent Organic pollutants (POPs), including agrochemicals such as organochlorine pesticides (OCPs) and industrial chemicals such as polychlorinated biphenyls (PCBs), are semi-volatile, persistent, bioaccumulative and toxic in nature (Jones and de Voogt, 1999; Zhang et al., 2016). POPs are ubiquitous worldwide, although they are primarily located in densely populated, subtropical and tropical regions (Simonich and Hites, 1995; Wang et al., 2016b). With favorable atmospheric circulation, POPs can be transported to places where they have never been used or produced (Hageman et al., 2015). Due to long range atmospheric transport (LRAT) and global condensation, POPs emitted in tropical and subtropical regions can disperse to polar and high mountain areas, where temperatures are cold enough to trap POPs (Mackay and Wania, 1995). In this regard, the emissions of POPs from low latitude countries (tropical and subtropical environments) have received global concern.

The climate of South Asia is dominated by tropical and subtropical monsoons. South Asia, which includes developing countries such as Nepal, India and Pakistan, is regarded as a POP source region (Sharma et al., 2014). Among these countries, India is the biggest consumer and producer of OCPs and PCBs; it has a long history of extensive use of OCPs in agriculture and vector control, and PCBs in the industrial sector (Sharma et al., 2014; Chakraborty et al., 2013). In agricultural regions of India, atmospheric OCP concentrations can reach 4000 pg/m^3, which is among the highest values reported in the literature (Pozo et al., 2011). POP emissions in Pakistan are also serious, with historical dichlorodiphenyltrichloroethane (DDT) sources and current applications of lindane and endosulfan (Syed et al., 2013; Nasir et al., 2014). All these sources contribute to the POP emissions in the subtropical and tropical region and are influential at the regional and global scales, due to the LRAT of POPs.

In comparison with studies from India and Pakistan, where relatively comprehensive sampling of POPs in air, soil, water, sediment and vegetation have been conducted (Sharma et al., 2014; Tariq et al., 2007; Eqani et al., 2012), POP research in Nepal remains limited. Nepal is a land-locked, subtropical, Himalayan country surrounded by China, to the north, and India. The lowland plain area (<100 m above sea level) in the south is relatively warm and densely populated, while the high Himalayan mountains (> 4000 m above sea level) in the north comprise the least populated area. Nepal is an agrarian country; 39% of its gross domestic product is based on agriculture. The

use of agrochemicals to protect crops and vegetables is common practice and most of the agrochemicals in Nepal are imported from India. Nepal has an open border with India that allows easy import (through both legal and illegal routes) of agrochemicals. Although Nepal banned most OCPs in 2001, the indiscriminate use of these chemicals in farmlands is common (Yadav et al., 2017). To date, although some studies have reported the concentrations of POPs in water, soils and sediments from Nepal (Aichner et al., 2007; Yadav et al., 2016), detailed analyses regarding the sources of POPs in the urban (populated) regions of Nepal are lacking.

Most recently, Yadav et al. (2017) conducted a short-term (8-week) passive air sampling study in urban areas of Nepal and suggested that new applications and illegal use of DDTs and lindane still occur. Due to the close proximity of Nepal to the Himalayas, several studies have demonstrated that POPs emitted from the low altitude source regions of Nepal might be transported to the higher Himalayan mountains (Guzzella et al., 2016; Gong et al., 2014; Sheng et al., 2013; Pokhrel et al., 2016). However, these studies have not illustrated the seasonality of POPs in the atmosphere of the Nepal source (urban) regions. In addition, the spatial distribution of POPs across different types of land cover (i.e., croplands, vegetable produce areas, industrial areas, tourist areas and residential areas, among others) within the cities is unclear; such information would be useful for identifying the source features and assessing the "hot spots" of personal exposure and health risks from air pollution. There has been notable absence of a systematic monitoring campaign for POPs in the urban areas of Nepal and this country has not been considered for any global or regional regular monitoring plan except for a year monitoring of atmospheric OCPs in a background region (Shunthirasingham et al., 2010). Thus, there is a need to obtain atmospheric concentrations of POPs, and understand the spatial variations and seasonal trends of the contaminants in Nepal, to highlight the potential contribution of the region to the POPs burden of the Himalayan mountains and the global environment.

In the present study, polyurethane foam (PUF)-based passive air samplers were deployed in three major cities of Nepal (Kathmandu, Pokhara and Hetauda). The objectives of the study were to: (i) obtain the levels of OCPs and PCBs in the urban regions; (ii) compare these levels with other cities in South Asia to identify the levels of pollution in Nepal in the regional and global context; (iii) elucidate the spatial distribution and sources of POPs in each city; and (iv) identify any seasonal

trends in the POP concentrations and predict the LRAT potential of POPs from the Nepalese tropical climatic region.

Materials and Methods

Study area and sampling design. Three major cities, Kathmandu (the capital city), Pokhara and Hetauda (agricultural cities) were chosen for this study. Descriptions of the cities, their climates and air circulation patterns are provided in the supporting information (SI; Text SI-1, Figure SI-1 and Figure SI-2).

Six sample sites (K1-K6) in Kathmandu, four (P1- P4) in Pokhara and three in (H1-H3) Hetauda were selected for air monitoring; the air was monitored for one year at each site (Kathmandu, August 2014–August 2015; Pokhara, August 2014–August 2015; and Hetauda November 2015–November 2016), covering three seasons (pre-monsoon, February to May; monsoon, June to September; and winter, October to January). Duplicate PUF-based passive air samplers were deployed at each site and changed every two months during each one-year sample period. Efforts were made to cover each type of land use present, including residential, industrial, croplands, vegetable production areas and tourist sites. Details of the sampling sites and sampling periods are presented in the SI (Table SI-1 and Table SI-2).

Sampler deployment, extraction and analysis. Prior to deployment, PUF disks were cleaned with dichloromethane using soxhlet extraction (24 h) and dried in a desiccator under reduced pressure for 24 h. The cleaned PUF disks were spiked with four depuration compounds (DCs; PCB-30, PCB-54, PCB-104, and PCB-188), to enable analysis of the site-specific sampling rates (Pozo et al., 2009). Each PUF disk was wrapped with clean aluminum foil, packed in a zip-lock plastic bag, and stored in an airtight tin container to protect it from contamination during transport. The samplers were mounted on the roof of a building at each sample site that was at least 6 m above the ground. During sampling, five field blanks for Kathmandu, three for Pokhara and three for Hetauda were prepared to assess possible contamination.

After collection, samples were transported to the Key Laboratory of Tibetan Environment Changes and Land Surface Processes, Beijing, China and stored at −20°C until extraction. The samples were soxhlet extracted with dichloromethane for 24 h after being spiked 20 µl (100 pg/µl) of recovery standard (2,4,5,6-tetrachloro-m-xylene). Details on the chemical extraction are included

in Text SI-2. Samples were analyzed using a gas chromatograph (GC) with an ion-trap mass spectrometer (MS) (Finnegan Trace GC/PolarisQ), using a CP-Sil 8CB capillary column (50 m, 0.25 mm, 0.25 mm) and operating under the MS–MS mode. Details on the gas chromatographic temperatures are given in Text SI-3. The target analytes were: PCB-28; PCB-52, PCB-101, PCB-138, PCB-153 and PCB-180; α-hexachlorocyclohexane (α-HCH), β-HCH, γ-HCH and δ-HCH; hexachlorobenzene (HCB); o,p′-dichlorodiphenyldichloroethylene (o,p′-DDE) and p,p′-DDE; o,p′-dichlorodiphenyldichloroethane (o,p′-DDD) and p,p′-DDD; o,p′-DDT and p,p′-DDT; α- and β-endosulfan; and heptachlor and heptachlor epoxide.

Quality control. All analytical procedures were monitored using strict quality assurance and control measures. Both laboratory and field blanks were extracted and analyzed exactly the same way as the samples. Method detection limits (MDLs) were derived as mean blank concentrations plus 3 times the standard deviation. When a target compound was not detected in the blanks, the concentration of the lowest calibration standard was substituted for the MDL. The derived MDLs ranged between 0.01 and 1.34 ng per sample for OCPs, and between 0.02 and 0.13 ng per sample for PCBs (Table SI-3). The recoveries of all samples were within 89 ± 14% (between 75% to 103%) for 2,4,5,6-tetrachloro-m-xylene. All reported values were blank corrected, but not corrected for the recovery.

Sampling rate determination. To convert derived concentration per sampler into volumetric concentration, site-specific sampling rates for each sampling site were determined as described by Moeckel et al. (2009). The average sampling rates for each city were 4.3 ± 0.64 m^3/day for Kathmandu, 4.3 ± 0.82 m^3/day for Pokhara and 3.3 ± 0.83 m^3/day for Hetauda; details of the calculation of the site-specific sampling rates are included in Text SI-4 and Table SI-4.

Calculation of air concentrations. In order to obtain the volumetric air concentrations, compound specific effective air volume were calculated using the formula given by Harner et al., (2013)

$$V_{Air} = (K'_{PSM-A}) + (V_{PSM}) \times \left\{1 - \exp\left[\frac{k_A}{K'_{PSM-A}} \times \frac{1}{D_{film}}\right] t\right\} \quad \ldots\ldots\ldots (1)$$

Where,

V_{Air} = effective air sample volume (m^3)

$K'_{PSM-A} = K_{PSM-A} \times \delta_{PSM}$

Shoeib and Harner, 2002 have used a relationship between K_{PSM-A} and K_{OA} as

$$logK_{PSM-A} = 0.6366logK_{OA} - 3.1774 \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (2)$$

δ_{PSM} = density of PUF (g/m^3)

V_{PSM} = Vol. of the passive air sampler (m^3)

k_A = air side mass transfer coefficient (m d^{-1})

D_{film} = effective film thickness (m)

t = time (day)

k_A = the sampling rate R (m^3 d^{-1}) divided by the surface area of the PUF disk (341cm^2)

The air concentrations of compounds were obtained by dividing the concentration (pg/sample) by effective air sample volume (V_{Air}).

Result and Discussion

Atmospheric levels of POPs. Due to the high POP concentrations in the atmosphere and the hot climate, South Asia countries (e.g., India) are generally regarded as source regions of POPs, from the global perspective (Nasir et al., 2014; Shunthirasingham et al., 2010; Chakraborty et al., 2010;Zhang et al., 2008). Concentrations of OCPs and PCBs at each sample site in the major Nepalese cities are provided in Tables SI 5a–c and SI 6a–c. Table 1 compares the OCP and PCB concentrations found in the current study with those in other cities in South Asia. Generally, atmospheric levels of HCHs and DDTs in Nepal were lower than in Indian cities (Chakraborty et al., 2010; Zhang et al., 2008), comparable with levels found recently in Bangladesh (Nost et al., 2015) and Vietnam (Wang et al., 2016a), but slightly higher than those reported in Pakistan (Nasir et al., 2014) and previous study in Nepal (particularly for DDT in Kathmandu and Pokhara) (Yadav et al., 2017) (Table 1 and Table SI-7). Given that previous studies in South Asia have shown that POPs are still used in agriculture, including cotton and rice paddy fields (Nasir et al., 2014; Chakraborty et al., 2010; Zhang et al., 2008), and public health (Li et al., 1998), the relatively high concentrations of HCHs and DDTs in Nepal suggest their probable ongoing application locally, which is a cause for concern.

The atmospheric concentrations of endosulfan in the major Nepalese cities were 2–3 times lower than those reported for Pakistan and Vietnam, but one-magnitude lower than those found in India

(Table 1), implying relatively low levels of usage of this POP in Nepal. Regarding HCB and PCBs, their concentrations were broadly lower than in other cities in South Asia (Table 1). Taken together, levels of POPs (OCPs + PCBs) in the different countries roughly followed the order: Nepal $\approx$ Pakistan < India. The data from this study were further compared with results of the Global Passive Air Sampling (GPAS) study, which used the same PUF-passive air samplers (Table SI-7). Overall, the levels of POPs in Nepal, especially DDTs and HCHs, were higher than those found in other tropical regions, including the Philippines, Africa and Mexico (Table SI-7). Therefore, Nepal could be considered a regional and/or global source of DDTs and HCHs.

Box and whisker plots allow an analysis of the characteristics of a dataset, including the central tendency, dispersion, asymmetry, and extremes. Figure 2 shows the average value and data dispersion of each class of POPs in Kathmandu, Pokhara, and Hetauda. Extremely high concentrations of HCHs (∑HCHs, 3.3×10^3 pg/m^3) were found in Hetauda, close to the highest concentration reported in Indian agricultural regions (4.0×10^3 pg/m^3) (Pozo et al., 2011). Given Hetauda is close to India, this similarity suggests the potential agricultural application of HCHs in Hetauda. In addition, large data dispersion of DDT, HCH and HCB levels were found (Figure 2); large high/low concentration ratios (Table SI-8) were therefore observed (∑DDT = 117, ∑HCH = 630, and HCB = 60), which means that among these three cities, the chemical concentrations varied by 1–2 magnitudes. Both Figure 2 and Table SI-9 indicate that the emission features of POPs in the three cities may be different (intercity differences). Confounding factors, such as the sources of the pollutants (i.e., new application or historical residues), different land uses and different seasonal applications have a combined effect on the concentrations of POPs, leading to the large data variation observed in Figure 2.

Intercity comparisons and compositional trends

DDTs. Similar to Figure 2, Figure SI-3 displays the data dispersion and average levels of DDT isomers in each city. *p, p′*-DDT, *p, p′*-DDE and *o, p′*-DDT were the dominant congeners, while *o, p′*-DDD was found at the lowest levels in all three cities (Figure SI-3). Large data dispersion of *p, p′*-DDT was found in Pokhara, with an outlier concentration of 6.2×10^2 pg/m^3 (Figure SI-3). The average concentrations of *p, p′*-DDT (small square in the box of Figure SI-3) followed the order: Pokhara > Kathmandu > Hetauda. The data highlight the difference between Pokhara and the other two cities, due to the high concentrations of *p, p′*-DDT. The ratio of *p, p′*-DDT/*p, p′*-DDE

can be used to differentiate new applications (>1) from past usage (<1) of technical DDT. The calculated *p, p′*-DDT/*p, p′*-DDE ratios were 1.06 ± 0.4 for Pokhara, 1.05 ± 0.6 for Kathmandu, and 2.2 ± 1.4 for Hetauda (Figure SI-4); this suggests recent application of p,p′-DDT occurred in Hetauda, while both recent applications and historical residues contributed to the atmospheric DDT levels in Pokhara and Kathmandu. The isomer ratios of o,p′-DDT/p,p′-DDT ranged between < 0.1 and 1.7 in all three cities (Figure SI-4), indicating the use of technical DDT, rather than dicofol-type DDT in Nepal. This is consistent with a recent study in Pakistan (Nasir et al., 2014), but contrary to results from India (Chakraborty et al., 2010; Zhang et al., 2008). In Nepal, the use of DDT started in 1956 to control malaria and it was extensively used in agriculture as a pesticide until its ban in 2001 (http://www.prmd.gov.np). Due to some illegal use and because Pokhara and Hetauda are both agricultural cities, the concentrations of DDTs in Pokhara and Hetauda are suggestive of extensive past or ongoing usage of technical DDT for agriculture, as well as an inability to enforce regulations to restrict DDT use and disposal.

HCHs. Figure SI-5 shows the data dispersion and average levels of HCH isomers in each city. γ-HCH was the dominant HCH detected. Given that technical grade HCH is dominated by the α-isomer, while lindane is almost completely γ-HCH (99%), the dominance of γ-HCH demonstrates the application of lindane in Nepal. The highest level of γ-HCH (3.3×10^3 pg/m^3) was found in Hetauda (Figure SI-5). Average values of γ-HCH and total levels of HCH isomers followed the order: Hetauda > Kathmandu > Pokhara. Hetauda is therefore distinct among the three cities for its high atmospheric γ-HCH concentrations. In South Asia, India still produces and exports HCHs as pesticides for agriculture and as pharmaceutical products, to control scabies and head lice (Chakraborty et al., 2010;Pozo et al., 2017). In Nepal, lindane (Gammaxene) was the first POP pesticide imported in 1952 to control malaria (Sharma DR, 2012) and its use was banned in 2001 (http://www.prmd.gov.np), following the ban of DDT. However, current observations demonstrate that the use of lindane is still common in Nepal; illegal trade between Hetauda and India is easy (due to the open border between Nepal and India), which may have enabled high application rates of lindane in Hetauda.

Endosulfan. The β-isomer of endosulfan was dominant (Figure SI-5); levels of total endosulfan (α- and β-isomers) in Hetauda (average = 14 pg/m^3) were highest, followed by Kathmandu (average = 10 pg/m^3) and Pokhara (average = 7.0 pg/m^3) (Figure SI-5). Technical endosulfan is a

mixture of 70% α-isomer and 30% β-isomer with an α/β ratio of 2.3 (Chakraborty et al., 2010). Therefore, an α/β ratio of near 2.3 implies recent application; in the current study, the α/β ratios were generally below 2.3 (Kathmandu = 0.27 ± 0.32, Pokhara = 0.42 ± 0.51, and Hetauda = 0.17 ± 0.05; Figure SI-4), indicating historical emissions of endosulfan as being the main source. In a recent study in the same region, Yadav et al. (2017) also found low α/β ratios (<2) in several samples. Similarly, low α/β ratios were observed in Mumbai, India (Chakraborty et al., 2010). Considering these results together, the low α/β ratios found in our study emphasize the higher persistence of the β-isomer, and that the historical application of endosulfan has left a residue that still remains in the environment.

PCBs. The six indicator PCBs measured (PCB-28, PCB-52, PCB-101, PCB-138, PCB-153, and PCB-180) are the most abundant congeners globally. PCB-28 and PCB-52 were dominant in the samples analyzed (Figure SI-6); Kathmandu, with a relatively large industrial manufacturing area, was characterized by high PCB concentrations (Figure SI-6). PCBs were widely used in transformers as a cooling liquid, in capacitors as dielectric fluids, and in other industrial applications (Vallack et al., 1998). Given that old transformers with PCB contaminated oil, which were manufactured before 1990, are still used in Nepal (Kattel and Devkota, 2014; K.C. and Devkota, 2015), the PCB-contaminated industrial waste is the likely source of PCBs in Kathmandu (Li et al., 2013).

HCB. Similar to PCBs, the highest atmospheric HCB concentrations (Figure 2) occurred in Kathmandu, while levels of HCB in Pokhara and Hetauda were almost the same (average around 18 pg/m^3). The HCB data also showed large dispersion with many outliers (Figure 2) in Kathmandu, implying diverse local emissions. This is similar to results observed by Syed et al. in Pakistan (Syed et al., 2013). HCBs are the most persistent and ubiquitous chemicals in the global environment (Bailey, 2001; Barber et al., 2005). They are released into the environment either as byproducts of chlorinated pesticides or through waste incineration (Bailey, 2001; Breivik et al., 2004; Liu et al., 2009). Thus, either emissions as byproducts of combustion or the use of fungicides containing HCB are possible sources of the HCB in Kathmandu.

Heptachlor. Among the target analytes, heptachlor was detected at the lowest concentrations. Chlordane (which contains 10% heptachlor in its technical mixture (Pozo et al., 2011)) usage and higher atmospheric levels of chlordane were reported by Yadav et al. (2017) suggesting the recent

use of technical chlordane in Nepal. Consequently, the detection of heptachlor in this study might be a result of chlordane usage. For confirmation, further analyses including chlordane should be carried out.

With regard to the concentrations of each analyte, Kathmandu and Pokhara showed similar trends (DDTs > HCHs > HCB > endosulfan > PCBs > heptachlor), while, the trend for Hetauda was a little different (HCHs > DDTs > endosulfan > HCB > PCBs > heptachlor). The dominancy of the agrochemical POPs (OCPs) over the industrial POPs (PCBs), indicates that the Nepali cities are still mainly influenced by agricultural pollutants (pesticides). According to a report by the government of Nepal, 90% of pesticides in Nepal are used for vegetable production (http://www.prmd.gov.np), suggesting that the vegetable production areas might be important source sites.

Intracity comparisons

The PUF-passive air samplers were deployed in almost all of the dominant land uses in the three cities; therefore, they can convey the emission souces and the extent of POPs emissions from the different land uses. The concentrations of individual POPs measured at different sites in Kathamandu, Pokhara and Hetauda are listed in Tables SI-5a–c and SI-6a–c. One-way analysis of variance (ANOVA) was performed to determine the statistical significance of differences in the mean values of individual chemicals among the sampling sites (Table SI-9). The ANOVA showed significant variation in the atmospheric levels of p,p′-DDT, o,p′-DDE, p,p′-DDE, γ-HCH, HCB and PCBs among the sites in Kathmandu, and significant variation for o,p′-DDE, p,p′-DDE, γ-HCH, and PCBs (Table SI-9) in Hetauda. There was no significant spatial variation for any analyte in Pokhara. Where significant differences were found, Tukey's post hoc tests were used to determine which sites differed significantly from the others (Tables SI-10 and SI-11).

Kathmandu. Six different land-use types were considered in Kathmandu (K1, cropland; K2, vegetable market; K3, industrial site; K4, tourist site; K5, residential site; and K6, a mix of agricultural and residential). Based on the Tukey's post hoc tests (Table SI-10), samples from K2 (a vegetable market) and K3 (an industrial site) in Kathmandu differed from the other sites. K2 was located at the major market and the biggest vegetable wholesale trade centre in Kathmandu; it was distinguished by high atmospheric concentrations of p,p'-DDT, p,p'-DDE and γ-HCH

313 (Table SI-10 and Figure 3). Given that the use of pesticides, even for vegetables ready for market,
314 is common practice in Nepal, higher concentrations of DDTs and γ-HCH at the market site can be
315 attributed to the usage of these chemicals for vegetable storage. Contrary to K2, K3 (an industrial
316 site) was different from the other sites due to the high HCB concentrations (Figure 3 and Table SI-
317 10). Among the three cities, only Kathmandu showed high HCB concentrations (Figure 2), while
318 within Kathmandu, the HCB levels at the industrial site were 3–4 times higher than at the other
319 sites. In addition to agricultural uses of HCB (as a fungicide), waste, coal, fuel, and biomass
320 combustion can also produce HCB (Bailey, 2001; Breivik et al., 2004; Liu et al., 2009). The
321 occurrence of high HCB levels at the industrial site (K3) may therefore be attributed mainly to the
322 fuel combustion processs used for generation of power to run the industries.

323 **Pokhara.** The ANOVA showed no significant variation in atmospheric concentrations of POPs
324 among the different sites in Pokhara (Table SI-9), indicating a homogeneous distribution of POPs
325 within the city. Despite this spatial distribution, P1 (cropland), and P2 (vegetable production and
326 market area) exhibited higher concentrations of DDTs (Figure SI-7). This re-emphasizes the broad
327 use of DDTs for agricultural purposes in Nepal.

328 **Hetauda.** Like Pokhara, Hetauda is an agricultural city. Table SI-11 shows that site H2 (a
329 vegetable growing area) was significantly different from H1 (farmland for crops) and H3 (an
330 industrial area) (Figure SI-8). High levels of γ-HCH made H2 significantly different from the other
331 sites. Again, local application of lindane in H2 may have resulted in the remarkably high γ-HCH
332 (3.3×10^3 pg/m^3) atmospheric concentrations.

333 Taking all the results into account, pesticide use in vegetable markets and vegetable production
334 areas is an obvious source of pollutants in Nepal; high levels were observed in these areas across
335 all three cities. Compared with the traditional belief that DDT and HCH are mainly used for crop
336 production, this study highlights the application of these chemicals for both vegetable growth and
337 storage (in markets). Thus, the health risks of pesticide-contaminated vegetables in Nepal should
338 be of great concern. With regard to tourism sites and regular residence sites, POP levels were
339 generally low, suggesting minor contamination. For industrial sites, DDT and HCH levels were
340 relatively low, while HCB levels were high in Kathmandu, implying pollution caused by fuel and
341 biomass combustion should also be a focus of concern.

Seasonality of OCPs

The above results indicate that the agricultural uses of DDTs and HCHs, and industrial combustion emissions of HCB, are the major emission sources of these chemicals in Nepal. Agricultural practices are generally seasonal; crops (rice paddy and maize) are sown at the beginning of the monsoon and harvested at the end of the monsoon in Nepal. During the same period (pre-monsoon and monsoon season), small areas of mixed planting (crops and vegetables together) and domestic vegetable planting in home gardens are common. After the harvest of crops (November), vegetables are largely planted in farmlands and produced mainly for commercial purposes. This seasonal application inspired us to test whether the atmospheric POPs in Nepal displayed seasonality. For clarity, only seasonality of the dominant chemicals (DDTs, HCHs and HCB) was considered.

DDT. Considering that high concentrations of DDTs were generally observed in vegetable production areas and markets, the seasonality of DDT congeners (p,p′-DDT, o,p′-DDT, p,p′-DDE and o,p′-DDE) at K2, P2 and H2 were analyzed (Figure 4), which showed that there were higher levels of atmospheric DDTs during the pre-monsoon and monsoon seasons at the sites. In some winter samples, the concentrations of p,p′-DDT and o,p′-DDT were around tens of pg/m^3, whereas their concentrations reached up to hundreds of pg/m^3 during the monsoon period (K2 and P2 in Figure 4). Similar trends were also observed for other sites (i.e., croplands; K1, P1 and H1; Figures SI-9–11). We expected low atmospheric DDT concentrations during the monsoon season, due to the potential of rain scavenging, caused by monsoon precipitation. However, the seasonal pattern in Figure 4 was opposite to our expectations, indicating that rain scavenging of DDT may not be enough to remove DDT from the air. Although the DDT sources in Kathmandu and Pokhara could be attributed to the combined effects of historic and new applications of technical DDT (ratios of p,p′-DDT/p,p′-DDE were close to 1; Figure SI-4), the high concentrations of DDTs at K2 (vegetable market) and P2 (vegetable production and market area) were more likely from recent applications. Similarly, DDT sources in Hetauda were from recent applications (ratio of p,p′-DDT/p,p′-DDE = 2.2 ± 1.4; Figure SI-4). The seasonality of DDT at K2 and P2 (Figure 4), indicated that new applications of DDT mainly occur during the monsoon season while for H2 it happens during pre-monsoon season. Given that rice, maize, tomatoes, and cabbage are the major

crops and vegetables grown during these seasons, the application of DDT to control pests in soils during land preparation likely resulted in the observed seasonality.

HCHs. The dominance of lindane (γ-HCH), compared with α-HCH, in the atmosphere for all seasons demonstrated the wide application of this chemical in Nepal. γ-HCH was found at high levels during the pre-monsoon and monsoon seasons in Kathmandu, similar to the seasonal pattern of DDTs (Figure SI-9). As the capital city of Nepal, agriculture plays only a minor role in the Kathmandu economy. Increasing atmospheric concentrations of DDTs and HCHs during the warmer period (from April to November, covering the pre-monsoon and monsoon seasons) in Kathmandu may have occurred because of the combined effects of new applications and re-evaporation of the chemicals from historic residues in the soils. Contrary to this trend, the levels of γ-HCH were highest in the winter for Pokhara and Hetauda (Figures SI-10 and SI-11). This pattern was especially obvious for H2, which was located in a famous commercial vegetable produce area (Figure 4). As described above, winter is the major season for vegetable production and excessive spraying of pesticides is common (particularly for commercial purposes); possible uses of lindane for vegetable production thus could be the major reason for higher levels of γ-HCH during winter.

HCB. Figure SI-12 presents the seasonality of HCB among the sampling sites. High concentrations of atmospheric HCB occurred during winter, especially at the industrial sites (K3, P3 and H3). During the sampling period, Nepal experienced a huge power deficit in the dry winter. To achieve the required levels of electricity, numerous power generators were needed during the winter season. In addition, extensive coal combustion for brick manufacture also happened during winter. All of these reasons likely contributed to the high HCB concentrations observed in winter. Similar seasonality patterns of HCB were also reported previously in Chinese cities (Liu et al., 2009).

LRAT potential

According to the seasonality data, we know that high DDT concentrations occurred during the monsoon season (Figure 4). Given that high precipitation levels and temperatures are major characteristics of the tropical monsoon climate, this raises the question of how long DDT and other POPs will be transported, and where will they go under the tropical monsoon climate?

Characteristic travel distance (CTD) was estimated to assess the field specific LRAT potential using residential time (τ_a)(van Pul et al., 1998). The higher the CTD is, the greater LRAT potential. τ_a was defined as the time needed for 50% of the initial concentration of a POP to be lost, which can be estimated using (van Pul et al., 1998):

$$\tau_a = \frac{ln2}{k_{degr} + k_{wet} + k_{dry}} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

Where, k_{degr} is the photochemical degradation rate in air (s^{-1}), k_{wet} is the wet deposition rate (s^{-1}), and k_{dry} is the dry deposition rate (s^{-1}). The CTD was then calculated using:

$$CTD(km) = \frac{\tau_a \times u}{1000} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (4)$$

Where, u is the wind speed (m/s), which was obtained from Figure SI-1.

Loss rates (k_{degr}, k_{wet} and k_{dry}) were re-calibrated according to the local temperature, precipitation and content of hydroxyl radical [OH] (details in Text SI-5, and Tables SI-12 and SI-13). The obtained average CTD (km) of the four most dominant atmospheric pollutants (p, p'-DDT, α-HCH, γ-HCH and HCB) in the three Nepalese cities were compared with results from global models (TaPL3 and ELPOS) and estimated for other specified regions (i.e., east/south China sea, Indian Ocean, and Atlantic Ocean, Table SI-14). The CTDs estimated in this study were similar to values calculated by Shen et al. (2005) using the TaPL3 model (Table SI-14), lower than values derived from the ELPOS model, but higher than those for the Indian Ocean and Atlantic Ocean (Gioia et al., 2012) (Table SI-14); they followed the order: HCB > α-HCH > γ-HCH > p, p′-DDT. Uncertainties of the CTD estimations are provided in Text SI-6. According to Beyer's classification (Beyer et al., 2003) (high LRAT potential was defined as CTD > 2000 km; and medium LRAT potential was considered to be between 700 and 2000 km), the CTDs of POPs from the Nepalese cities fell in the range of medium and high levels of LRAT potential, suggesting that HCB and HCHs (CTD > 2000 km) emitted in Nepal could be dispersed globally and p, p′-DDT (average CTD = 1000 km) could easily reach adjacent regions, such as the high Himalayas. Due to the lack of measured data on atmospheric chemical concentrations, the LRAT potential of POPs under tropical climates has never been systematically estimated. The outcomes of this study therefore reveal that Nepalese emissions of the POPs are important globally, in respect of the transport and distribution of legacy POPs.

To test where the atmospheric POPs from the Nepalese cities could spread, 3-day air mass forward trajectories were generated; details of the generation of these trajectories are provided in Text SI-7. Forward trajectories approximate the directions and position that an air particle will travel. During the monsoon season (from June to September 2014), 60% of the trajectories (sum of clusters 1, 2 and 3; Figure SI-13) moved northward, crossing the Himalayas and reaching the southeast of the Tibetan Plateau. Combining this result with the CTD estimations, it could be considered that POPs generated from the Nepalese cities could easily be transported to the Himalayas and Tibet, which was further confirmed by the seasonality of atmospheric DDTs (peak occurred during the monsoon season) and γ-HCHs (peak occurred in March) in the southeastern Tibetan Plateau (Sheng et al., 2013; Wang et al., 2012; Ren et al., 2014). The similar seasonality in these POPs between the southeastern Tibetan Plateau and Nepalese cities indicates strongly that Nepal can be regarded as a source region of POPs that can contaminate the pristine Tibetan environment (Pokhrel et al., 2016; Gong et al., 2014).

Conclusions and Implications.

This study has provided a baseline of the atmospheric concentrations of Stockholm Convention POPs in urban regions of Nepal, which should be integrated with regional and global atmospheric monitoring campaigns for evaluating the effectiveness to reducing POP levels globally. The study indicates the continued application of DDTs and lindane in vegetable production regions and markets in Nepal, which needs further investigation. Considering the ongoing use of DDTs and lindane in Nepal, the continuous atmospheric transport of the monsoon system, and cold trapping caused by low temperatures, the high Himalayas and the Tibetan Plateau are likely key receptors of Nepalese POP emissions. In addition, increasing HCB atmospheric concentrations in the Arctic have been reported (Hung et al., 2016), and continuous HCB emissions from low latitude regions were attributed to this increase. On the basis of its volatility and the estimated CTD (> 10,000 km), we consider that once HCB is emitted in Nepalese cities, it might transfer among multiple transport systems to reach high latitude regions like the Arctic.

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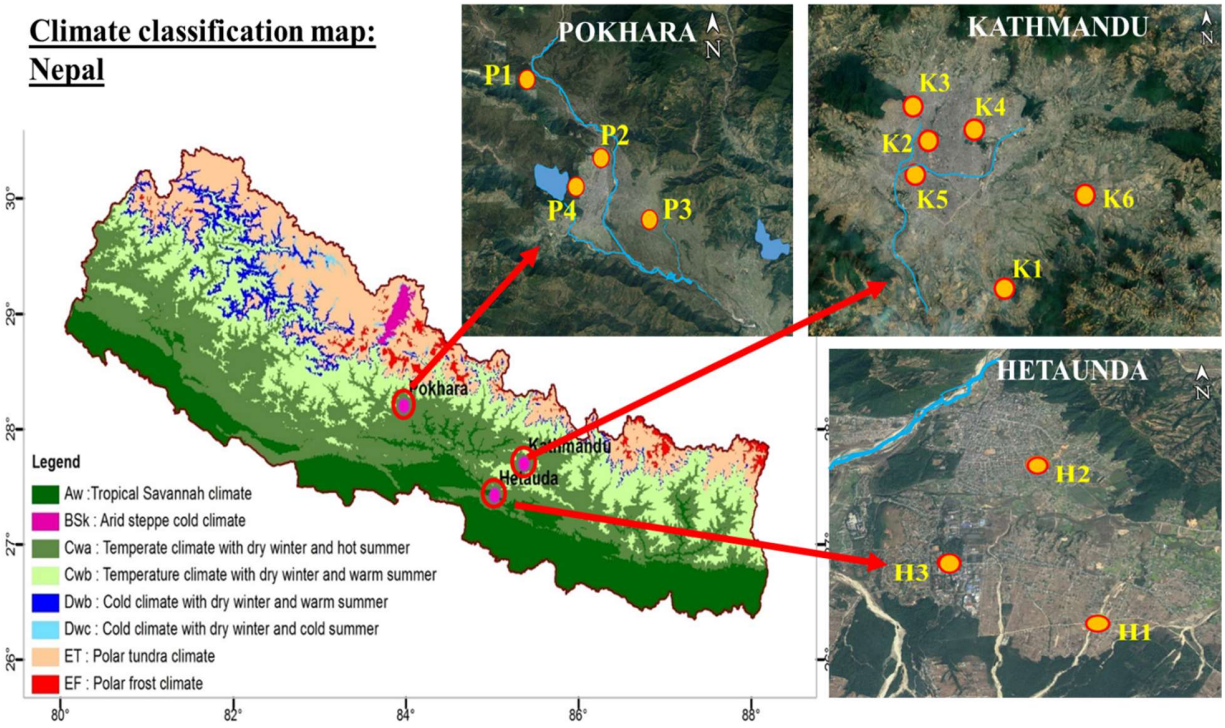

Figure 1. Map showing study area and sampling locations [modified from (Karki et al., (2016)

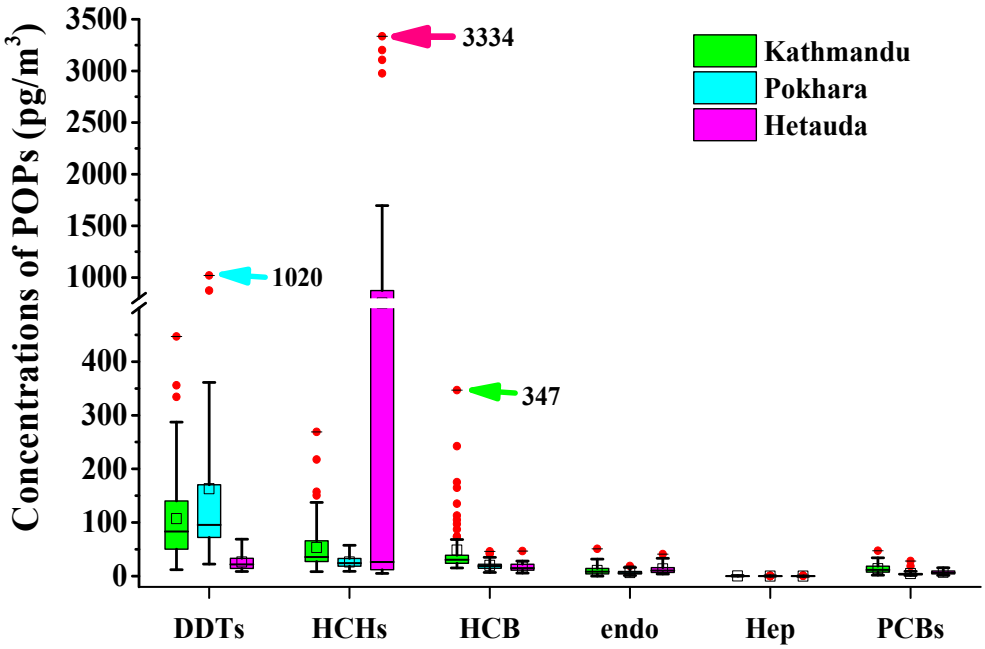

Figure 2. Box and whisker plot to show general distribution of POPs in Kathmandu Pokhara and Hetauda (Lower and upper limits of whisker indicate minimum and maximum, Lower and upper limits of the box indicate 25th and 75th percentiles, horizontal line in the box indicates median, small square in the box represents mean, red circle denotes outlier)

472 **Table 1. Air Concentrations (pg/m^3) of OCPs in different cities of Nepal and their comparison with other Asian cities**

	o, p'-DDT	p, p'-DDT	o, p'-DDE	p, p'-DDE	α-HCH	γ-HCH	HCB	α-endo	β-endo	∑PCBs	Sampling time
This Study											
Kathmandu[ϕ]	3.1–90	3.4–1.4×10^2	1.0–12	4.0–1.8×10^2	3.2–73	2.0–2.3×10^2	15–3.5×10^2	BDL–16	BDL– 35	2.1–47	
Pokhara[ϕ]	3.0–69	6.1–6.2×10^2	1.3–12	9.2–3.6×10^2	3.1–14	5.0–39	7.2–46	BDL–13	1.3–12	1.2–28	
Hetauda[ϕ]	1.2–7	2.3–49	BDL–1.4	2.2–22	1.3–11	3.1–3.3×10^3	6.3–47	1.2–5	3.3–36	2.4–16	
Previous studies											
Pakistan											
Karachi[ş]	6–66	3–57	7–19	24 – 116	6–61	27–66		24–147	4–58	37–293	Jan -Oct, 2011
Lahore[ş]	43–82	14–77	9–22	68–154	54–115	75–108		188–228	51–112	117–274	Jan -Oct, 2011
India											
Chennai[ı]	620	220		2061	1691	3562		680			Jul-Sept, 2006
Mumbai[ı]	524	188		925	637	912		498			Jul-Sept, 2006
Kolkata[θ]	34–67	56–95	3–6	23–38	28–137	34–83					Dec-Mar, 2014
New Delhi[i]	410–4430	100–1050	–	ND	600–2330	1130–3400	120–620				Nov-Dec, 2008
Urban[ı]										172	Jul-Sept 2006
Bangladesh[η]	68–380	111–692		44–393	18–62	12–242	70–685			7–1836	Feb-Mar 2013
Vietnam											
Winter[σ]	88–678	215–2035	9–70	140–912	47–184	24–206	125–752	2–58	5–95	136–615	Dec-Feb, 2013
Summer[σ]	25–424	49–809	7–43	66–327	33–501	17–122	304–998	3–98	1–36	144–1518	Jun-Aug, 2012

473 *[ϕ] This study; [ş] Nasir et al., 2014; [ı] Zhang et al., 2008; [θ] Pozo et al., 2016; [i] Chakraborty et al., 2010; [η] Nøst et al., 2015; [σ] Wang et al., 2016;*

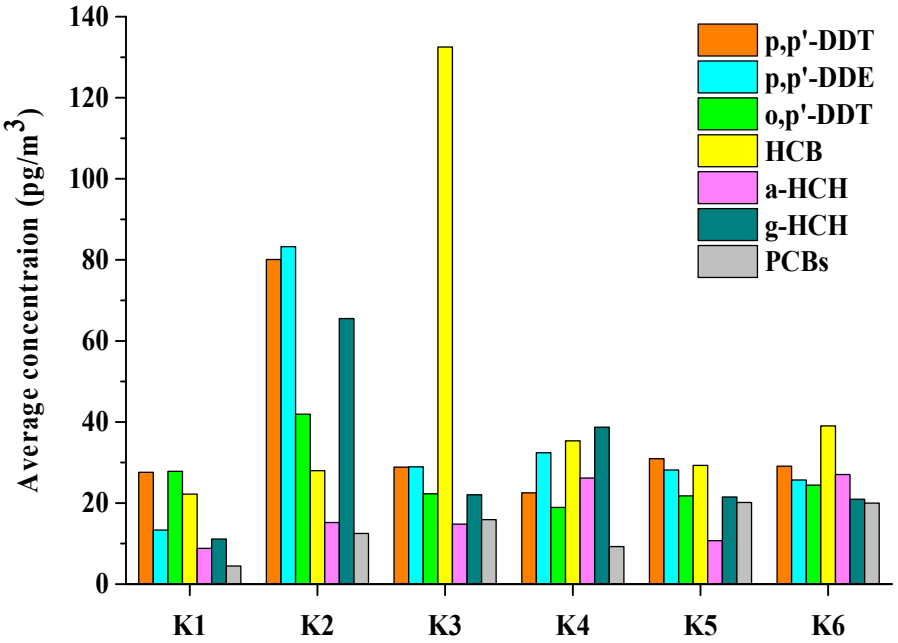

Figure 3. Distribution of OCPs and PCBs in the atmosphere of different sites of Kathmandu (K1-Cropland; K2-Market; K3-Industrial; K4-Tourist; K5-Residential; K6-Industrial and Farmland Mixed)

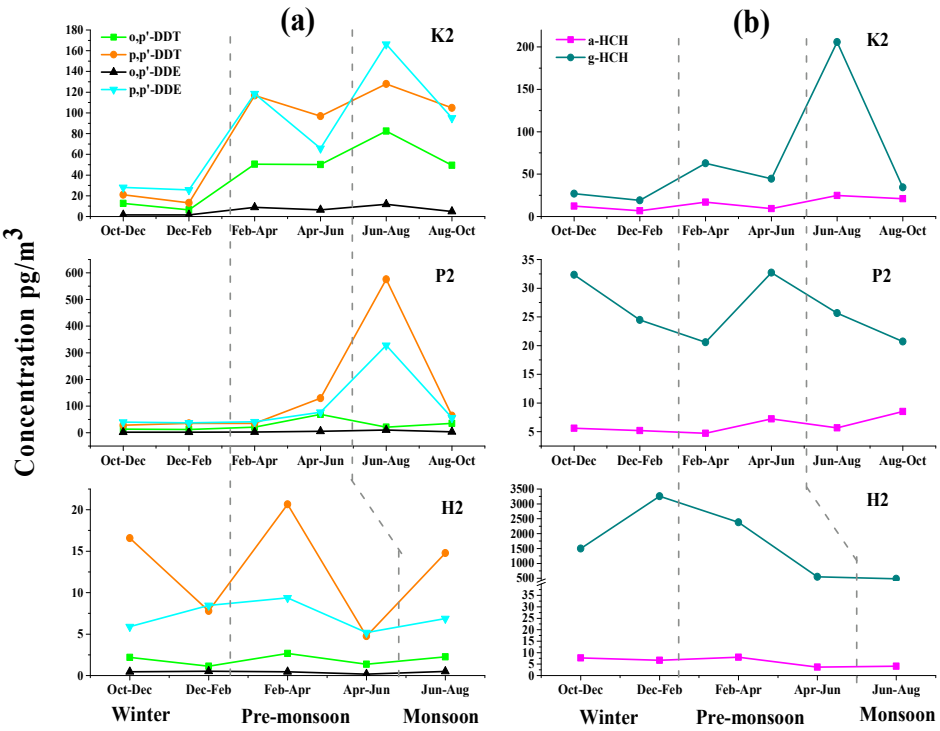

Figure 4. Seasonal variation of (a) DDTs and (b) HCHs in vegetable market or production area (K2, P2 and H2).

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