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Occurrence, profile and spatial distribution of organochlorines pesticides in soil of Nepal: Implication for source apportionment and health risk assessment

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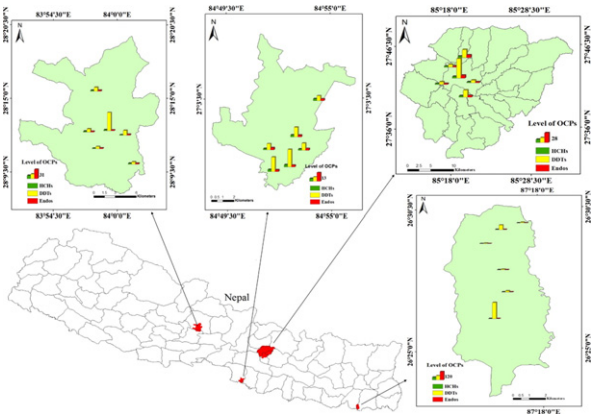
HIGHLIGHTS

- Biratnagar measured highest OCPs in soil samples.
- DDTs and endosulfans were the most abundant OCP chemicals in soil samples.
- Isomeric ratio of DDTs suggested ongoing usages of technical DDT as well as dicofol.
- TOC and BC were weakly correlated with OCPs.
- Moderate cancer risk was associated with high concentration of OCP in soil.

GRAPHICAL ABSTRACT



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ABSTRACT

Nepal is a landlocked country located between world's two most populous countries-India and China where high level of organochlorines pesticides has been reported from multi-environmental matrices. In this study, we investigated the occurrence, distributions and profile of selected OCP chemicals in surface soil samples (N = 72) from four major cities of Nepal. Overall, the sum of total OCPs in soil ranged from 20 to 250 ng/g with Biratnagar being the most polluted site in Nepal. DDTs and endosulfans were the most abundant OCP chemicals in soil samples. The concentration of DDTs ranged from 8 to 230 ng/g, 8-56 ng/g, 8-63 ng/g, and 8-27 ng/g in surface soil, while endosulfans were in the range of 2.9–3.3 ng/g, 2.8–8.7 ng/g, 2.8–3.4 ng/g, 2.8–3.2 ng/g in Biratnagar, Kathmandu, Pokhara and Birgunj, respectively. The isomeric ratio of DDT and their metabolites suggested the ongoing usages of technical DDT as well as dicofol in this region. Lower ratio of α/β-endosulfan indicated past application of endosulfans in Nepal. HCHs were less detected OCPs in soil sample accounting only 4–9% of ∑ OCPs. The isomeric ratio of α-/γ-HCH indicated that the HCHs may be originated from mixed source of technical HCH as well as lindane use. Scattered plot of TOC and BC showed they were weakly and positively related with concentration of OCPs in soil. Health risk assessment modeling study of OCPs in soil suggested moderate cancer risk with ingestion being the most potential pathway of OCPs exposure.

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1. Introduction

Organochlorine pesticides (OCPs) such as dichlorodiphenyltrichloroethane (DDT) and hexachlorocyclohexane (HCH) are diverse group of toxic substances (Jiang et al., 2009). They are highly toxic, stable and resistant to degradation in natural environment and are of global concern (Jones and de Voogt, 1999; Li et al., 2016). Because of their persistent, bio-accumulative, and toxic (PBT) behaviors, OCP chemicals pose significant threat to human health and the environment (Jones and de Voogt, 1999). In order to reduce the possible environmental threat and human harm, Stockholm Convention on persistent organic pollutants (POPs) was initiated and signed by >150 member countries, which seeks help and coordination from member countries toward elimination and reduction of POPs (UNEP, 2001). It was realized and established the fact during Stockholm Convention on POPs that a group of "dirty dozen" chemicals that are persistent must be eliminated or strictly restricted (UNEP, 2001). In order to develop effective control strategy, more information on occurrence, distribution, fate and transport of POPs from all the region is required. The participating countries were asked to conduct source inventories on POPs. Historically, DDT and HCH are most common OCPs that were widely practiced in agriculture field. The production and consumptions of POPs were banned worldwide in late 1970s. As of now, despite the ban on consumptions, production, import and export of OCPs since 1970s in many countries, they have been reported in multi-environmental matrices across the world (Ge et al., 2013; Yang et al., 2014; Whitehead et al., 2015; Yadav et al., 2015).

Soil is one of the important environmental medium which play vital role in fate, behavior and dissemination of POPs at global level (Meijer et al., 2003). Such soils can acts as significant environmental reservoirs, sinks or sources for these chemicals. It also receives plenty of POPs chemicals through atmospheric deposition and re-emits them into atmosphere (Schuster et al., 2011). POPs in surface soil can also act as secondary sources of pollutants in the air through air-soil exchange mechanism (Cabrerizo et al., 2011). In order to expand the accuracy in estimating the rate of emission, knowledge on residual level in soil is equally significant as quantities of pesticides used. Moreover, knowledge on the distribution of POPs in soil can also help to assess levels of contamination, classify the sources of emission, and assess the human health impact and environmental risks (Peng et al., 2011; Wu et al., 2011; Yang et al., 2012; Yuan et al., 2014).

Nepal is a landlocked country located in between two most populous countries India and China where elevated level of POPs in multi-environmental matrix have been reported (Jiang et al., 2009; Chakraborty et al., 2010, Devi et al., 2011; Wong et al., 2010; Ni et al., 2011; Zhang et al., 2011; Devi et al., 2013, Devi et al., 2014; Devi et al., 2015; Yadav et al., 2015). Nepal has signed the Stockholm Convention on POPs on April 5, 2002 and ratified on October 13, 2006 (NIP, 2007). Realizing and reaffirming the commitment made as a signatory and party to implement the provision of Stockholm Convention, the usage, import and trade of all POP pesticides were banned in Nepal. Nepal has neither produced POP pesticides nor sought any exemptions at the Stockholm Convention. However, due to porous border with India, large quantities of OCPs and other pesticides may enter Nepal illegally. Nepal is primarily an agrarian country of which 88% live in rural areas, where nearly 80% of young population engaged in agricultural profession. The agricultural farming shares 39% of the national gross domestic product (GDP) (DOA, 2014). In order to increase higher yield, the consumptions of POP pesticides have steadily increased in past 4–5 decades. It is estimated that about two-third of agriculture production gets destroyed during pre- and post-harvesting due to pest attacks in the field and at storage facilities (Palikhe, 2002). Thus, it is obvious for Nepalese farmer to use disproportionate quantities of pesticides to avoid pest attacks. In Nepal, the national level usages of pesticides were estimated to be about 151 g active ingredient per hectare of arable land in 2008 (Atreya and Sitaula, 2010). Unlike other countries, active ingredient pesticides are not used on per hectare basis in cropland (FAO, 2014). However, evidence suggests that pesticide use in Nepal is on increasing trend which poses significant exposures and risk to Nepalese farmer (Baker and Gyawali, 1994; PPD, 2003). Nevertheless, environmental concentration of POPs in Nepal has not been well studied. Past efforts in this region seem to have targeted long-range atmospheric transport into high altitude areas (e.g. Himalayas) rather than possible primary source regions in this lesser studied area (Gong et al., 2014; Guzzella et al., 2016). This study therefore aims to investigate the occurrence, distributions and profile of OCPs in Nepal in suspected source areas/more densely populated regions. Further, potential adverse effect of OCPs on human health in soil was estimated. The finding of this study will expand our knowledge on POPs contamination and provide useful information to local policy maker to manage health risk and implement Stockholm commendation.

2. Materials and methods

2.1. Study area

Four major cities (Kathmandu, a capital city together with 3 sub-metropolitan cities (Pokhara, Birgunj and Biratnagar)) of Nepal were selected for collection of surface soil (Fig. S1, Supplementary information). Kathmandu is the only metropolitan city in Nepal. It spreads over 50 sq·km area at elevation of 1400 m from MSL with population of 1 million (CBS/NPCS, 2011). Pokhara is the second largest city of Nepal and the headquarters of Kaski District, in Western Development Region. It is located in northwestern corner of Pokhara valley and about 200 km west of the capital city Kathmandu. The altitude varies from 827 m in the southern part to 1740 m in the north. It occupies 225 sq·km area and host 0.3 million population (CBS/NPCS, 2011). Birgunj is a sub-metropolitan municipality located in South-Central part of Nepal in the Terai plains bordering to Indian state of Bihar. It is also the headquarters of the Parsa District. It host about 0.21 million population (CBS/NPCS, 2011). It is also known as gateway to Nepal as major points of entry of goods from Bihar and Kolkata pot. Biratnagar is another sub-metropolitan city and the fourth largest city of Nepal after Kathmandu, Pokhara and Patan. It is spread over an area of 58.5 km^2 hosting 0.24 million population (CBS/NPCS, 2011). It is located in Morang District in the Kosi Zone of eastern Nepal. It is located about 399 km east to the capital city Kathmandu and only 6 km north to Indian state of Bihar. Biratnagar is also known as Industrial capital of Nepal. The details about study sites are given in Table S1, Supplementary information.

2.2. Sample collection

A total of 72 surface soil samples (18/city) were collected during August–October 2014. Each 18 soil samples were composited to 6 samples. In this way, each sample was composite of 3 sub samples. Altogether, 24 representative samples (6/city) were obtained. Soil samples were collected using stainless steel scoops at depth ranging from 0 to 20 cm in all four cities. The soil samples were then wrapped in aluminum foil, packed in zipper polythene bags and transported to laboratory keeping in ice-box. To minimize cross contamination during sampling, surgical gloves were used in hand. In laboratory, samples were freeze-dried and ground to powder and sieved through 1 mm sieve and stored at −4 °C until analysis.

2.3. Determination of total organic carbon (TOC) and black carbon (BC)

About 2–3 g of each soil sample was mixed with 3 mL of 10% HCl and transferred to 50 mL beaker and let for 8 h. Then the soil samples were washed thrice with MilliQ water and dried in oven at 45 °C overnight. A small fraction of the dried soil samples were used to measure TOC by using Elemental Carbon-Hydrogen- Nitrogen Analyzer (Elementar VARIO EL III). Black carbon (BC) was analyzed in soil samples as per

chemo-thermal oxidation (CTO-375) method detailed elsewhere (Gustafsson et al., 2001; Elmquist et al., 2008). Briefly, 2–3 g of dried soil samples were subjected to thermal oxidation (375 °C, 18 h) in a muffle furnace under constraint air flow. The soil samples were then digested with 1 N HCL and the residual organic carbon was determined as BC using Elemental Carbon-Hydrogen-Nitrogen Analyzer (Elementar VARIO EL III).

2.4. Extraction and analysis

All the solvents used under experimentation were purchased from fisher Scientific, USA and Tedia Co. USA. OCP standards 2, 4, 5, 6-tetrachloro-*m*-xylene (TCmX), decachlorobiphenyl (PCB 209) and 2, 2′, 6, 6′-tetrachlorobiphenyl (PCB54) were ordered from Ultra Scientific. All the glassware was dipped in $K_2Cr_2O_7$-H_2SO_4 solution for 24 h, properly cleaned and oven dried prior to experimentation. About 20 g of well dried and homogenized soil samples were used for Soxhlet extraction. The OCP chemicals were extracted with DCM for 24 h. A known quantity (20 ng) of TCmX and PCB209 were added as surrogate standard before extraction. To remove elemental Sulphur, small granules of copper were added in extraction flask. After Soxhlet extraction, the extract was concentrated to about 2–3 mL by a rotary evaporator. Then, the extracted samples were cleaned by alumina/silica column. From the bottom to the top, the column was packed with neutral alumina (3 cm, 3% deactivated), neutral silica gel (3 cm, 3% deactivated), 50% acid silica (3 cm), and anhydrous sodium sulphate (1 cm). The column was eluted with 30 mL solvent of DCM/hexane (1:1). The eluted fraction was concentrated and reduced to 0.2 mL using gentle nitrogen stream. About 25 μL of dodecane was added as a solvent keeper. A known quantity of PCB-54 was added as an internal standard prior to GC–MS analysis.

The eluted samples were injected in to an Agilent 7890A GC coupled with an Agilent 7000 A GC/MS Triple quadrupole in EI mode. Five hexachlorocyclohexanes (HCHs, including α-HCH, β-HCH, γ-HCH, δ-HCH, and ε-HCH) and six DDTs (including *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT, and *p,p′*-DDT), two endosulfan (α-endosulfan and β-endosulfan), heptachlor (HEPT), chlordane, hexachlorobenzene (HCB), aldrin, dieldrin, endrin, isodrin, methoxychlor and mirex were quantified using an Agilent 7890GC-7000A triple quadrupole mass spectrometer equipped with a CP-Sil 8CB capillary column (50 m × 0.25 mm × 0.25 μm) in SIM mode. Helium was used as the carrier gas at a flow of 1 mL/min. The initial oven temperature was set at 60 °C for 1 min, and raised to 290 °C at the rate of 4 °C/min for OCPs.

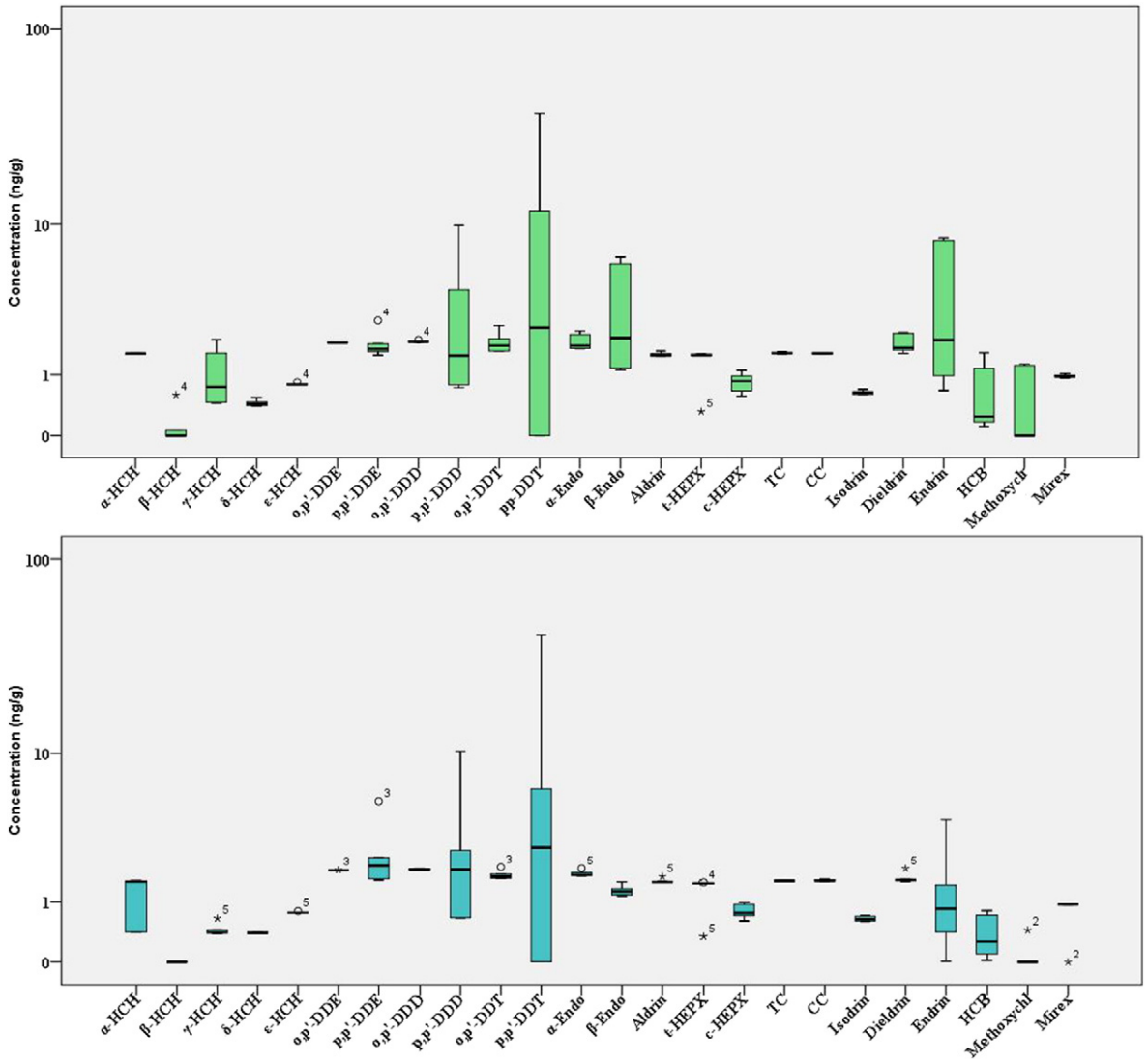

Fig. 1. Box plots showing average concentration of different OCP chemicals in soil samples of Kathmandu (Top) and Pokhara (bottom). Box plots showing average concentration of different OCP chemicals in soil samples from Birgunj (Top) and Biratnagar (bottom).

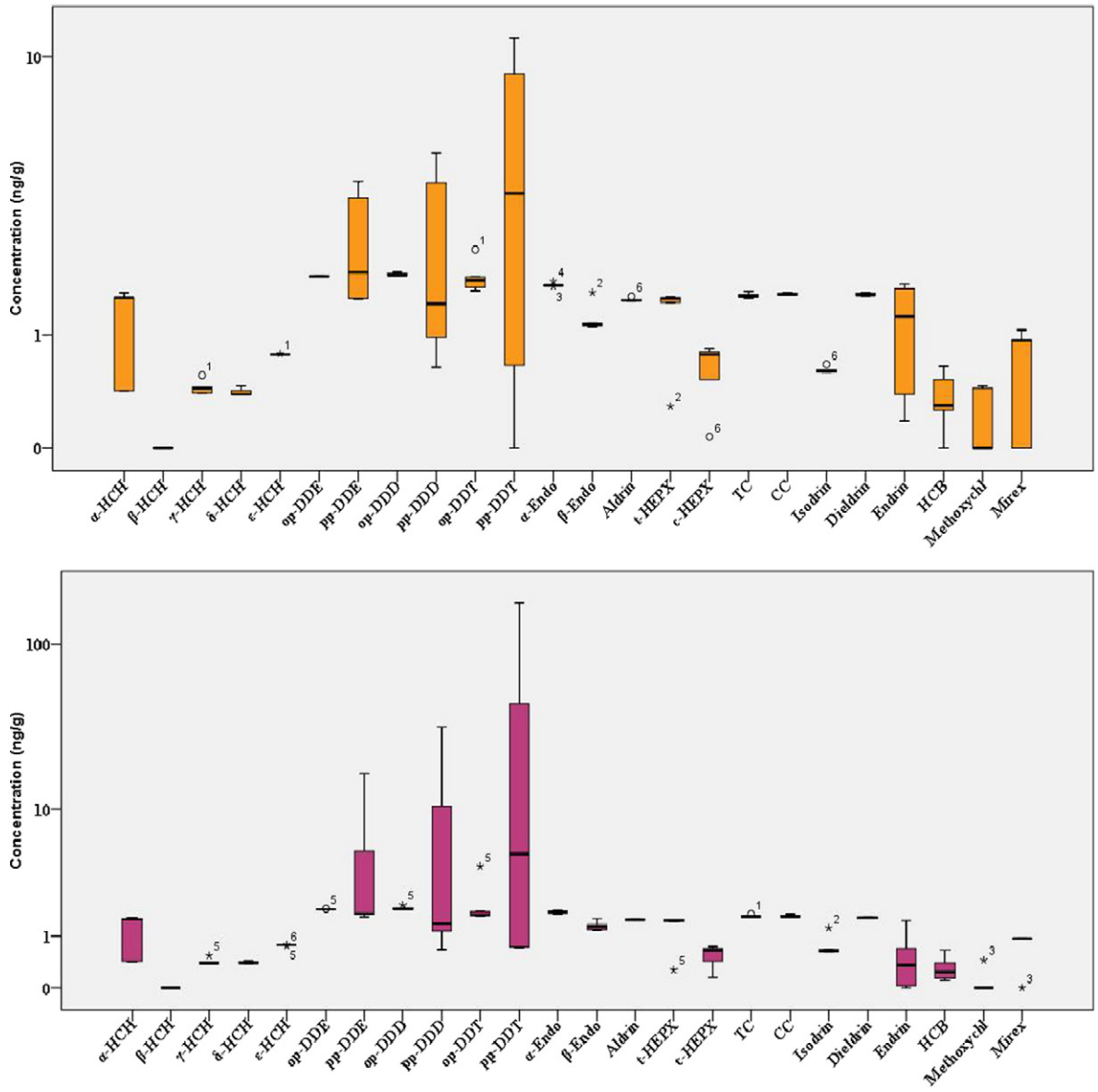

Fig. 1 (*continued*).

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

After every 12 batch of samples, a set of calibration standards solutions were run to check sort of interference and cross contamination. All the reagents used in this experiment were of analytical grade. Field, procedural and solvent blank were analyzed in the same way adopted for original sample analysis. Blank detected negligible concentration of DDT, HCH and endosulfans. The chromatogram and peak of the blank solution and standard solution were clearly identified without overlapping. The method detection limits (MDLs) of OCPs was 3 times signal versus noise value (S/N). The surrogate recoveries for TCmX and PCB 209 in all soil samples were in the range of 72 ± 11 to 88 ± 15%. OCP concentrations were expressed on dry weight basis and were not corrected for recoveries.

2.6. Statistical analysis

Descriptive statistics scattered plots and box plots were performed using IBM SPSS statistics (version 21). The Arc GIS (Version 9.3) was used to make spatial distribution maps. Samples with BDL concentration were set as zero for calculation and analysis purpose.

2.7. Health risk assessment

Ingestion, dermal contact, and inhalation are the three important routes of OCPs exposure to the human. The cancer risk and non-carcinogenic risk were calculated for OCPs pollutants in this study as per the US EPA Exposure Factors Handbook - 1997 (USEPA, 1997). The cancer risks to humans are estimated using the average daily doses (ADD) multiplied by a slope factor (SF).

$$\text{Cancer risk} = \text{ADD} \times \text{SF} \tag{1}$$

The average daily doses (ADDs, mg/kg/day) for three different routes of exposure can be calculated using Eqs. (2)–(4).

$$\text{ADD}_{\text{ingest}} = \frac{C_{\text{soil}} \times \text{IRS} \times \text{EF} \times \text{ED}}{\text{BW} \times \text{AT}} \times \text{CF} \tag{2}$$

Where,

C_{soil} is the concentration of the chemical in soil (mg/kg); IRS is the ingestion rate of soil (mg/day); EF is the exposure frequency (350 days/yr);

Table 1
Comparison of OCP chemicals in surface soil in this study with previous studies across the world (ng/g).

Location	*o,p*-DDT	*p,p*-DDT	*o,p*-DDE	*p,p*-DDE	*p,p*-DDD	∑ DDTs	α-HCH	γ-HCH	∑ HCHs	α-Endos	β-Endos	∑ Endos	References
Central Europe	nd-3.5	0.19–11.6	0.01–0.08	0.49–8.12	0.02–1.77	0.73–25.4	nd-0.16	nd-0.25	nd-0.76	–	–	–	Degrendele et al., 2016
Beijing, China	nd-533	nd-1925	–	0.22–832	0.15–50.4	0.77–2179	0.32–8.2	nd-12.86	1.36–57	–	–	–	Zhu et al., 2005
Shenzhen, China	0.31–25.1	0.41–119	0.53–3.87	0.27–50.5	–	1.93–199	0.03–9.86	0.03–90.7	0.35–281	0.26–46.8	0.25–0.39	0.51–47.2	Ni et al., 2011
Shanghai, China	nd-21.01	nd-24.52	–	0.17–77.8	0.15–124.7	0.44–247.4	nd-1.46	nd-4.15	nd-10.4	nd-1.77	nd-2.33	nd-4.1	Jiang et al., 2009
Mexico	nd-20	nd-110	nd-1.6	0.03–203	nd-23	nd-360	nd-0.09	nd-0.13	nd-0.14	nd-243	nd-440	0.01–909	Wong et al., 2010
Kihihi, Uganda	11–20	7–34	6–14	2–10	–	26–78	–	–	–	4–8	5–10	12–23	Ssebugere et al., 2010
Narok, Kenya	–	1.53–9.13	–	0.04–1.62	nd-1.72	1.88–11.33	nd-1.62	nd-1.31	nd-7.31	nd-0.53	0.13–2.56	0.13–9.16	Sun et al., 2016
Hyderabad, Pakistan	0.01–0.15	4.2–55	0.6–1.1	5.9–11.3	11–20	28–41	44–134	8.3–15	68–164	–	–	–	Alamdar et al., 2014
Korba, India	1.2–36	0.8–116	8.5–15	1.6–135	1.2–21	2.1–315	0.9–9.3	0.9–11	0.9–16	–	–	–	Kumar et al., 2014
Kurukshetra, India	0.59–10.9	0.55–7.1	–	0.54–8.49	0.66–18	0.5–37	0.56–2.1	0.75–2.24	0.6–8.5	–	–	–	Kumar et al., 2013
IHR, India	nd–484	0.28–1448	nd-1.97	nd-78.8	nd-103	0.28–2127	nd-0.33	nd-1.28	nd-2.79	nd-0.54	nd-1.26	nd-2.83	Devi et al., 2015
Manipur, India	0.07–1.44	nd-0.77	nd-0.05	0.02–1.91	0.03–1.01	0.5–5.68	nd-0.10	nd-1.05	0.01–2.9				Devi et al., 2013
Kathmandu	1.6–2.47	nd-37.4	1.86–1.88	1.5–2.67	0.71–9.87	7.6–56	1.51–1.55	0.44–1.96	3.1–5	1.68–2.3	1.1–6.53	2.8–8.7	This study
Pokhara	1.61–1.97	nd-41.4	1.86–1.87	1.55–5.31	0.66–10.3	7.5–62.7	0.42–1.54	0.39–0.66	1.97–3.4	1.68–1.9	1.14–1.5	2.8–3.4	This study
Birgunj	1.62–2.37	nd-11.3	1.86–1.87	1.49–4.12	0.64–5.10	7.5–26.7	0.42–1.59	0.4–0.56	1.98–3.4	1.69–1.8	1.1–1.59	2.8–3.2	This study
Biratnagar	1.61–4.1	0.71–173	1.86–1.89	1.59–16.7	0.67–32.7	8.4–230	0.42–1.56	0.39–0.54	1.97–3.3	1.69–1.8	1.1–6.53	2.93–3.4	This study

ED is the exposure duration (24 yr); CF is the conversion factor (1×10^{-6} kg/mg); BW is body weight (70 kg); and AT is the average lifetime (25,550 days).

$$ADD_{dermal} = \frac{C_{soil} \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \times CF \quad (3)$$

Where, SA is soil surface area (cm^2); AF is the skin adherence factor for soil (mg/cm); and ABS is the dermal absorption factor from the soil (chemical specific);

$$ADD_{inhale} = \frac{C_{soil} \times IhR \times EF \times ED \times \frac{1}{PEF}}{BW \times AT} \quad (4)$$

Where, IhR is the inhalation rate (m^3/day); and PEF is particulate emission factor (1.36×10^9 m^3/kg).

The hazard quotient (HQ) is used to estimate the non-carcinogenic risks of OCPs in soils of different exposures. It is the ratio of the ADD and the specific reference dose (RfD) and can be estimated using following Eq. (5).

$$HQ = \frac{ADD}{RfD} \quad (5)$$

RfD (mg/kg/day) is the daily maximum allowable dose of pollutants without posing noncarcinogenic risk to human during life-span. Generally, there are three RfDs for three different exposure pathways as follows: reference dose (RfDo, mg/kg/day) for ingestion, RfD_{ABS} ($RfD_{ABS} = RfD_o \times ABS_{GI}$, mg/kg/day) for dermal contact and reference dose of inhalation (RfCi, mg/m^3) for inhalation. ABS_{GI} is the fraction of the pollutant absorbed in the gastrointestinal tract in the critical toxicity study (unit less).

The total risk of specific chemicals through multiple exposure routes is expressed as the hazard index (HI). The total risks from exposure to OCPs in soil are calculated as per Eq. (6).

$$HI = \sum HQ_i \quad (6)$$

Where, i = different exposure pathways.

The specific and constant parameter used in this health risk assessment model is given in Table S2, Supplementary information.

3. Results and discussions

3.1. General comments on OCPs concentration

The average concentration of individuals OCPs analyzed in soil samples are plotted in Fig. 1. The highest concentration of total OCPs was measured in Biratnagar. Overall, the average concentration of ∑OCPs in Biratnagar was almost 2 fold higher than rest of the sites. The levels of individual OCP isomers/metabolites in all four cities were assessed and compared using box-plots (Fig. 1). DDTs and endosulfans were most dominant OCP chemicals in the soil. DDTs concentrations were higher than the concentration of HCHs observed in this study. This is because of large quantities of DDTs used in agricultural activities than HCH formulations and less extent due to difference in physicochemical properties (Zhang et al., 2015). HCHs have lower lipophilicity and particle affinity can biodegrade rapidly compared with DDTs (Loganathan and Kannan, 1994). Specifically, the elevated level of DDTs as observed in Biratnagar, were higher than urban soil from Shanghai (China), Kurukshetra (India) and Central Europe (Table 1), however, it was much lower than reported from Beijing (China), IHR (India) and Mexico. High level of DDTs was also observed in surface soil from Indian cities, indicating the past and ongoing use of DDTs (Kumar et al., 2014; Devi et al., 2016). With regards to endosulfans, the concentration in Kathmandu was much lower than reported Mexico and Shenzhen, China (Wong et al.,

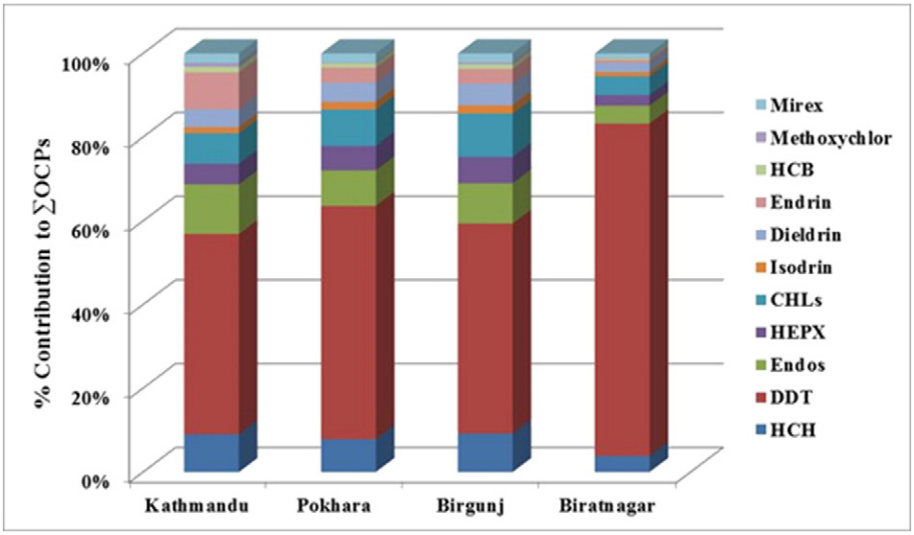

Fig. 2. % contribution of individual chlorinated pesticides to total OCPs.

2010; Ni et al., 2011), and were comparable to India, Shanghai China and Kenya (Jiang et al., 2009; Devi et al., 2015; Sun et al., 2016). Heptachlor was not detected either of soil samples. However, heptachlor epoxide (t-HEPX and c-HEPX) were less detected and were higher than global studies (Fu et al., 2009; Devi et al., 2015; Sun et al., 2016).

3.2. Intercity variation and compositional profile

3.2.1. DDTs

DDTs accounted 48–80% of total OCP chemicals in this study (Fig. 2). Highest concentrations of DDTs were detected in Biratnagar in the range of 20.4–250.3 ng/g (mean 73.2 ng/g). The concentration was doubled times greater than concentration detected in Kathmandu, Pokhara and Birgunj. The elevated level of DDTs in Biratnagar attributed to large amount of DDTs used in intensive farming and malaria control (Tarcau et al., 2013). DDTs and its metabolites were detected in all study sites (Fig. 3). The relative abundance of DDT metabolites showed abundance of pp.-DDT (Fig. S2, Supplementary information). Technical DDT is a mixture of 85% *p,p′*-DDT and 15% *o,p′*-DDT (Zheng et al., 2009). The isomeric ratio of *o,p′*-DDT/*p,p′*-DDT is used to differentiate the contribution of dicofol from technical DDT. The ratio of *o,p′*-DDT/*p,p′*-DDT between 0.2 and 0.3 shows the presence of technical DDT, ratio between 1.9 and 9.3 or higher suggests contribution from dicofol (Qiu et al., 2005). In this study, the ratio of *o,p′*-DDT/*p,p′*-DDT ranged between 0.05 and 0.37 except three samples, suggesting the usages of technical DDT. One sample in Birgunj (BRS-2) and two samples in Biratnagar (BTS-3 and BTS-4) comparatively showed higher *o,p′*-DDT/*p,p′*-DDT ratio. This corresponds with dicofol usage in this region (Fig. 4). The ratio of *p,p′*-DDT and their metabolites (*p,p′*-DDE and *p,p′*-DDD) are used to assess the amount of DDT degradation in the environment (Devi et al., 2015). DDTs can be degraded to DDD through reductive de-chlorination under anaerobic environment; while it changed to DDE under aerobic environmental conditions. The ratio of *p,p′*-DDT/(*p,p′*-DDD + *p,p′*-DDE) >1 is indication of current ongoing use, while ratio <1 infers historical use. In this study, the ratio of *p,p′*-DDT/(*p,p′*-DDD + *p,p′*-DDE) ranges between 1 and 3, specifying the ongoing usage of technical DDT (Fig. 4). Similar finding were reported from Indian cities (Devi et al., 2015).

3.2.2. Endosulfans

Endosulfan comprised 4–12% of total OCPs in all the study sites (Fig. 2). Average higher concentrations of endosulfan were observed in Kathmandu in the range of 2.8–8.7 ng/g (mean 5 ng/g). The concentration of endosulfan detected in Pokhara, Birgunj and Biratnagar were similar and ranged 2.83–3.43 ng/g, 28–3.28 ng/g and 2.93–3.35 ng/g, respectively (Fig. 1). Fig. 3 represents spatial distribution of OCPs. Highest concentrations of endosulfans were detected at KTS-4 and KTS-5 in Kathmandu. Commercial technical endosulfan is mixture of α-endosulfan (70%) and β-endosulfan (30%) (WHO, 1984; Sun et al., 2014). The α-endosulfan decomposed more rapidly than β-endosulfan in soil, while α-/β-endosulfan isomers can be easily reduced to endosulfan sulphate by microbes, which is difficult to be further degraded (Qiao et al., 2010). The ratio of α-/β-endosulfan <2.3 is indicative of historical use of endosulfan whereas ratio >2.3 states recent use. In this study, α-/β-endosulfan ratio were below 2.3 in all the study sites, suggesting past application of endosulfans in this region.

3.2.3. HCHs

The concentrations of HCHs were relatively low accounting only 4–9% of ∑OCPs in all study sites (Fig. 2). Kathmandu detected maximum concentration of HCHs and ranged 3.1–5 ng/g. Concentration of HCHs was similar in Pokhara, Birgunj and Biratnagar. Comparatively, α-HCHs were the most abundant congeners observed in soil (Fig. S2). All the HCH isomers were detected in soil samples except β-HCHs. β-HCH were only detected at two sampling sites (KTS-4 and KTS-5) in Kathmandu (Fig. S2). The highest concentration of ∑HCHs detected at KTS-4 and KTS-5 in Kathmandu. The isomeric ratio of α-HCH and γ-HCH is used as indicator of HCHs sources. α-/γ-HCH ratio is <3 advocates application of lindane, whereas ratio between 3 and 7, represents technical input of HCHs (Jiang et al., 2009). In this study, the α-/γ-HCH ratio in 50% of total sites showed >3, while in remaining 50% of sampling sites, the α-/γ-HCH ratio were below 3, demonstrating that HCHs may be originated from mixed source of technical HCH as well as lindane use. Further, comparatively lower relative abundance of γ-HCH than α-HCH confirms usage of technical HCHs in Nepal. The finding can be supported by the fact that wide spread lindane was used in Nepal before they were banned. Lindane is also used as essential drug in treatment of scabies disease in Nepal (NIP, 2007).

3.2.4. Chlordane-related compounds

Although, the import, trade and usage of all persistent organic pollutants (POPs) including chlordane were banned in Nepal in 2001, the transboundary movement and illegal importation due to open border with India many banned pesticides is still found in local market. Chlordane pesticides are popular pesticides among Nepalese farmer and are widely used in cash crops such as sugarcane, tomato, potatoes, and vegetables. The main constituents in technical chlordane are TC (13%), CC

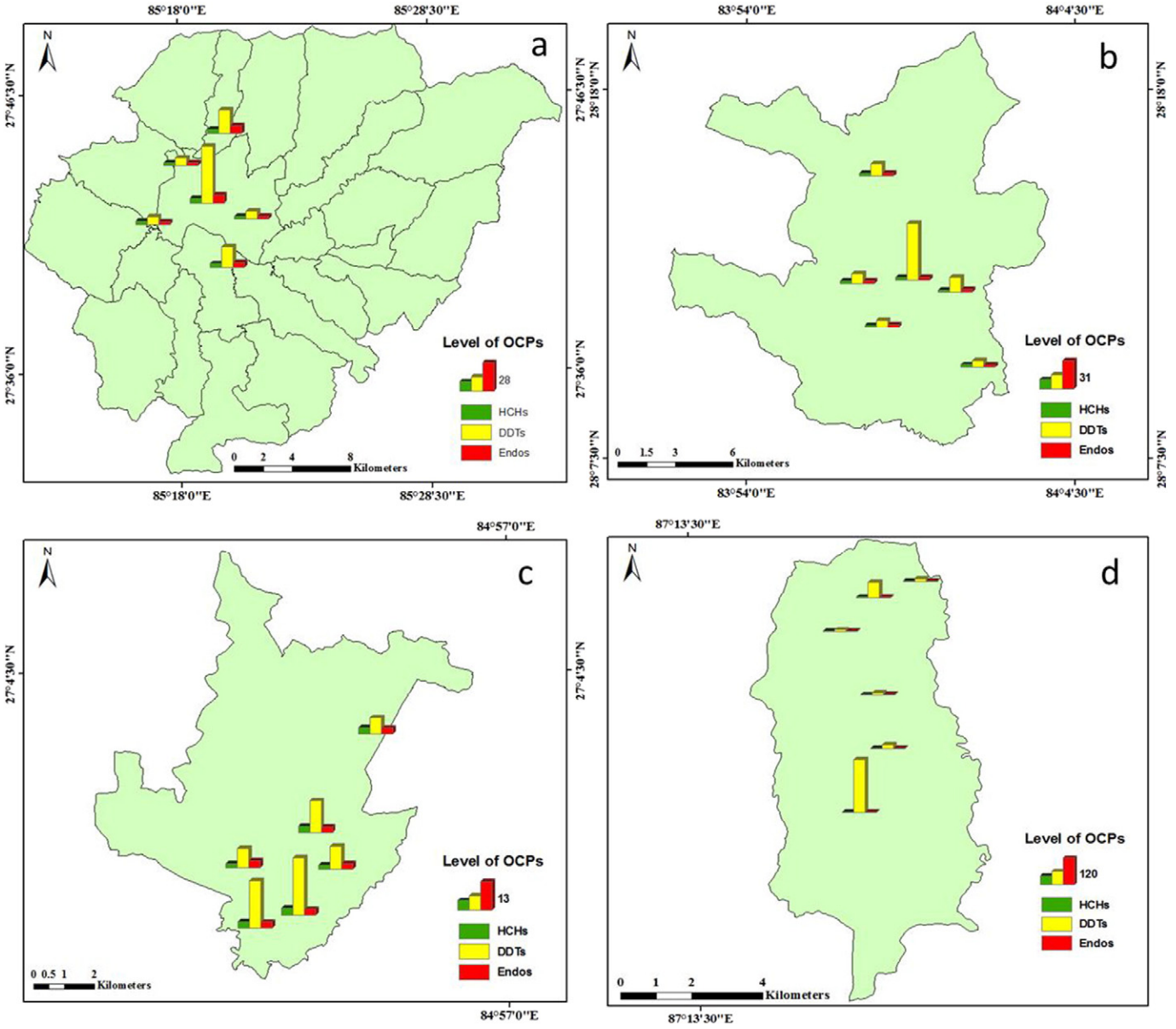

Fig. 3. Spatial distributions of DDTs, HCHs and endosulfans in (a) Kathmandu, (b) Pokhara (c) Birgunj and (d) Biratnagar.

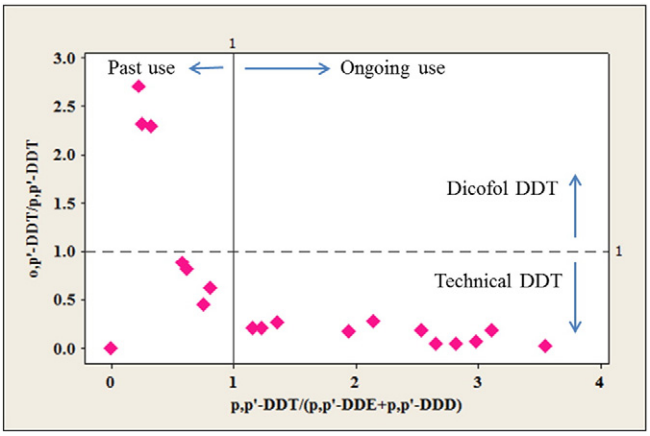

Fig. 4. Scattered plots of *o,p′*-DDT/*p,p′*-DDT and *p,p′*-DDT/(*p,p′*-DDD + *p,p′*-DDE).

(11%), HEPT (5%) and *trans*-Nonachlor (5%) (Zhang et al., 2012). Chlordane chemicals were less detected OCPs and accounted only 4–9% of ∑OCPs (Fig. 2). The relative abundance of cis-chlordane (CC) and trans-chlordane (TC) showed stable concentration irrespective of sampling sites in all the study sites (Fig. S2). The CC and TC concentration were in the range of 1.52–1.62 ng/g and 1.50–1.69 ng/g, respectively.

TC/CC ratio is used to indicate the recent use and/or historical usage of chlordane. Likewise, TC/CC ratio 1.17 represents technical chlordane (Chakraborty et al., 2010). In this study, the TC/CC ratio was near unity, suggesting the past application of technical chlordane. Low TC/CC ratio observed in this region might also due to remission from weathered chlordane source (Devi et al., 2015). HEPT is a chlorinated cyclodiene insecticide which was segregated from technical Chlordane in 1946 (Zhang et al., 2012). It may be degraded to HEPX subject to photolysis or epoxidation in soils, plants and animals. HEPT were not detected, while HEPX were detected in the range of 0.36–2.62 ng/g, in all study sites, indicating the complete degradation of HEPT.

3.2.5. Hexachlorobenzene (HCB)

Industrial and agricultural usage of HCB, together with emissions from incomplete combustion of waste, coal, fuel, and biomass results in elevated level of HCB (Bailey, 2001). HCB can also release into the environment during various chemicals process such as production of chlorinated solvents and chlorinated aromatics and pesticides as byproduct. HCB was ubiquitous and detected in all study sites except BRS-2 site in Birgunj. The soil concentration of HCB ranged from 0.11–1.56 ng/g, 0.02–0.81 ng/g, nd-065 ng/g and 0.1–0.66 ng/g in Kathmandu, Pokhara, Birgunj and Biratnagar, respectively. Highest concentration of HCB was detected in soil samples at site KTS4 and KTS-5 in Kathmandu.

3.3. Relationship with TOC and BC

Because of hydrophobicity behavior, OCPs get adsorbed by soil organic matter when in the surface soil. Soil organic matter provides sufficient amount of carbon to soil microbes in order to facilitate OCPs degradation process (Meijer et al., 2003; Nam et al., 2008). Accordingly, TOC significantly influence the fate and behavior of OCPs in soil. In this study, TOC in soil samples varied from 0.10% to 2.47%. To investigate the role of TOC, concentration of OCPs were plotted with % TOC content in soil using scatter plot (Fig. S3). OCPs were weakly but positively correlated with TOC contents in soil (DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, *p,p′*-DDE, *p,p′*-DDT, δ-HCH, ε-HCH, HCB and CC (R^2 = 0.065, 0.068, 0.090, 0.072, 0.061, 0.061, 0.049, 0.111, 0.065, 0.01 at $p \le 0.05$, Fig. S3). This suggests other factors such as land use type, particle size and soil chemistry played more important role than organic carbon in adsorbing OCPs in soil (Jiang et al., 2009; Zhang et al., 2013). The linear correlation between OCPs and TOC in this study is in line with previous study from Central Europe and Central Tibetan Plateau (Degrendele et al., 2016; Yuan et al., 2014).

Incomplete combustion of fossil fuel or biomass generates Black carbon (BC) (Ni et al., 2014). It is an important type of carbonaceous geosorbent (Accardi-Dey and Gschwend, 2002) that affects the overall behavior of POPs in soils, partially because of its porous nature (Sun et al., 2008) and due to high adsorptive capacity. Low level of BC is quantified in soil samples. The % BC in soil samples in this study ranged between 0.03% and 0.58%. BC content in soil samples in this study didn't show significant correlation with OCPs. Like TOC, BC content in soil was also weakly correlated with OCPs, especially for HCHs, Endos and HCB (R^2 = 0.012, 0.019, 0.026 at $p \le 0.05$, Fig. S4) suggesting adsorption onto BC is not the important mechanism of distributions of OCPs in Nepal.

3.4. Health risk assessment

Although, EPA factors for the human exposure may not be entirely adequate for Nepal, it provides evidence of exposure of carcinogenic and non-carcinogenic OCPs in Nepalese environment. The average daily dose (ADD), non-carcinogenic risk (HQ) and total lifetime carcinogenic risk (HI) of OCP chemicals Nepalese soil through ingestion, dermal contact and inhalation is given in Table S3 and S4, Supplementary information. Total cancer risk was calculated only for those chemicals for which SF and IUR are available. As can be seen from Table S2, the total lifetime risk in all the study sites ranged between 1.63×10^{-7} and 7.66×10^{-5}. All the chemicals in all study sites didn't exceed the target risk level of 1×10^{-6}, except *p,p′*-DDT. *p,p′*-DDT is the only chemicals exceeded the target risk value indicating potentially high health risk is associated with *p,p′*-DDT in soil. Also, *p,p′*-DDD and *p,p′*-DDE exceeded the target risk value in Biratnagar, suggesting moderate level cancer risk. Therefore, the local authorities and policy makes are advised to focus on DDT pollution when implementing Stockholm Convention's commendation. For different exposure pathways, the higher cancer risk for OCPs was through ingestion followed by dermal contact and inhalation.

4. Conclusions

In this study, we assessed the contamination level of OCPs in surface soil collected from four major cities of Nepal. The soil in Nepal is contaminated with OCPs especially, at Biratnagar. The new input of DDTs was still present in Nepalese soil that might be due to easy access of such pesticides in local market because of open border with India. High concentration of DDTs and endosulfans in soil suggests their application in both agricultural and health sector for malaria control. The soil was moderately contaminated with OCPs, especially by DDT at Biratnagar. Therefore, the local authorities and policy makes are suggested to focus on DDT pollution when implementing Stockholm Convention's commendation.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2016.09.133.

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