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Polychlorinated biphenyls and organochlorines pesticides in indoor dust: An exploration of sources and health exposure risk in a rural area (Kopawa) of Nepal

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ABSTRACT

While contamination of indoor environment with organochlorine compounds (OCs) is well documented worldwide, only a few studies highlighted the problem of indoor pollution in Indian sub-continent, including Nepal. This study insight the contamination level, distribution pattern, and sources of OCs in indoor dust from a rural area of Nepal. Additionally, daily exposure risk through different intake pathways was estimated in order to mark the potential risk of OCs to local residents. Results indicated the predominance of organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) in dust. Σ_{26}OCPs (median 87 ng/g) in dust was about 7–8 times greater than Σ_{30}PCBs (median 10.5 ng/g). DDT was the most abundant chemical among Σ_{26}OCPs, followed by HCHs and endosulfan, and accounted for 73%, 7%, and 4% of Σ_{26}OCPs, respectively. A relatively high level of ΣDDT than other OCPs suggests the existence of DDT source in the Nepalese environment. Among PCB, tetra-CBs were most prevalent, trailed by penta-CBs, hexa-CBs, and hepta-CBs, and comprised 28%, 21%, 17% and 17% of Σ_{30}PCBs, respectively. Dioxin like-PCBs (median 3.48 ng/g) was about two times higher than the total indicator-PCB (median 1.63 ng/g). High *p,p*-DDT/*p,p*-DDE ratio (median 2.89) suggested fresh application and minimal degradation of DDT in the local environment of Kopawa. While lower α-/γ-HCH ratio (median 0.75) indicated lindane contamination as the primary sources of HCH. Moreover, the low α-/β-endosulfan ratio (median 0.86) specified the fresh use of commercial endosulfan. Among OCPs, only DDT positively related to total organic carbon (TOC) ($Rho = 0.55$, $p < 0.05$) but not black carbon (BC), proposing minimal or zero impact of TOC and BC. For PCBs, PCB-126 was moderately and negatively correlated with TOC ($Rho = -0.49$, $p < 0.05$), but not BC. The daily risk exposure (DRE) assessment showed that children are more vulnerable to OCs than the adult. The DRE of OCs in this study were 2–4 order of magnitude lower than their corresponding reference dose (RfD), proposing insignificant risk.

1. Introduction

Organochlorine compounds (OCs) are synthetic chemicals that constitute one or more chlorine element linked with carbon. Polychlorinated biphenyls (PCBs) and organochlorine pesticides (OCPs) are two crucial class of OCs (Pelletier et al., 2003). Both PCBs and OCPs are stable chemicals because of their chemical structure (Kutz et al., 1991). They are lipophilic and can remain in the environment and fatty tissue for quite a long time because of their long half-lives (Pelletier et al., 2003). Globally, PCBs are more commonly utilized in capacitor and transformer in the form of dielectric fluid. Industrially, it is also used as flexible sealants, fire retardant coatings, and in several other household materials, for instance, paints, finishes, and light apparatus (DellaValle et al., 2013: IARC, 2016). Realizing the environmental toxicity and persistence characteristics of OCs, they were banned globally in 1960–1980 (Kutz et al., 1991; ATSDR, 2004, 2005; Darnerud, 2003). Although the usages of dichlorodiphenyltrichloroethane (DDT) (one of the most popular insecticide) in agribusiness has been banned worldwide, a limited amount of DDT still being used specially in tropical region for vector-born disease control such as

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malaria and bubonic plague (Loomis et al., 2015). Chlordane was used in a residential lawn, garden, and few crops such as potatoes and corn. Furthermore, it was also utilized for eradicating termites by professional applicators (IARC, 2016). The worldwide production of DDT and PCBs is estimated to be about one million tons, separately (Voldner and Ellenton, 1987; Breivik et al., 2004).

Indoor dust is an essential sink of various organic pollutants in the residential condition. Hence, the degree of OCs in indoor dust can be utilized as an intermediary to evaluate the contamination level and human exposure in the residential state (Lioy et al., 2002; Hwang et al., 2008). Furthermore, it serves as a good reservoir of OCs contaminants. OC chemicals is known to persist more under residential condition than the outdoor environment due to their minimal biotic and abiotic degradation. Dust in indoor environment has been considered as a significant medium of exposure, particularly children population due to their regular hand to mouth contacts (Mercier et al., 2011). A number of studies have demonstrated significant health risk due to exposure of OCs in indoor environment (Mitchell et al., 2007; Wu et al., 2007). Ingestion of dust is an essential medium of human exposure to different types of organic pollutants, especially countries with less industrialization where flame retardant chemical is less prominent (Dirtu and Covaci, 2010; Harrad et al., 2010; Whitehead et al., 2011; Yadav et al., 2019a). Despite the ubiquitous presence of OCs in the indoor environment, very little is known about the degree of OCP and PCB contamination, their distribution characteristics, and sources in the whole of the Indian subcontinent, especially in case of Nepal. Accordingly, characterizing the fate, distribution, and sources of OCs in the indoor condition of Nepal required significant attention. It is equally imperative to explore the connection between the degree of indoor pollution and socioeconomic status.

Albeit authoritatively prohibited, there are reports of unauthorized utilization of DDT and HCHs in few developing nations, including Nepal, due to improper implementation law (Yadav et al., 2016). The transfer of OCs from outside through window/ventilation and discharge from building structure, such as floors carpets and wood, are the another critical medium of OCs occurrence inside the house. (Harrad et al., 2010; Hwang et al., 2008). Even though PCBs are never again being used, their vaporization, abrasion of building items, and consumer goods can result in substantial dust pollution in the house (Herrick et al., 2004; Rudel et al., 2008). Additionally, long-range atmospheric transport and deposition processes can relocate OCPs and PCBs to a remote location far away from the source (Iwata et al., 1993). OCs may also reach to the residential domain through particulate matter stored on human clothing and many other related items.

Although few sporadic studies have detailed significant level of OCP/PCBs in indoor dust (Yadav et al., 2019b), soil (Yadav et al., 2016, 2017a) and air (Yadav et al., 2017b) from urban area in the Himalayan state of Nepal, the plausibility of longstanding contamination of OCPs/PCBs in house dust in rural condition remains unexplored. While dietary exposure is commonly perceived as a significant source of OCs and may explain the human level of OCs, the current knowledge, and information about the influence of combined exposed source for the non-occupational group are still limited. The selection of the indoor environment for studying the legacy POPs was based on the fact that human spends most of their times inside houses. It is, therefore, essential 1) to evaluate and investigate the presence of the legacy POPs in the indoor environment of rural Nepal, 2) to compare the magnitude of legacy POPs in this study with worldwide studies; and 3) to estimate the extent of human exposure (adults and children) of these pesticides. This is a subsequent study of the previous one where brominated, chlorinated, and organophosphate flame retardants were investigated in indoor dust (Yadav et al., 2019a).

2. Materials and methods

The comprehensive experimental protocol for the determination of OCP and PCBs in dust has been discussed elsewhere (Yadav et al., 2019b) and also in the supplementary information (SI).

2.1. Study area and sampling

Detail of study area and sampling sites have been discussed elsewhere (Yadav et al., 2019a) and also presented in Table S1. Briefly, house dust samples were collected by vacuuming the domestic, residential, and public places in Kopawa village (a lower administrative division) of Nepal. The kitchen room, study room, bedroom, living room, and passage of the households were vacuumed to have a representative sample. A single dust bag was used in a vacuum cleaner for collecting dust in individual houses. In this way, a total of 21 dust samples (roughly 50 g each) were collected and transported to laboratory keeping in zipper airtight plastic bags. Later, dust samples were dried under freeze dryer, well mixed, and screened with a mesh size of 500 μm. The pretreated dust samples were preserved at −20 °C before chemical investigation.

2.2. Extraction and GC-MS analysis

Extraction, purification and chemical analysis of dust samples for OCshas been discussed previously (Yadav et al., 2016, 2017b, 2019b). Briefly, 10 g dust samples were used for extracting the OCs chemicals in dichloromethane (DCM) by soxhlet extraction unit for 24 h. Two recovery standards (tetrachloro-m-xylene; TCmX, and PCB 209) were added (20 ng each) in the extracting solvent prior to extraction. After soxhlet extraction, the extractant was concentrated on rotary evaporation and subjected to acid silica-alumina column purification. Later, the eluent was reduced to about 0.2 μL under gentle nitrogen flow before transferring to GC vial. Accurately, 20 ng of PCB-54 and 10 ng of ^{13}C-PCB-141 was added as the injection standard before GC-MS investigation.

OC chemicals were analyzed on Agilent 7890 A GC coupled with an Agilent 7000 A MS Triple quadrupole. OCP and PCB chemicals were separated using CP-Sil 8CB capillary column (50 m length, 0.25 mm internal diameter, × 0.25 μm film thickness) with a column flow of helium (99.9999%) of 1 mL/min. The mass spectrometer was operated using electron impact (EI) mode with selected ion monitoring (SIM). One μL sample was introduced in pulsed splitless mode. The GC oven program used was: 60 °C for 1 min, 30 °C/min to 220 °C held for 0 min, and 5 °C/min to 300 °C held for 15 min All26 target compounds of OCPs and 30 out of 32 PCBs were detected in dust. The complete nomenclature and other details of individual OCP and PCBs have been detailed in SI.

2.3. QA/QC

Blank samples were used to check cross-contamination and interference for laboratory or field conditions (Yadav et al., 2019b). Ten laboratory and three field blanks were investigated with GC-MS in a similar manner the original samples analyzed. The method detection limits (MDLs) read 3 times standard deviation plus mean of all the blank samples. The blank concentration of OCP and PCB varied from nd-2.65 pg/g and nd-1.03 pg/g, respectively. In the absence of OC compounds in blanks, the MDLs read 3 times S/N of the lowest spiked standard. OCP MDL varied from 1.31 to 7.30 pg/g while PCB had MDL in the range of 0.40–7.10 pg/g. The % recoveries of recovery standards in dust ranged from 88 to 110 and 99–121 for TCmX and PCB 209, respectively. OCs concentration in the dust is taken on a dry weight (DW) basis. OCs concentration in blank samples was subtracted from the level of target compounds. However, they were not adjusted for recoveries.

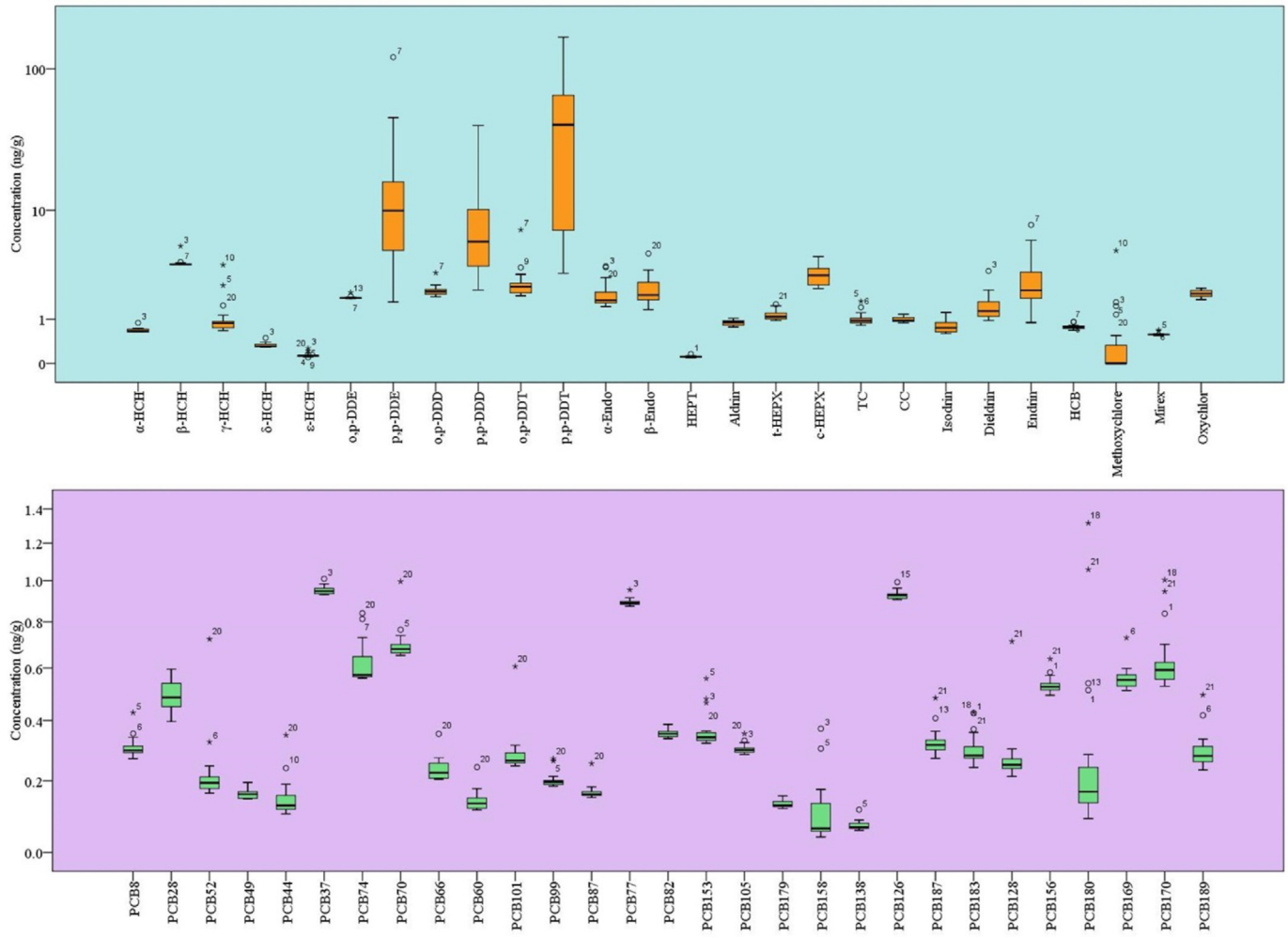

Fig. 1. Box and whiskers plots are showing contamination levels of different OCP and PCB chemicals in dust from Kopawa. Y-axis is in logarithmic scale.

2.4. Determination of TOC and BC

Total organic carbon (TOC) and black carbon (BC) content in dust were obtained from our previous study (Yadav et al., 2019a). The sample pretreatment and analysis of TOC and BC in the dust is detailed somewhere else (Yadav et al., 2019a) and also discussed in SI.

2.5. Statistical analysis

The statistical parameters (minimum, maximum, median, and average) were calculated using MS Excel 2016. The box and whisker's plot and bar diagram were plotted using the Statistical Package for Social Science software (IBN SPSS, version 21). ESRI Arc GIS software was used to draw a spatial distribution map. The non-detected value was replaced with zero for statistical calculation.

2.6. Health risk estimation

Dermal absorption and ingestion of dust are the two significant pathways of OCs exposure to human inside houses. (Alves et al., 2014). While the exposure risk of OCs via dust to the general population is widely reported, data on health risk to adult and children population due to exposure of OCs via dust inside houses in developing countries is limited. In this study, we estimated daily health risk exposure of adults and children individually due to dust ingestion and dermal contact. The dust ingestion and dermal absorption risk due to OCs exposure in dust was determined using USEPA's risk assessment guideline (USEPA, 2011, 2014; Cequier et al., 2014; Yadav et al., 2018). Mathematically, it is expressed as:

$$\text{Dust ingestion} = \frac{CD \times DI}{BW} \quad (1)$$

$$\text{Dermal contact via dust} = \frac{CD \times DAS \times ESA \times AF}{BW} \quad (2)$$

Where CD represents OCs level in the dust (ng/g), Di is everyday dust uptake (60 mg/day in case of children; and 20 mg/day in case of an adult). DAS is a dust sticking rate to the skin (0.01 mg/cm^2); ESA denotes the surface area of skin exposed to OCs (4970 cm^2/day in case of children and 8620 cm^2/day in case of an adult). AF is sorption quantity (0.17%). In the absence of fundamental data on exposure to legacy POPs, 100% assimilation uptake was considered in compliance with the previous study (Jones-Otazo et al., 2005). The average body weight adult (70 Kg) and children (15 Kg) were utilized for estimating the OCs risk. Three exposure situation (low-end, typical, and high-end) was determined based on 5th percentile, median, and 95th percentile estimation.

3. Results and discussion

3.1. Overall summary

The concentration of 26 OCPs and 30 PCBs determined in dust in this study is summarized in Table S2 and S3. Generally, Σ_{26}OCPs in dust was about 7–8 times greater than Σ_{30}PCBs. Σ_{26}OCPs and Σ_{30}PCBs in

dust ranged from 32 to 388 ng/g (median 87 ng/g) and 9.64–16.5 ng/g (median 10.5 ng/g), respectively. This level of Σ_{26}OCP in this investigation is about 2 folds greater than that reported in Pakistan (median 54.7 ng/g) (Ali et al., 2012), but 4–12 folds lower than those reported in Germany (310 ng/g) (Walker et al., 1999) and Romania (1050 ng/g) (Dirtu and Covaci, 2010). ΣOCPs in dust were mostly influenced by DDT, followed by HCHs and endosulfan (Endos), and accounted for 73%, 7% and 4% of Σ_{26}OCPs, respectively. This percentage of OCP in dust in this investigation is consistent with a past study from the urban sites of Nepal (Yadav et al., 2019b). Several studies have also shown a significant contribution of DDTs and HCHs among other chlorinated OCs in indoor dust (Hwang et al., 2008; Brauner et al., 2011; Ali et al., 2012). Among PCB homologous, tetra-CBs was most prevalent, followed by penta-CBs, hexa-CBs, and hepta-CBs, and comprised 28%, 21%, 17% and 17% of Σ_{30}PCBs, respectively (Fig. S1).

3.2. Organochlorine pesticides(OCPs)

A pesticide used inside the house is the same as those used in agriculture and farming. However, pesticides in outdoor environment get decomposed rapidly due to microorganisms' action, hydrolysis, and presence of UV light. The lack of microbial, photolytic, and dissipation activities inside the house can result in the higher persistence of OCP inside the indoor environment (Brauner et al., 2011). The environmental concentration of different classes of OCPs determined in dust has been illustrated in Fig. 1. DDT was identified as the most dominant chemical among different types of OCPs. The concentration of OCP in dust in this study were arranged in decreasing order of DDT > HCH > Endos > CHLs. The abundance of DDT than other OCP could be possibly due to large consumption of DDT in agriculture and health sector. DDT was one of the most popular and effective pesticides among Nepalese farmer before it was banned in 2001. Despite banned for agribusiness, DDT still being used for malaria control.

3.2.1. DDX (DDT, DDD, DDE)

The usage of DDX (sum of *o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, and *p,p′*-DDE) as natural pesticides formulation to control malaria and pests results in contamination of the indoor environment (Butte and Heinzow, 2002). In this investigation, the median concentration of different DDT metabolites measured in the dust is shown in Fig. 1. ΣDDT in dust ranged from 12.4 to 338 ng/g (median 62.9 ng/g). ΣDDT level analyzed in dust in this investigation was matched with past studies reported in Nepal and other location around the world (Table S4). ΣDDT in dust in this study is roughly 2–6 times lower than that discussed in house dust from urban sites of Nepal (Yadav et al., 2019b). This level of ΣDDT in dust is in line with the previous study from Pakistan (53.2 ng/g), but slightly greater than those observed in Canada (46 ng/g) (Audy et al., 2018) and Czech Republic (42 ng/g) (Audy et al., 2018) (Table S4). Moreover, several folds high level of ΣDDT than this investigation has been reported in dust from Romania (1130 ng/g) (Dirtu et al., 2012) and Denmark (1250 ng/g) (Brauner et al., 2011). A relatively high level of ΣDDT and their metabolite than other OCPs in indoor dust suggests the existence of DDT source in the Nepalese environment.

The profile of the DDT and their metabolites in dust has been illustrated in Fig. 2. It is evident from Fig. 2 that *p,p′*-DDT is the main component (65%) to the ΣDDT, while *p,p′*-DDE contributed about 16% of ΣDDT. This profile of DDX is consistent with past studies (Covaci et al., 2003; Dirtu et al., 2012; Yadav et al., 2019b). The *p,p′*-DDT/*p,p′*-DDE ratio is more commonly used to differentiate technical DDT sources from other sources. The *p,p′*-DDT/*p,p′*-DDE ratio greater than 1 indicates the fresh application and minimum degradation, while *p,p′*-DDT/*p,p′*-DDE ratio < 1 specify long-range transport (LRAT) from other regions. In this study, *p,p′*-DDT/*p,p′*-DDE ratio in dust ranged from 0.8 to 7.6 (median 2.89), suggesting fresh application and minimal degradation of DDT in the local environment of Kopawa. Although usages of DDT have been officially banned in Nepal since 2001, the porous border with India allow illegal entry of DDT from India, where large quantities of DDT still being produced (MOEST, 2007). A higher proportion of *p,p′*-DDT/*p,p′*-DDE has been reported in indoor dust (Zhang et al., 2010; Yadav et al., 2019b), and air (Yadav et al., 2017b) previously. The elevated level of DDT in house dust could be also due to their past use, specifically for malaria eradication and pest treatment (Neupane, 1995). The location wise distribution of ΣDDT in dust in this investigation has been shown in Fig. 3 and S3. It is evident from Fig. 3 that the extremely high level of ΣDDT is determined at KPD-7 (323 ng/g). A more significant level of ΣDDT was additionally seen at KPD-13 (234 ng/g) and KPD-9 (171 ng/g). All (KPD-7, -13 and −9) sites are medium-income residential houses. Likewise, a low level of ΣDDT was seen at KPD-21 (12 ng/g) and KPD-18 (14 ng/g). KPD-21 and KPD-18 are low-income, and medium-income residential houses, respectively.

3.2.2. Hexachlorocyclohexane (HCH)

Information on HCH contamination in indoor dust is either unknown or little explored. In this study, all the HCH isomers were detected in all dust samples with 100% detection frequency (DF). The ΣHCH in dust in this study ranged from 5.39 to 10.6 ng/g (median 5.69 ng/g). This level of ΣHCH in this investigation is about 4–5 folds greater than that reported in house dust from Pakistan (1.4 ng/g) (Ali et al., 2012), but consistent with past studies from Canada (7 ng/g) (Audy et al., 2018), Denmark (11 ng/g) (Brauner et al., 2011) and China (8.2 ng/g) (Zhang et al., 2010) (Table S4). This level of ΣHCH is also consistent with ΣHCH level measured in dust sampled from urban houses of Nepal (Yadav et al., 2019b). The profile of the HCH isomers shown in Fig. 2 suggests *β*-HCH as the most dominant isomer, followed by γ-HCH and *α*-HCH, and accounted for 65%, 16% and 12% of ΣHCH, respectively. This profiling pattern of HCH isomer indicated the usage of technical lindane instead of pure lindane. The ΣHCH in dust decreased in the order *β*-HCH > γ-HCH > *α*-HCH > *δ*-HCH > *ε*-HCH.

The isomeric ratio of *α*- and γ-HCH is used to differentiate lindane contamination from commercial HCH sources. The isomeric molecular ratio of *α*- and γ-HCH between 3 and 7 indicates usage of commercial HCH, while α-/γ-HCH ratio close to 1 indicates lindane contamination. In this study, the α-/γ-HCH ranged from 0.18 to 0.98 (median 0.75), well below 1, suggesting HCH source from lindane contamination. The abundance of lindane contamination has also been reported in air and dust samples from Nepal (Yadav et al., 2017b, 2019b). The located graph of HCH showed the highest level of ΣHCH at KPD-10 (8.56 ng/g), KPD-3 (8.07 ng/g), and KPD-5 (7.43 ng/g). The lowest ΣHCH was measured at KPD-18 (5.40 ng/g) and KPD-12 (5.44 ng/g) (Fig. S3). KPD-10 was high-income residential house, while KPD-18 was medium-income residential houses.

3.2.3. Endosulfan

Both *α*- and *β*-isomers of endosulfan were detected in all dust samples. The concentration Σ_2Endos ranged from 2.76 to 8.19 ng/g with a median of 3.61 ng/g (Fig. 1). Σ_2Endos level in dust in this investigation is about 3–6 folds smaller than that observed in house dust from Nepal (10–21 ng/g) (Yadav et al., 2019b). This level of Σ_2Endos in the dust is consistent with that reported in dust from Portugal (3.1 ng/g) (Arnold et al., 2018). Moreover, ΣEndos in dust in this investigation is about 5–6 folds lower than that reported in house dust from the USA (15 ng/g) (Arnold et al., 2018). The profile of the Endos showed a slightly higher proportion of *β*-endosulfan than α-endosulfan and accounted for 53% of Σ_2Endos, while α-endosulfan comprised 47% of Σ_2Endos (Fig. 2). The percentage of *α*- and *β*-endosulfan is utilized to distinguish the past application and influence of the LRAT source (Qu et al., 2015). The *α*-/*β*-endosulfan ratio closed to 2.3 is indicative of the current use of commercial endosulfan, while *α*-/*β*-endosulfan ratio higher than 2.3 implies the influence of LRAT. In this study, the α-/β-endosulfan rate in dust varied from 0.59 to 1.20 (median 0.86), specifying the fresh use of commercial endosulfan. This is true in fact that

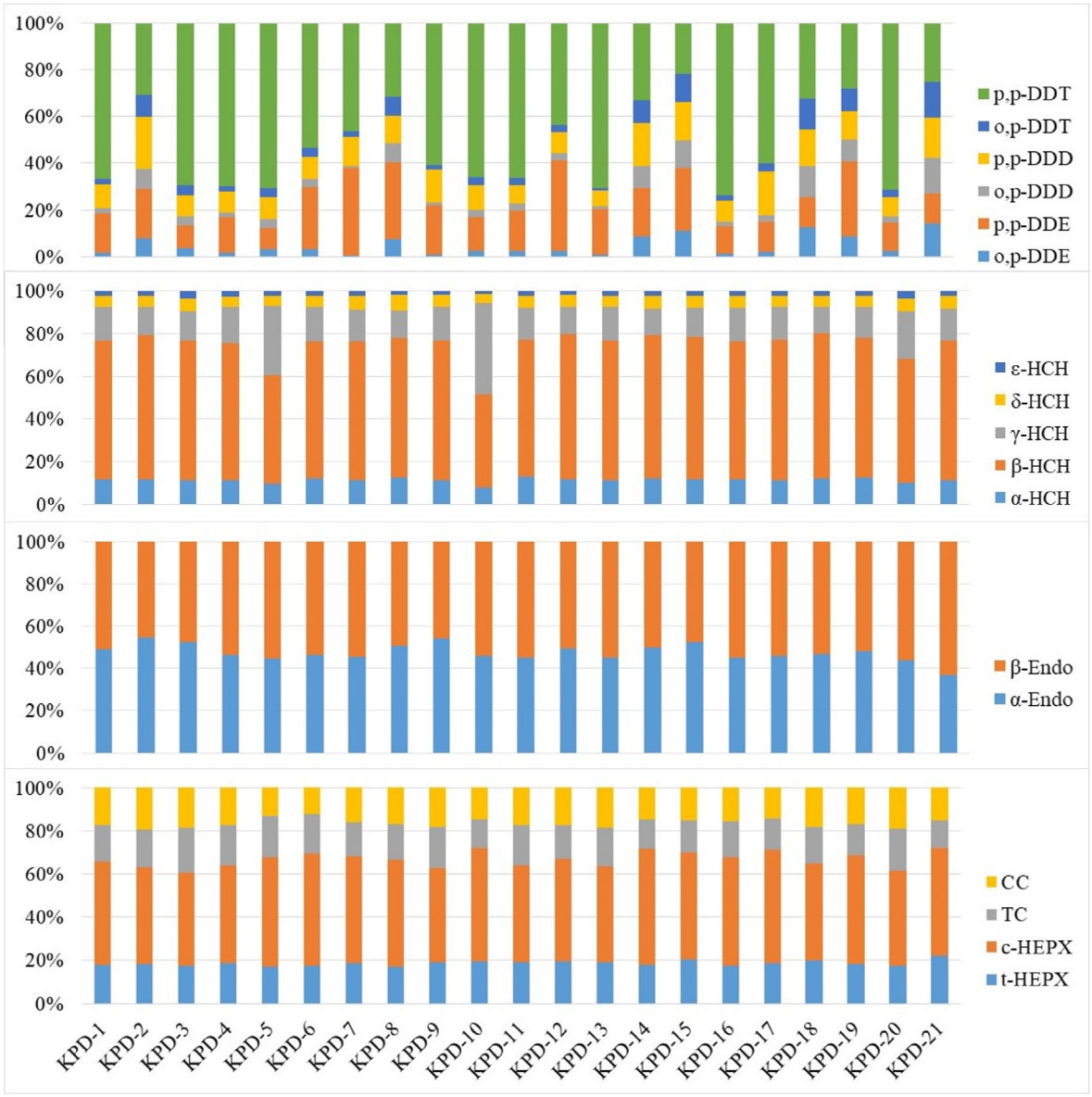

Fig. 2. Profile of the OCP showing contribution of individual chemicals in dust from Kopawa.

large amounts of endosulfan still being used in Nepal in varieties of crops (Yadav et al., 2017b). The spatial distribution of endosulfan is illustrated in Fig. 3 and S3. A more significant level of ΣEndos was determined at KPD-20 (8.19 ng/g), KPD-3 (6.71 ng/g), and KPD-10 (6.16 ng/g), while the low level was measured at KPD-15 (2.76 ng/g) and KPD-14 (2.97 ng/g).

3.2.4. Others OCPs

Different OCPs such as chlordane, HCB, HEPT, HEPX measured in this study had low detection frequencies and concentration in dust samples (Table S2). This result is consistent with our past research where not detectable to a small degree of OCPs was reported in dust from urban sites of Nepal (Yadav et al., 2019b).

3.3. Polychlorinated biphenyls (PCBs)

PCBs are indeed stabilized compound with a half-life of about 20 years (Erickson, 1997). The dissemination of PCB in the indoor condition is delayed because of the absence of moisture, daylight, and microbial exercise, thereby allowing them to be persistent for a longer duration (Hwang et al., 2008). In this study, of 32 targeted, only 30 PCB congeners were detected in the dust. The interquartile range, together with a median level of PCB determined in the dust, has been shown in Fig. 1. The Σ_{30}PCBs level in dust ranged from 9.64 to 16.5 ng/g (median 10.5 ng/g), which is about ten times higher than ΣPBDEs reported earlier from the same study sites (Yadav et al., 2019a). Also, the Σ_{30}PCBs in dust in this study are about 5 times to several orders of magnitude lower than ΣNBFRs and ΣOPFRs reported previously from the same sites, respectively (Yadav et al., 2019a). This level of Σ_{30}PCB is consistent with ΣPCB level seen indoor dust from Nepal (7–11 ng/g) (Yadav et al., 2019b) and Singapore (6.0 ng/g) (Tan et al., 2007). Σ_{30}PCBs measured in dust in this investigation is multi times smaller than those reported in dust from USA (37 ng/g) (Audy et al., 2018), Portugal (98 ng/g) (Arnold et al., 2018) and Romania (35 ng/g) (Dirtu et al., 2012), but roughly 15–16 folds greater than that reported in indoor dust from Pakistan (0.67 ng/g) (Ali et al., 2012).

Dioxin like-PCB (DL-PCBs) (sum of PCB-77, -105, −126, −156, −169, and −189) in dust in this investigation was about 2 times higher than the indicator-PCB (Ind-PCBs) (sum of PCB-28, -52, −101, −153, −138, and −180) (Table S3). A more significant level of DL-PCBs than

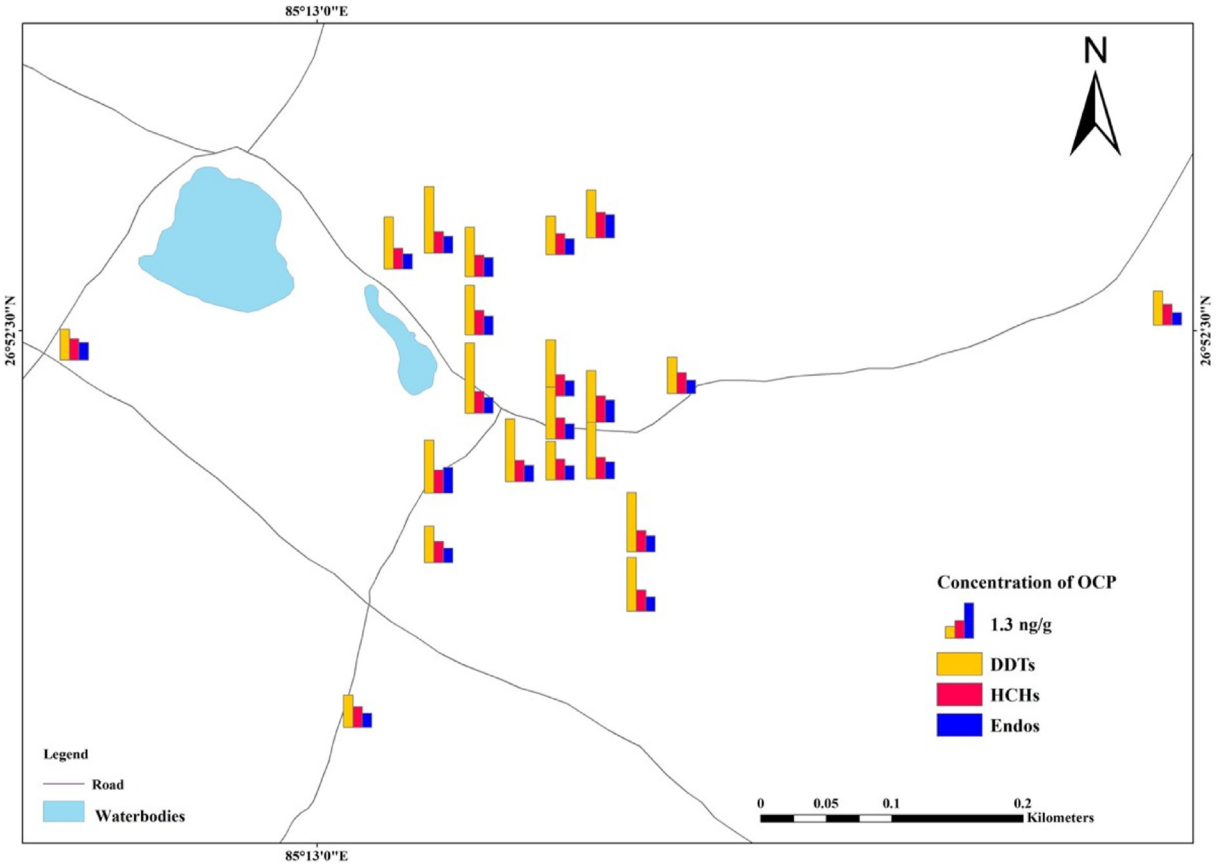

Fig. 3. Spatial distribution of OCP chemicals (ng/g) in indoor dust from Kopawa. PCB concentration is in logarithmic scale.

Ind-PCBs has also been reported previously in house dust and surface soil from Nepal (Yadav et al., 2017b, 2019b). ΣDL-PCBs and ΣInd-PCBs in dust ranged from 3.35 to 3.82 ng/g (median 3.48 ng/g) and 1.35–2.67 ng/g (median 1.63 ng/g), respectively (Table S3). The relative abundance of ΣPCBs measured in dust has been shown in Fig. 4 and **S4.** Individually, PCB-37 was recognized as the most prevalent PCB congeners, trailed by PCB-126 and PCB-77, and accounted for 9.1%, 8.9%, and 8.5% of Σ_{30}PCB, respectively. This profile of PCBs is consistent with the PCB profile reported in indoor dust from Nepal (Yadav et al., 2019b). The abundance of PCB-126 was also seen in indoor dust from Nepal, and workshop station in China (Xing et al., 2011). PCBs are used as joint sealant, paints, varnishes, cable insulation, and plasters in building to make more effective and longevity (ATSDR, 2000), thus there is severe chance of indoor contamination with PCBs released from these structure materials, through volatilization and consequent adsorption in house dust (Hwang et al., 2008).

The spatial distribution of ΣPCBs, DL-PCBs, and Ind-PCBs in dust in this investigation have been illustrated in Fig. 5. Spatially, Σ_{30}PCB, ΣDL-PCBs, and ΣInd-PCBs in dust varied little from one house to another house. The highest degree of ΣPCBs was detected at KPD-20 (13.2 ng/g), while the least concentration was measured at KPD-12 (10 ng/g) (Fig. S5).

3.4. Comparison of legacy POPs with other OCs

In order to examine the difference between contamination level of legacy POPs with those still being used or as of late been restricted, dust sample used in this study were also analyzed for polybrominated diphenyl ethers (PBDEs), dechlorane plus (DPs), novel brominated flame retardants (NBFRs) and organophosphate esters (OPEs) (Yadav et al., 2019a). ΣOPEs showed the highest concentration by order of magnitude (1300 ng/g), followed by ΣOCPs (86.7 ng/g) and ΣNBFRs (51.4 ng/g) (Fig. 6). This implies a more significant application of OPEs in consumer materials and building products where they are included in a higher rate than brominated flame retardants. However, ΣOCPs and ΣPCBs were second and fourth to those of ΣOPEs, individually. A similar proportion of legacy POPs and other recently used OCs has been reported earlier in England (Harrad et al., 2006), Japan (Takigami et al., 2009), Czech Republic, and Canada (Audy et al., 2018). ΣNBFR concentration was about 5 times greater than ΣPCBs and about 2 times lower than ΣOCPs. The high degree of legacy POPs in indoor suggests their ecological persistence or ongoing use of POPs in building materials (in case of PCBs) or due to intentional use. Dust residual cleaning and structural ventilation system can hardly decrease these chemicals from the indoor condition. The elevated level of legacy POPs also highlights the ineffective management of these chemicals in Nepal. It is realistic to note the inefficient management of chemicals even 15 years after the implementation of the Stockholm Convention, the “dirty dozen” POPs continue to be detected in the indoor environment.

Relatively low levels of OCs (PCBs and OCPs) in this investigation than from more developed nations suggest their restricted use in the rural area of Nepal. This could be due to the open way of housing and dusting habits in the rural area of Nepal. People sweep and mop their houses once or twice daily, thereby removing settled dust particles from the indoor surface. Moreover, low levels of OCs could be due to the fast degradation of and evaporation of target chemicals due to hot high temperatures and more extended sunlight (Davis and Stapleton, 2009; Stapleton, 2006; Zeng et al., 2008).

3.5. Linkage of TOC, BC with OCPs/PCBs

A Spearman rank's correlation coefficient analysis was performed using IBN SPSS software over the full dust data set of OCP and PCB to establish the linkage of TOC and BC with that of OCP and PCB. Result

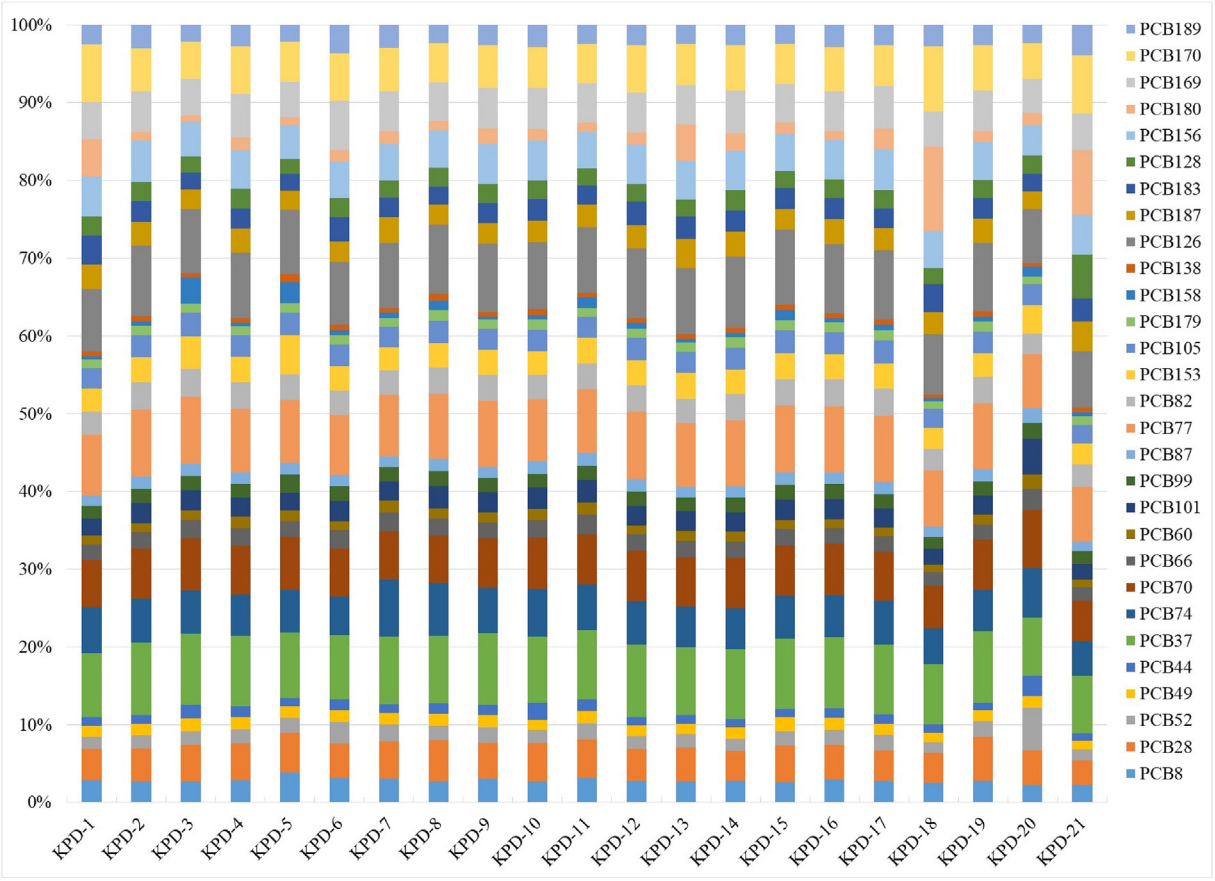

Fig. 4. Relative abundance of individual PCB to ΣPCBs in dust.

showed that only DDT metabolites (*o,p′*-DDT, *p,p′*-DDT, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDE, and *p,p′*-DDE) among OCPs were moderately and positively linked with TOC (*Rho* = 0.45, 0.49, 0.51, 0.55, 0.53, and 0.46, $p < 0.05$) but not BC (Table S5). This suggests practically zero impact of TOC on the fate of OCP in the dust. None other OCPs were linked with either TOC or BC. For PCBs, except PCB-126, neither of PCB congeners were linked with TOC/BC (Table S6). PCB-126 was moderately and negatively correlated with TOC (*Rho* = −0.49, $p < 0.05$), but not BC. Weak linkage of BC with OCPs and PCBs in dust could be due to the low level of BC in the dust. A similar finding was reported earlier (Yadav et al., 2019a). TOC contains only 4% of the BC. Hence, a low BC/TOC ratio may inhibit the sorption of BC because of natural attenuation (Nam et al., 2008).

Additionally, an interrelationship of OCP and PCB chemicals were investigated in the dust using spearman's rank correlation coefficient. DDT metabolites were strongly and positively linked with each other, suggesting similar sources in the dust. *β*- and γ-HCHs were also moderately to strongly connected with DDT metabolites. Endosulfan showed a moderate to strong correlation with different HCH isomers (Table S5). For PCBs, only high molecular weight PCB congeners were interlinked with each other. DL-PCBs, Ind-PCBs, and ΣPCBs were also moderately to strongly interrelated in the dust.

3.6. Health risk exposure

Even though indoor dust has been known as one of the significant sources of human exposure to different types of OC chemicals, the exposure fate of OCPs and PCBs has been little known in indoor dust (Yadav et al., 2019a). In this investigation, the daily health exposure of OCPs and PCBs to adult and children population through dust ingestion and dermal absorption via dust were estimated and presented in Table 1. The result indicated that dermal absorption via dust as the primary routes of OCs exposure. Children were more vulnerable to OCs exposure than the adult population. The daily exposure risk (both intake and dermal contact) of PCBs was lower than OCPs, indicating a relatively minimal risk of PCB. *p,p′*-DDT, and *β*-HCH showed relatively higher exposure risk than other OCPs. The estimated daily exposure risk (ingestion and dermal contact) of PCBs (0.5046 ng/kg/BW/day and 0.7106 ng/kg/BW/day) and OCPs (10.0145 ng/kg/BW/day and 14.1021 ng/kg/BW/day) for children in this study were consistent with previous health risk studies from urban house dust of Nepal (0.1641 ng/kg/BW/day and 2.3111 ng/kg/BW/day for PCBs and 6.9190 ng/kg/BW/day and 97.4312 ng/kg/BW/day for OCPs) (Yadav et al., 2019a). Moreover, this result is also in line with previous studies around the world (Harrad et al., 2006; Wang et al., 2010; Ali et al., 2013; Dirtu et al., 2012). The typical (median) and worst-case scenario (95th percentile) intake of OCP/PCBs were about 2–4 orders of magnitude lower than their respective reference dose (RfD), indicating insignificant risk (Table 1). However, other means of OCs exposure such as dietary exposure may still be significant in case of rural Nepal which need to be further investigated as95% of the consumption of chemicals can mainly enter through food ingestion (Yu et al., 2012). .

4. Conclusions

In this study, the environmental concentration of OCPs and PCBs was investigated in indoor dust from a rural village (Kopawa) of Nepal to understand the fate and behavior of OCs. The result showed that OCP contamination was more prominent than PCBs. ΣOCPs level determined in dust was about 7–8 times greater than ΣPCBs. DDT was identified as the most abundant chemical among OCPs, while PCB-37 exceeded among PCBs. Relatively high levels of ΣDDT than other OCPs in indoor

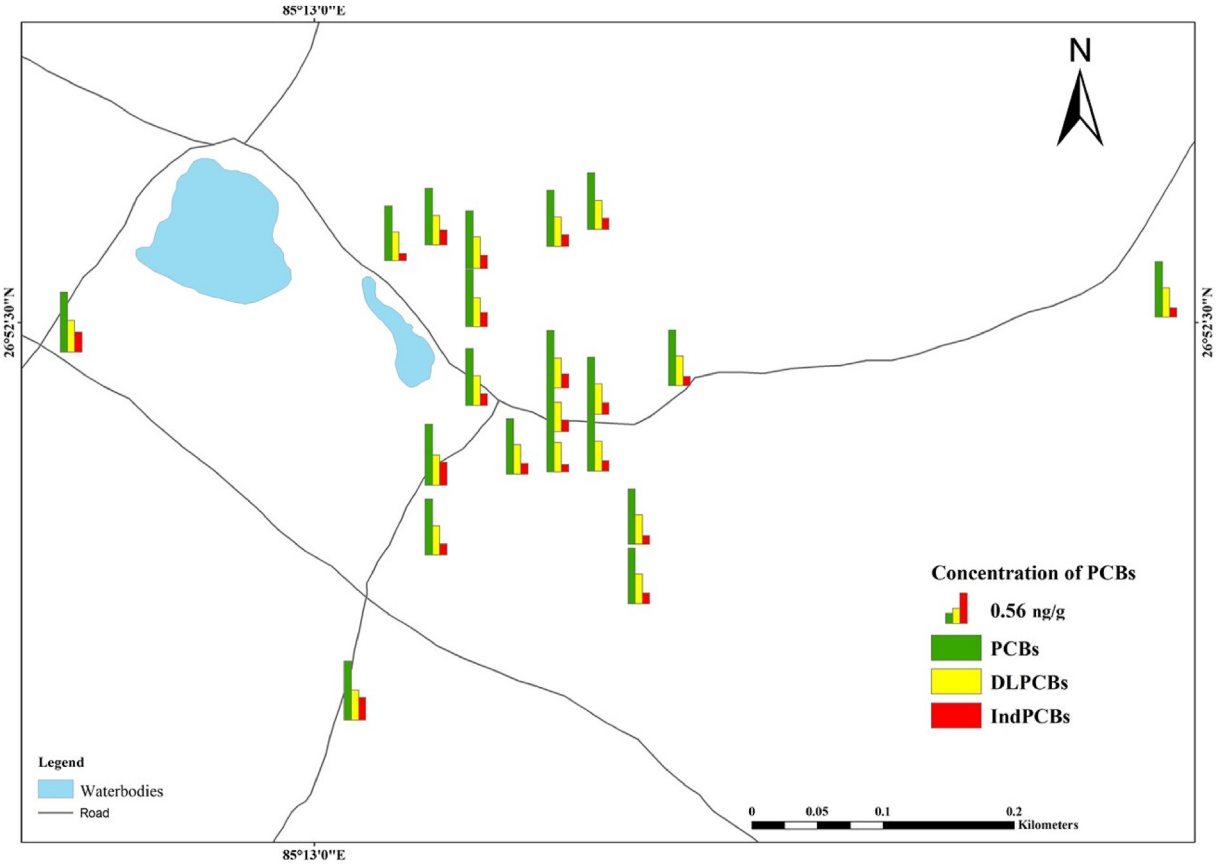

Fig. 5. Spatial distribution of ΣPCB, DL-PCB, and Ind-PCB(ng/g) in dust from Kopawa. PCB concentration is in logarithmic scale.

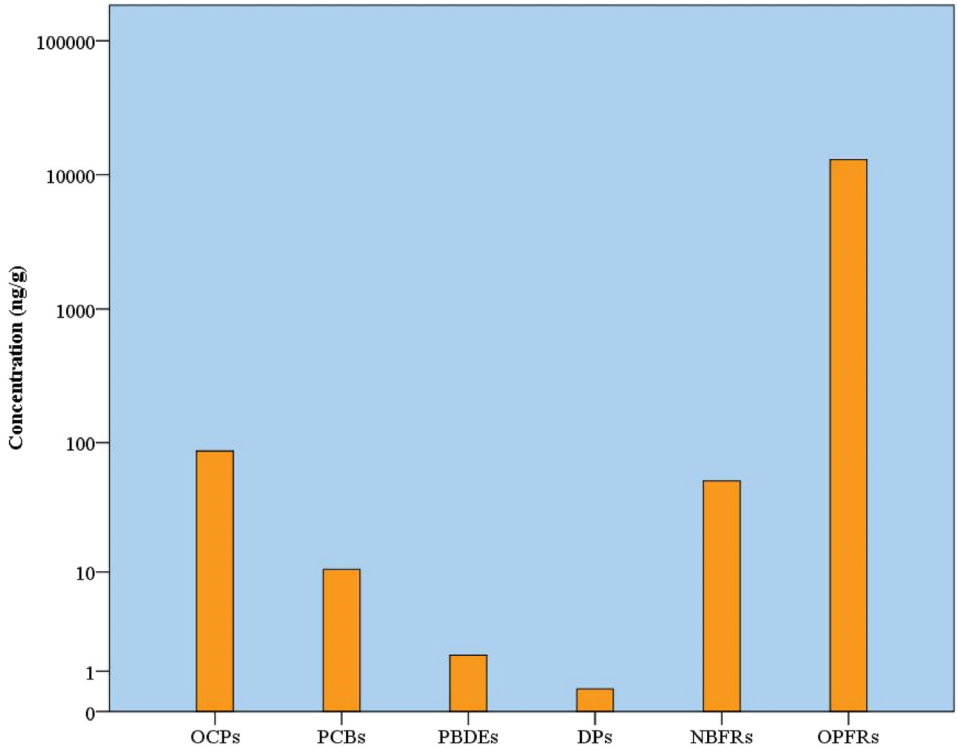

Fig. 6. Comparison of the median ΣOCP and ΣPCB in dust in this study with other POPs. The concentration of PBDEs, DPs, NBFRs and OPFRs were obtained from Yadav et al. (2019a).

Table 1
Daily exposure risk of OCP and PCB (ng/kg BW/day) to adult and children population via dust ingestion and dermal contact.

Adult	Dust ingestion			Dermal absorption			RfD
	5th percentile	Median	95th percentile	5th percentile	Median	95th percentile	
α-HCH	0.0018	0.0019	0.0021	0.0134	0.0137	0.0155	300[a]
β-HCH	0.0106	0.0106	0.0111	0.0773	0.0778	0.0817	300[a]
γ-HCH	0.0020	0.0025	0.0069	0.0147	0.0185	0.0503	300a
δ-HCH	0.0008	0.0009	0.0012	0.0060	0.0066	0.0084	300[a]
ε-HCH	0.0003	0.0004	0.0006	0.0022	0.0026	0.0047	300[a]
o,p-DDE	0.0050	0.0051	0.0053	0.0369	0.0375	0.0390	500[a]
p,p-DDE	0.0053	0.0284	0.1318	0.0388	0.2079	0.9654	500[a]
o,p-DDD	0.0053	0.0060	0.0069	0.0389	0.0437	0.0507	500[a]
p,p-DDD	0.0064	0.0164	0.0680	0.0466	0.1201	0.4985	500[a]
o,p-DDT	0.0055	0.0066	0.0100	0.0401	0.0487	0.0733	500[a]
p,p-DDT	0.0101	0.1173	0.4272	0.0741	0.8593	3.1298	500[a]
α-Endo	0.0042	0.0048	0.0100	0.0308	0.0354	0.0734	
β-Endo	0.0042	0.0055	0.0095	0.0306	0.0403	0.0696	
CHLs	0.0050	0.0056	0.0067	0.0368	0.0407	0.0495	
HEPX	0.0094	0.0118	0.0153	0.0686	0.0865	0.1122	
HCB	0.0019	0.0022	0.0026	0.0143	0.0158	0.0192	800[a]
ΣInd-PCBs	0.0039	0.0047	0.0074	0.0288	0.0341	0.0539	
ΣDL-PCBs	0.0098	0.0099	0.0109	0.0716	0.0729	0.0797	
ΣPCBs	0.0288	0.0310	0.0360	0.2109	0.2269	0.2641	1000[b]
Children							
α-HCH	0.0256	0.0262	0.0295	0.0360	0.0369	0.0416	300[a]
β-HCH	0.1477	0.1487	0.1561	0.2080	0.2093	0.2198	300[a]
γ-HCH	0.0282	0.0353	0.0962	0.0396	0.0497	0.1354	300a
δ-HCH	0.0115	0.0125	0.0161	0.0163	0.0176	0.0227	300[a]
ε-HCH	0.0042	0.0050	0.0089	0.0060	0.0071	0.0126	300[a]
o,p-DDE	0.0705	0.0716	0.0745	0.0992	0.1008	0.1050	500[a]
p,p-DDE	0.0742	0.3973	1.8446	0.1045	0.5595	2.5975	500[a]
o,p-DDD	0.0743	0.0835	0.0969	0.1047	0.1176	0.1365	500[a]
p,p-DDD	0.0891	0.2295	0.9525	0.1254	0.3232	1.3413	500[a]
o,p-DDT	0.0765	0.0931	0.1400	0.1078	0.1311	0.1971	500[a]
p,p-DDT	0.1415	1.6419	5.9802	0.1993	2.3121	8.4211	500[a]
α-Endo	0.0589	0.0676	0.1403	0.0830	0.0952	0.1975	
β-Endo	0.0585	0.0769	0.1330	0.0824	0.1083	0.1873	
CHLs	0.0703	0.0778	0.0945	0.0990	0.1096	0.1331	
HEPX	0.1311	0.1653	0.2144	0.1845	0.2328	0.3019	
HCB	0.0273	0.0302	0.0368	0.0384	0.0426	0.0518	800[a]
ΣInd-PCBs	0.0549	0.0651	0.1030	0.0774	0.0917	0.1450	
ΣDL-PCBs	0.1369	0.1393	0.1523	0.1927	0.1961	0.2145	
ΣPCBs	0.4030	0.4335	0.5046	0.5675	0.6104	0.7106	1000[b]

[a] Dirtu et al., 2012.
[b] Oostam et al., 1999

dust suggest the existence of DDT source in the Nepalese environment. A higher ratio of *p,p*-DDT, and *p,p*-DDE proposed fresh application and minimal degradation of DDT in the local climate of Kopawa. While low α-/γ-HCH recommends lindane contamination as the primary source of HCH. Likewise, the low α-/β-endosulfan ratio in dust specified the recent use of commercial endosulfan. Only DDT among OCPs and PCB -126 among PCBs, showed moderate correlation with TOC but not BC, recommending little or no influence of TOC/BC in the fate of OCs in the dust. The estimated daily health risk exposure showed that children were more vulnerable to OCs than the adult population. The estimated daily ingestion and dermal absorption of OCs were about 2–4 orders of magnitude lower than their respective oral reference dose (RfD), suggesting minimal risk. However, other routes of exposure, for instance, food intake still may be important which needs further investigation.

CRediT authorship contribution statement

Ishwar Chandra Yadav: Formal analysis, Writing - review & editing. **Ningombam Linthoingambi Devi:** Software, Writing - review & editing. **Jun Li:** Methodology. **Gan Zhang:** Methodology.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

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