Possible emissions of POPs in plain and hilly areas of Nepal: implications for source apportionment and health risk assessment

Ishwar Chandra Yadav [a,*], Ningombam Linthoingambi Devi[b], Jun Li[a], Gan Zhang[a], Knut Breivik[c,d]

[a]*State Key Laboratory of Organic Geochemistry, Guangzhou Institute of Geochemistry, Chinese Academy of Sciences, Guangzhou- 510640, China*

[b]*Centre for Environmental Sciences, Central University of South Bihar, BIT Campus, Patna-800014, Bihar, India*

[c]*Norwegian Institute for Air Research, Box 100, NO-2027 Kjeller, Norway*

[d]*University of Oslo, Department of Chemistry, Box 1033, NO-0315 Oslo, Norway*

*Corresponding author

Tel. no. +86-15626134294

E-mail: icyadav.bhu@gmail.com (I C Yadav)

Capsule: POPs in Nepal; source apportionment and health risk assessment

Abstract

Ambient air is a core media chosen for monitoring under the Stockholm Convention on POPs. While extensive monitoring of POPs in ambient air has been carried out in some parts of the globe, there are still regions with very limited information available, such as some developing countries as Nepal. This study therefore aims to target the occurrence of selected POPs in Nepal in suspected source areas / more densely populated regions. Four potential source regions in Nepal were furthermore targeted as it was hypothesized that urban areas at lower altitudes (Birgunj and Biratnagar located at approximately 86 and 80 m.a.s.l.) would be potentially more affected by OCPs because of more intensive agricultural activities in comparison to urban areas at higher altitudes (Kathmandu, Pokhara located 1400 and 1135 m.a.s.l). As some of these areas could also be impacted by LRAT, air mass back trajectories during the sampling period were additionally evaluated using HYSPLIT. The concentrations of overall POPs were twice as high in plain areas in comparison to hilly areas. DDTs and HCHs were most frequently detected in the air samples. The high *p,p′*-DDT/(*pp′*-DDE+ *pp′*-DDD) ratio as well as the low *o,p′*-DDT/*p,p′*-DDT ratio observed in this study was inferred as continuing use of technical DDT. High levels of $\sum_{26}$PCBs were linked to proximity to highly urbanized and industrial areas, indicating the potential source of PCBs. The measured concentrations of legacy POPs in air from this study is assumed to represent a negligible health risk through inhalation of ambient air, however, other modes of human exposure could still be relevant in Nepal. The air mass backward trajectory analysis revealed that most of the air masses sampled originated from India and the Bay of Bengal.

Keywords: legacy POPs**;** Nepal; Stockholm convention; lindane; technical DDT

1. Introduction

Persistent organic pollutants (POPs) including organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs), are a group of highly hazardous chemical contaminants which may pose a risk to environmental and human health (Hoff et al., 1992). Both OCPs and PCBs have similar physicochemical characteristic in terms of persistence and tend to bio-accumulate in fatty tissues within living organisms (Jones and de Voogt, 1999). Evidence suggests atmospheric long range transport of these chemicals into remote regions where they have neither been used nor produced (Kallenborn et al., 1998).

In the past several decades, POPs have been released into the environment and are now widely distributed in different environmental matrices due to their persistence, bio-accumulative and long-range transport behaviors. The Stockholm Convention on POPs to which more than 150 countries are members at present, calls for parties to identify the source of POPs (e.g. OCPs and PCBs) and to monitor these chemicals in environment. Under this agreement, signed in 2001, every party is required to develop their own national implementation plan on POPs (Weinberg, 2008). While some countries have already collected information on historical usages and developed national inventories for selected POPs, such data are not always readily available or even known in other countries. Monitoring of these chemical in diverse environmental matrices represent a complementary approach to help trace the sources and fate of these chemicals (Du et al., 2009; Liu et al., 2009; Zheng et al., 2010; Yadav et al., 2015). PCBs are industrial chemicals which were extensively produced during the last century (e.g. for electrical equipment), as well as a byproduct of various industrial processes. High levels of PCBs have been detected in industrialized and urban areas (Jamshidi et al., 2007; Du et al., 2009). Dichlorodiphenyl trichloroethane (DDT), hexachlorocyclohexane (HCH), aldrin, dieldrin, endrin, heptachlor, chlordane, toxaphene, and

mirex are often collectively referred to as organochlorine pesticides (OCPs). These OCPs are among the "dirty dozen" that were initially regulated under the Stockholm Convention on POPs because of environmental and human health concerns (Yadav et al., 2015). Nepal is among some developing countries, where individual OCPs may still be used in spite of international regulations (Yadav et al., 2016).

The atmospheric transport pathway plays a significant role in movement of POPs. It is the most effective medium for regional and global distribution (Wang Y et al., 2015). The potential for atmospheric transport and deposition of POPs is dictated by various meteorological parameters (eg., temperature, precipitation, wind speed, and wind direction), fate properties of individual POPs, as well as spatial and temporal variability in emissions (Reisen and Arey, 2005; Tsapakis and Stephanou, 2007; Ohura et al., 2013). Ambient air monitoring and subsequent data analysis is the best available technique for assessing atmospheric burdens of POPs released from primary as well as secondary sources (Wang Y et al., 2015). OCPs and PCBs are synthetic compounds and have great chemical stability. Because of their wide application throughout the world, they are ubiquitous in the environment and pose significant environment and human risk via ingestion, inhalation and dermal contact (Jones and Voogt, 1999; Ben et al., 2012). Some of these pollutants are highly toxic and have a large variety of chronic effects, including endocrine dysfunction, mutagenesis and carcinogenesis (Wang et al., 2011).

Nepal Himalaya is the highest part of the Himalayan mountain range in South Central Asia. It occupies most of Nepal including Central, Eastern and Southern Region (Encyclopedia Britannica, 2016).Nepal is primarily an agrarian country, of which more than 66% of total populations are engaged in agricultural activities (DOA, 2014; Neupane et al., 2014). This attributes to 39% of the national gross domestic product. Comparatively, pesticides are mostly

used in plain areas (also called Terai) because of higher % share of cropland in the country (Pariyar, 2008). Nepalese farmer usage disproportionately amounts of pesticides to avoid pest attacks in their crops. There are reports of increasing use of pesticides in Nepal (Palikhe, 2002; Yadav et al., 2016). Use of older, non-patented, more toxic, environmentally persistent and inexpensive chemicals are widely practiced in Nepal (Ecobichon, 2001). However, very few studies report data on concentrations of POPs in air from Nepal. Secondly, 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 (Aichner et al., 2007; Gong et al., 2014; Guzzela et al., 2016). This study therefore aims to target the occurrence and possible atmospheric sources and source regions of selected POPs in Nepal in suspected source areas / more densely populated regions. Our primary interest was on legacy POPs which could still be emitted from contemporary sources in Nepal, such as PCBs and various OCPs, and for which passive air samplers are suitable as sampling media. Four potential source regions in Nepal were furthermore targeted as it was hypothesized that urban areas at lower altitudes (Birgunj and Biratnagar located at approximately 86 and 80 m.a.s.l., respectively) would be potentially more affected by OCPs because of more intensive agricultural activities in comparison to urban areas at higher altitudes (Kathmandu, Pokhara located 1400 and 1135 m.a.s.l, respectively). As some of these areas could also be impacted by LRAT, air mass back trajectories during the sampling period were additionally evaluated using HYSPLIT.

2. Materials and Methods

Detailed descriptions of the materials and methods are described in Supplementary Information.

2.1. Air sampling

Four major cities i.e. Kathmandu, Pokhara, Birgunj and Biratnagar of Nepal were selected for collection of atmospheric samples (**Fig. S1, Table S1**, Supplementary information).A total of 34 polyurethane foam passive air sampling (PUF-PAS) samplers (8 each in Pokhara, Birgunj, Biratnagar and 10 in Kathmandu) were deployed at different locations in open areas within each city and >3m above the ground. After exposure to sufficient duration, all PUF disk were retrieved, resealed and transported to the laboratory where it was and stored at −20 °C till analysis. The details about design and deployment of PUF-PAS sampler has been described elsewhere (Shoeib and Harner 2002; Jaward et al., 2005).

2.2. Extraction and GC-MS analysis

All the 34 PUFs were spiked with known concentration of TCmX and PCB209 as surrogate standards and were soxhlet extracted for 24 h with DCM as solvent. The extract was concentrated to 2-3mL (approx.) by a rotary evaporator after extraction. Then the extract was cleaned by multilayer silica gel/ alumina column followed by anhydrous sodium sulfate. About 30-40ml mixture of DCM/hexane (1:1volume) was used to elute the column and concentrated to 0.2mL under gentle nitrogen stream. About 25µl of Dodecane was added to the GC vial as solvent keeper. A known amount of PCB-54 and ^{13}C-PCB141 were added as internal standards for OCPs and PCBs, respectively, before GC-MS analysis.

The eluted samples were injected in to an Agilent 7890A GC coupled with an Agilent 7000A 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,

146 methoxychlor, mirex and 26 out of 32 PCBs congeners were quantified using an Agilent 7890GC-
147 7000A triple quadrupole mass spectrometer equipped with a CP-Sil 8CB capillary column (50 m
148 ×0.25 mm × 0.25 µm). The GC-MS parameters of individual OCP and PCB are given in **Table**
149 **S2, (**Supplementary Information**).**

150 *2.3. QA/QC*

151 A set of calibration standards were run after every ten samples to check any interference
152 and cross contamination. Field, procedural and solvent blank were analyzed in the same manner
153 as the real samples. The method detection limits (MDLs) for both OCPs and PCBs was 3:1 signal
154 versus noise value (S/N). The average surrogate recoveries in all samples for TCmX and PCB 209
155 were in the range of 72±11 to 80±15%. The concentration of OCPs and PCBs were blank corrected
156 but not corrected for recoveries and expressed on a dry weight basis (amount per sample).

157 *2.4. Statistical Analysis*

158 Descriptive statistics were performed using IBM SPSS statistics (version 21). Arc GIS
159 were used to draw spatial distribution maps. Samples below detection limits were set as zero in
160 the numerical analysis.

161 *2.5. Backward trajectory analysis*

162 A 5-days backward trajectory analysis was performed for each city in order to understand
163 the transport of POPs from potential sources region. The National
164 Oceanic and Atmospheric Administration's (NOAA's) Hybrid-Single Particle Lagrangian
165 Integrated Trajectory (HYSPLIT) model available from National Air Resource Laboratory, USA
166 was used to carryout backward trajectory analysis (http:// www.arl.noaa.gov/ready/hysplit4.html).
167 Meteorological data were obtained from the online database of NOAA (NCEP/NCAR Global
168 Reanalysis data). Backward trajectories at 500 m above ground level were calculated during Sept-

Oct, 2014 for every 00:00, 06:00, 12:00 and 18:00 UTC. A height of 500m from ground level is considered standard elevation which significantly influences the investigation of well-mixed convective boundary layer for regional transport (Jin et al., 2013). HYSPLIT cluster analysis was used to elucidate different patterns of air mass arriving at all the four cities.

2.6. Human health risk assessment

The inhalation exposure and potential health risk to general population due to atmospheric contamination of OCPs and PCBs were estimated using equation 1, 2, and 3, that were adopted from US environmental protection agency (EPA) (USEPA,1998; 2009). As per US EPA (1998), inhalation exposure of contaminant through air can be estimated as:-

$$Exp_{inh} = \frac{C_{air} \times IR \times EF}{BW \times 365} \quad \text{..... (1)}$$

Where, Exp_{inh} is the inhalation of contaminants through air (pg/kg/day), C_{air} is air concentration (pg/m^3), IR is inhalation rate (20m^3/day), EF is exposure frequency (350days/year), BW is body weight (70 kg).After exposure calculation, the carcinogenic and non-carcinogenic risk will be estimated. According to new intake methodology, the amount of contaminants that reaches the target sites through inhalation is directly related to exposure concentration (US-EPA, 2009). Hence, the risk can be estimated as follows:-

$$EC_{inh} = \frac{C_{air} \times ET \times EF \times ED}{AT \times 365 \times 24} \quad \text{..... (2)}$$

$$Cancer\ risk_{inh} = EC_{inh} \times IUR \quad \text{..... (3)}$$

Where, EC is exposure concentration (µg/m^3), C air is the air concentration (µg/m^3), ET is exposure time (24h/day), EF is exposure frequency (350days/year), ED is exposure duration (30

193 years), and AT is average time (70 and 30 years for carcinogenic and non-carcinogenic substances,
194 respectively. IUR is the inhalation unit risk $(\mu g/m^3)^{-1}$). An upper exposure limit of contaminant
195 (also known as carcinogenic benchmark level) is an exposure that poses an upper-bound lifetime
196 excess cancer risk. The exposure of contaminants which exceed the risk factor of 1×10^{-6} (one in 1
197 million people) is taken as significant score for assessing human cancer risk (EPA, 2003).

198 **3. Results and discussions**

199 *3.1. Introductory remarks*

200 As Kathmandu and Pokhara are hill areas, whereas Birgunj and Biratnagar are plain areas
201 bordering to India, the comparison of level and profile of POPs may provide insights into possible
202 altitudinal differences in primary source areas of Nepal. For example, climatic differences may
203 affect the potential for atmospheric emissions as most POPs are semi-volatile organic
204 contaminants. Hill areas are furthermore considered as more pristine regions compared to plain
205 areas in relation to POPs contamination, due to their distance from pesticide sources. Due to
206 differences in altitudinal and climatic variation in these two areas, we aspire to know the possible
207 influence of primary emission from plain areas for the atmospheric burden in hill areas. Further,
208 the level of POPs emission in selected cities of Nepal could be achieved by comparing the
209 atmospheric level of POPs contaminations around the globe.

210 *3.2. General comments on POPs concentration in air*

211 Concentrations of OCPs and PCBs measured in air (ng/PAS) at each site are presented in
212 **Table S3 and S4** (Supplementary Information). For comparison purpose with previous studies, a
213 default uptake rate of 3.5 m^3/day was used to convert the concentration of chemicals held in PAS
214 into volumetric air concentration (pg/m^3). The overall concentration of POPs (OCPs + PCBs) in

Birgunj (average 1307 pg/m^3) and Biratnagar (average 2023 pg/m^3) were about twice as high as those measured in Kathmandu (913 pg/m^3) and Pokhara (724 pg/m^3). The average concentration of HCHs (sum of α-HCH, β-HCH, γ-HCH, δ-HCH and ε-HCH), DDTs (sum of *o,p′*-DDE, *p,p′*-DDE, *o,p′*-DDD, *p,p′*-DDD, *o,p′*-DDT and *p,p′*-DDT), endosulfans (sum of α- and β-endos), HEPT, heptachlor epoxide (HEPX) , trans-chlordane (TC), cis-chlordane (CC), aldrin, dieldrin, isodrin, endrin, HCB, methoxychlor and mirex is presented in **Table 1**. Twenty six different congeners of PCBs (PCB-8, -28, -37, -44, -49, -52, -60, -66, -70, -74, -77, -87, -99, -101, -105, -118, -126, -153, -156, -158, -169, -170, -180, -183, -187, and -189) were also detected in air samples and are given in **Table 2**.

DDTs (647±610) and HCHs (201±258) were the most abundant OCPs detected in the air samples (**Fig. 1**). Specifically, the level of DDTs observed in Biratnagar and Birgunj were much higher than in Japan and South Korea (Jaward et al., 2005), but comparable to urban cities of China (eg., Guangzhou and Zhaoqing) (Ling et al., 2011) (**Table S5,** Supplementary Information). Elevated concentrations of DDT have also been detected in Indian cities (eg., New Delhi, Kolkata, Chennai and Bangalore) (Chakraborty et al., 2010), and were attributed to the ongoing use of DDTs in the agricultural areas and for malaria control (Zhang et al., 2008). The level of HCHs detected in Kathmandu is comparable to Azerbaijan and most Indian cities (eg., New Delhi, Kolkata, Mumbai and Chennai) (Zhang et al., 2008; Chakraborty et al., 2010; Aliyeva et al., 2012), but much higher than in Vietnam and Pakistan (Nasir et al., 2014; Wang et al., 2016).The overall concentration of endosulfans in Biratnagar is much lower than the urban center of Pakistan (eg., Karachi and Lahore) (Nasir et al., 2014) and China (eg., Wuhan) (Qu et al., 2015), but comparable to Vietnam (Wang et al., 2016). The heptachlor and its metabolite were least detected among all OCP chemicals in Nepal, however the levels were comparable with urban area of Wuhan, China

238 (Liu et al., 2009). Regarding PCBs, the average concentration level in Kathmandu and Birgunj are
239 higher than Pakistan and Azerbaijan (Aliyeva et al., 2012; Nasir et al., 2014), whereas they are
240 comparable to those reported in urban center of Vietnam and India (Zhang et al., 2008; Wang et
241 al., 2016). High level of PCBs in Nepalese cities may in part be due inadequate management of
242 PCB containing equipment.

243 *3.3. Intercity variation*

244 *3.3.1. DDTs and their metabolites:*

245 In Nepal, chlorinated pesticides was introduced in 1950 for the first time by importing DDT
246 and pyrethrum from USA, exclusively for malaria control (Neupane, 1995). Later, the success of
247 DDT in controlling the vector of malaria encouraged Nepalese farmers to apply DDT in the
248 agricultural sector as well. The long-term usage of DDT for controlling malaria and its usage in
249 agricultural sector resulted rapid increase in import of chlorinated pesticides and other groups of
250 pesticides in Nepal before DDT was banned in 2001. Despite the replacement of DDT with other
251 non-POPs pesticides for malaria control since 1995, and in the ban to follow in 2001, large
252 quantities of DDTs may still enter Nepal illegally from India, where DDT is still being produced
253 and used (MOEST, 2007). The concentration of ∑DDTs was highest in Biratnagar and Birgunj
254 and ranged from 365-5838 pg/m^3 and 211-1639 pg/m^3, respectively (**Fig. 2**). The high level of
255 DDTs in these two plain cities might in part be due to intensive farming and/or continuing use of
256 DDTs for malaria control (Liu et al., 2009; Zhang et al., 2012; Hu et al., 2014).The level of ∑DDTs
257 was much lower in Kathmandu (44-194pg/m^3), the only metropolitan city in the country, followed
258 by Pokhara (51-202pg/m^3). The similar trend of DDT level in these two hill cities indicates that
259 these sites are likely to have similar sources of DDT. DDTs have been used for several years both

in Nepal and bordering India for controlling malaria, which could be the possible reason for DDT contamination through long range atmospheric transport.

The relative abundance of parent compound and their metabolites (*p,p′*-DDT/*p,p′*-DDE + *p,p′*-DDD) has been used to assess either recent application (if ratio >1)or historical use (if ratio<1) (Zhang et al., 2008).The ratio of *p,p′*-DDT/(*pp′*-DDE+ *pp′*-DDD) were found to be greater than unity in all cities (**Fig. 3**) suggesting the recent application of DDT. The ratio of *o,p′*-DDT/*p,p′*-DDT can be used to differentiate the technical DDT (if ratio < 0.3) from "dicofol-type DDT" (if ratio >1.9) (Devi et al., 2015).The ratio of *o,p′*-DDT/*p,p′*-DDT were low (**Fig. 3**), irrespective of study sites, also suggesting the use of technical DDT. Low *o,p′*-DDT/*p,p′*-DDT ratio observed in this study is contrasting to trend observed in urban center of China and India, where much higher *o,p′*-DDT/*p,p′*-DDT ratio were associated with application of dicofol-type DDT. Moreover, lower *o,p′*-DDT/*p,p′*-DDT was also reported in Pakistan (Nasir et al., 2014).

3.3.2. Hexachlorocyclohexanes (HCHs)

HCHs and DDTs have been used for several years in India and China for agricultural purpose before it was banned in 1997 and 1983, respectively (Zhang et al., 2011; Yadav et al., 2015). Although, the usages of HCHs were banned for agricultural purposes, exemptions allowed for some continued uses, which may in part help explain the continued presence of these chemicals. Nepal is not an exception to this, and because of the open border with India, Nepalese farmers can easily buy HCH as much they need. HCH dust was the most frequently sold chemical pesticide in Nepal before it was officially banned in 2001 (SKJ, 2003).

HCH has two formulations; technical-grade and lindane. Technical-grade HCH contains 60-70% α-HCH, 5-12% β-HCH, 10-15% γ-HCH, 6-10% δ-HCH and 3-4% ε-HCH, while lindane

constitutes >99% of γ-HCH. Concentrations of ∑HCH was highest in Kathmandu (13-2820 pg/m^3) followed by Birgunj (38-305 pg/m^3), Biratnagar (51-199 pg/m^3) and Pokhara (16-60 pg/m^3) (**Table 1**). Generally, the high ratio of α/γ-HCH (between 3 and 7) is indication of fresh input of technical grade HCH, whereas as ratio below 3 indicate lindane application. In this study, the ratio of α/γ-HCH ranged from 0.02-0.52 in all study sites, suggesting lindane contamination in atmosphere (**Fig. S2**, Supplementary Information). Further, high relative abundance of γ-HCH confirm lindane contamination (**Fig. S3**, Supplementary Information). This is consistent with the GAPS study, which found lindane as the principal source of HCH in global atmosphere (Shunthirasingham et al., 2010). However, historical use of technical HCH cannot be totally ignored in Nepal on the basis of observed β-HCH isomers in air.

3.3.3. Endosulfans

Endosulfan is the most popular and lonely organochlorine pesticides which are still being used in Nepal to protect varieties of crops especially rice, potato, tomato, vegetables and tea plant both in hill and plain area of the country. Sometimes endosulfans are also put in rivers and streams by fisherman to catch fish easily. Dahal (1995) found that about 95% of Nepalese farmer used endosulfan and other organochlorine pesticides to control pests in crop and to store food grain after harvesting. ∑endosulfans ranged from 16-71pg/m^3 (average 40 pg/m^3), 26-51 pg/m^3 (average 38 pg/m^3), 14-48 pg/m^3 (average 29 pg/m^3) and 12-46 pg/m^3 (average 9 pg/m^3) in Birgunj, Biratnagar, Pokhara and Kathmandu, respectively (**Table 1**). Comparatively, the plain areas (Birgunj and Biratnagr) showed higher concentrations of endosulfan in comparison to the hill areas (Kathmandu and Pokhara). Technical endosulfan constitutes 70% of α-endosulfan and 30% β-endosulfan (Chakraborty et al., 2010). β-endosulfan is more reactive in atmosphere and gets converted to α-endosulfan after post application (Nasir et al., 2014).The higher ratio of α/β-endosulfan (if ratio

>2.3) is the indication of past usage of endosulfan or influence of long range transport, while ratio closer to 2.3 suggests recent use of technical endosulfan (Qu et al., 2015). The overall ratio of *α/β*-endosulfan in this study ranged between 0.72-2.67 (average 1.81) (**Fig. S2,** Supplementary Information), suggesting current usage of technical endosulfan in Nepal (Chakraborty et al., 2010; Pozo et al., 2011). Moreover, a high *α/β*-endosulfan ratio in Kathmandu (2.67) may suggest historical use and possible influence of long range transport of endosulfan from India (Nasir et al., 2014).

3.3.4. Chlordane-related compounds

Since 2001, Nepal banned all POPs pesticides including chlordane for their import, export and use. However, due to trans-boundary movement and illegal importation, some of these banned pesticides are still found in local markets. It is one of the most popular pesticides among Nepalese farmers and widely used in cash crops such as sugarcane, tomato, potato and other vegetables. High concentration of TC and CC were detected in the hill areas. The level of TC in Pokhara and Kathmandu ranged 7-197pg/$m^3$and 4-70pg/m^3, respectively (**Table 1**). Technical chlordane is a mixture of TC (13%), CC (11%), HEPT (5%) and trans-Nonachlor (Zhang et al., 2012) with TC/CC ratio of 1.2 (Chakraborty et al., 2010). TC is more easily degradable than CC in the environment. Hence, TC/CC ratio can be utilized to trace aged or fresh chlordane. The TC/CC ratio in technical chlordane is reported to be 1.2 (Chakraborty et al., 2010). Moreover, the ratio of TC/CC would increase to 1.63 and 1.95 at 20°C and 25°C, respectively in air arising from volatilization from soil or technical chlordane (Park et al., 2011; Qu et al., 2015). The TC/CC ratio in hill area (Kathmandu and Pokhara) was calculated to be 3.25 and 2.17 (**Fig. S2,** Supplementary Information), higher than ratio 1.95, suggesting recent application of technical chlordane (Qu et al., 2015). However, low TC/CC ratio was observed in plain area (Birgunj and Biratnagar) with

328 1.64 and 1.4, respectively indicating their past use in these areas (Zhang et al., 2013). The low
329 TC/CC in plain area might also due to remission from weathered chlordane source (Chakraborty
330 et al., 2010).

331 HEPT is a chlorinated cyclodiene insecticide, which is used for controlling soil insect,
332 termites, grasshoppers, and mosquitoes. HEPT is degraded to more stable HEPX in air through
333 photolysis or/epoxidation in plants, soil and animals (Nasir et al., 2014).HEPT was the least
334 detected chemical among all OCPs. The concentration of HEPT together with HEPX ranged from
335 3-24 pg/m^3, 13-27 pg/m^3, 13-20 pg/m^3, and 5-15 pg/m^3 in Pokhara, Kathmandu, Birgunj and
336 Biratnagar, respectively (**Table 1**).Low level of HEPT in the atmosphere of this study might be
337 due short half-life of HEPT compared to other OCPs (Baek et al., 2013).HEPX is a breakdown
338 product of HEPT that can remain in soil and water for a long time. Hence, higher ratio of
339 HEPT/HEPX (if ratio>1) is the indication of fresh application of HEPT. The HEPT/HEPX ratio
340 was below unity in all cities except Kathmandu, indicating the dominance of HEPX. However,
341 HEPT/HEPX ratio was greater than 1 in Kathmandu. Although HEPT was deregistered in Nepal
342 in 2001, the detection of parent compound in Kathmandu might be because of ongoing use of
343 HEPT in home lawn and garden as termiticides (Nasir et al., 2014). Besides, the low level of HEPT
344 in this study could also due to application of TC (Baek et al., 2011).

345 *3.3.5. Hexachlorobenzene (HCB)*

346 Hexachlorobenzene was introduced for the first time in 1933 and mainly used as fungicide
347 to protect wheat and other seeds. It is also utilized in the production of fireworks and synthetic
348 rubber. Although, the usage of HCB was banned in 1980s in most countries after global production
349 of HCB exceeded 100,000 tons, it is still present in the environment as a byproduct and/or impurity

in the manufacture of chlorinated solvents, chlorinated pesticides and produced during incomplete combustion processes (Barber et al., 2005).Comparatively, highest concentration of HCB was detected in Kathmandu (average 37 pg/m^3), than in Birgunj (average 25 pg/m^3), Biratnagar (average 24 pg/m^3) and Pokhara (average 14pg/m^3).Concentrations of HCB observed in this study is much lower than those reported in Vietnam (589 pg/m^3), China (158 pg/m^3), South Korea (148 pg/m^3), and Norway (Ling et al., 2011; Park et al., 2011; Halse et al., 2012; Wang et al., 2016) (**Table S5**, Supplementary Information).However, these level were consistent with those reported in Italy (4 pg/m^3), Pakistan (33 pg/m3) and Ghana (24 pg/m^3) (Estellano et al., 2012; Syed et al., 2013; Hogarh et al., 2014). Air concentration of HCB observed in this study was fairly constant suggesting the air concentration of HCB is mainly associated with diffused continental sources rather than local sources (Estellano et al., 2012). HCB has a relatively high volatility and atmospheric half-life and therefore a compound with high potential of LRAT (Liu et al., 2010).

3.3.6. Other OCPs

Aldrin, dieldrin and endrin are among the 12 ‘dirty dozen’ POPs on the Stockholm Convention. They were widely used as insecticides to control termites, textile pests and rodents such as mice and voles. Dieldrin is the main metabolite of Aldrin and its active compound. Aldrin itself is not toxic to insects, but is rapidly converted to dieldrin in plants and animals. The photo-degradation rate of dieldrin is much lower than the Aldrin (Rowland et al., 2011).Endrin is a stereoisomer of dieldrin which get removed from the environment by bacterial degradation (WHO, 2004; UNIDO, 2013). Endrin and dieldrin is the most detected “drin” OCPs in plain areas. The average drin concentration were (aldrin 10 pg/m^3 and 9 pg/m^3), dieldrin (11 pg/m^3 and 11 pg/m^3), endrin (25 pg/m^3 and 19 pg/m^3) and (isodrin 4 pg/m^3 and 4 pg/m^3) in Biratnagar and Birgunj, respectively (**Table 1**). These concentrations are consistent with previous study in Chiapas

Mexico, Chile and Costa Rica (Pozo et al., 2004; Daly et al., 2007; Alegria et al., 2008).The low concentration of dieldrin suggest they are no longer being used in this region. Dieldrin and other drin OCPs have been banned in Nepal as well as in India and China, which may explain the low levels in this study (PRMS, 2010; Zhang et al., 2011; Yadav et al., 2015). Concentrations of mirex were consistently low irrespective of study sites, with an average of 5 pg/m^3 suggesting background level as mirex was not known to have been used in Nepal.

3.3.7. Polychlorinated biphenyls (PCBs)

PCBs have never been intentionally produced in Nepal. However, the grants assistance by various donor countries in developing the hydropower (HP) stations in Nepal could have been a possible entry of PCBs in the country. Another possible entry of PCBs might be import of large quantities of dielectric fluid and transformer oil by Nepal Electricity Authority (NIP, 2007).Among 32 targeted PCBs, only 26 congeners(PCB-8,-28,-37,-44,-49,-52,-60,-66,-70,-74,-77,-87,-99,-101,-105,-118,-126,-153,156,-158,169,-170,-180,-183,-187, and -189) were detected in air samples. The level of individual PCBs are presented in **Table 2**. The concentration of $\sum_{26}$ PCBs ranged from 65-1002 pg/m^3(average 289 pg/m^3), 78-534 pg/m^3 (average 242 pg/m^3), 62-127 pg/m^3 (average 89 pg/m^3) and 30-76 pg/m^3 (average 51pg/m^3) in Kathmandu, Birgunj, Biratnagar and Pokhara, respectively. High concentrations of $\sum_{26}$PCBs were mostly observed in highly urbanized (in case of Kathmandu) and industrial cities (in case of Birgunj) (Jamshidi et al., 2007; Du et al., 2009). High levels of PCBs are typically associated with urban centers that are known to be potential sources of PCBs (Pozo et al., 2006). The most volatile PCBs with low molecular weight tends to be present in gaseous phase, hence can easily be transported in air, while less volatile PCBs with higher molecular weight tends to partition to aerosols and deposit near source (Pozo et al., 2006; Nasir et al., 2014). This concept is supported by decreasing trend of atmospheric PCBs

level with increasing molecular weight of PCB congeners (**Fig.1**). An additional explanation is that PUF-PAS mostly capture gaseous phase chemicals, and the uptake of high chlorinated PCB might be low by PUF-PAS (Wania et al., 2003). Among the so-called indicator congeners, PCBs -28 and -52 (most volatile) were the most abundant congeners detected in air at all sampling sites and ranged 3-331pg/m^3 and 1-67 pg/m^3, respectively (**Fig. 4**), while PCB 101(0.2-39 pg/m^3), PCB118(nd-15 pg/m^3), PCB153 (0.6-29 pg/m^3), and PCB180(0.6-10 pg/m^3) were present in lesser amounts. The elevated level of PCBs congener -28 in air might also in part be due re-emission and volatilization of PCBs from soil (Aichner et al., 2007).

Depending on the numbers of chlorines, PCBs in this study may be classified into six groups. These are di-CBs (PCB-8), tri-CBs (PCB-28, and -37), tetra-CBs (PCB-44,-49,-52,-60,-66,-70,-74, and -77), penta-CBs (PCB-87,-99,-101,-105,-118, and -126), hexa-CBs(PCB-153,-156,-158,and -169) and hepta-CBs (PCB-170,-180,-183,-187,and-189). About 70% of total PCBs produced globally were made up low chlorinated congeners especially tri-, tetra- and penta-CBs (Breivik et al., 2002). The individual composition profile of chlorinated PCBs showed that PCBs were mostly dominated by tri-, tetra-, and di-CBs, accounting 38%, 34% and 11% of ∑PCBs (**Fig. S3**, Supplementary Infomation). Abundance of tri-CBs was also reported in surface soil from Kathmandu (Aichner et al., 2007). Marginally, higher contribution of hexa- and hepta-CBs were observed in Kathmandu (4.8% and 3.6% of ∑PCBs, respectively) and Birgunj (9.9% and 4.5% of ∑PCBs, respectively).Because of low potential of atmospheric transport of these congeners, they are more likely to remain at sources region (Choi et al., 2008). Thus, trend of these congeners suggest Nepalese urban area are sources of atmospheric PCBs, most likely due to PCBs emission from dielectric fluid, electrical transformer and contamination from hydraulic fluid oil (Aichner et

418 al., 2007; NIP, 2007). However, hexa- and hepta-CBs were significantly contributed to ∑PCBs in
419 Pokhara and Biratnagar suggesting possible influence from local source.

420 The backward trajectory of air mass over all four cities is similar except in Biratnagar.
421 Back trajectory analysis suggest that most of air mass (90-95%) in Kathmandu, Pokhara and
422 Birgunj during sampling originated from India and less than 5% were from Bay of Bengal (**Fig.**
423 **5**).Unlike to these, 43% air mass that arrived at Biratnagar during sampling were associated with
424 Bay of Bengal and rest from India.

425 *3.4. Altitudinal variations*

426 The altitudinal variation of chlorinated POPs (HCHs, DDTs, Endos and PCBs) in air
427 concentration was investigated by plotting the concentration vs altitudinal gradient. Among
428 chlorinated OCPs, higher concentrations of DDTs and occasionally Endos were observed in
429 Birgunj and Biratnagar (low altitude areas), while DDTs were less detected in Kathmandu and
430 Pokhara (high altitude areas), suggesting local application of these chemicals mainly in the former
431 areas mentioned (**Fig. S5**, Supplementary Information). Unlike DDTs and Endos that increased
432 with decreasing altitude, HCHs did not show any clear trend of either increasing or decreasing
433 concentrations with altitude. This may indicate rather well-mixed conditions for HCHs in air and
434 a general lack of clear source-receptor relationships. Homogeneous concentration of HCHs
435 observed in this study is in good agreement with previous study by Miere et al. (2012) and Liu et
436 al. (2010) in Brazilian and Chinese mountains, respectively.

437 Higher concentrations of PCBs were detected at KTM (hill) and BRG (plain) (**Fig. S5**,
438 Supplementary Information) emphasizing the likely influence from local sources at these sites

(Meire et al., 2012).The similar altitudinal trend of PCBs have also reported in Souther Brazil, Italy and Bolivian Andes (Jaward et al., 2005; Estellano et al., 2008; Miere et al., 2012).

3.5. Cancer risk assessment

The maximum inhalation exposure through air was calculated for DDTs, HCHs, endosulfans, chlordane and PCBs. Maximum environmental exposure were calculated for DDTs, HCHs and chlordane chemicals with average exposure level ranging between 11-463 pg/kg/day, 11-136 pg/kg/day and 2-11 pg/kg/day, respectively (**Table S6**, Supplementary Information). The inhalation exposure for $\sum_{26}$PCBs ranged between 14-80 pg/kg/day. The total human health risk associated with inhalation was estimated following USEPA inhalation dosimetry guideline (OEHHA, 2002; EPA, 2003). Total health risk was only evaluated for those compounds for which inhalation unit risk factor (IUR) is available (OEHHA, 2002; USEPA, 2009). Total cancer risk was not estimated for endosulfan because of non-availability of its IUR value. With respect to total cancer risk, none of the OCPs and PCBs exceeded the carcinogenic bench mark level described by USEPA (1×10^{-6}), suggesting the safe level. Even after considering the total cancer risk as cumulative, it would not surpass the threshold limit. This suggests that the measured concentrations of legacy POPs in air from this study represent a negligible health risk through inhalation of ambient air. However, other modes of human exposure could still be relevant in Nepal. The occasional high concentrations of DDT reported in this study warrants for further studies to assess implications for human exposure through other modes than inhalation.

4. Conclusions

The occurrence and possible source of legacy POPs was studied in four potential source region in Nepal targeting urban areas at both lower and higher altitudes. The concentrations of

overall POPs at lower altitudes were approximately twice that of higher altitudes. DDTs and HCHs were the most abundant OCPs measured in air. The DDT fingerprints observed in this study reflected influence from fresh use of technical DDT. High levels of $\sum_{26}$PCBs occasionally detected in this study was linked to possible emissions in more highly urbanized / industrial areas. The measured concentrations of legacy POPs in air from this study is assumed to represent a negligible health risk through inhalation of ambient air, however, other modes of human exposure could still be relevant in Nepal. The air mass backward trajectory analysis revealed that most of air mass during sampling period at present study sites originated from India and Bay of Bengal.

Supplementary Information

Detailed information on sampling and experimental procedure, QA/QC, GC-MS parameter, study area map, site specific abundance of OCPs and PCB congeners, altitudinal trend of OCP and PCBs, statistical summary of OCPs and PCBs, comparison with global studies, isomeric ratio table and overall health risk summary table.

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Figure captions

Fig.1 Box plot of OCPs (top) and PCBs (bottom) in air. The concentration in Y-axis is in logarithmic scale

Fig.2 Spatial distribution of DDTs, HCHs and endosulfans in air. The concentrations are plotted in logarithmic scale

Fig.3 Scattered plots of *o,p′*-DDT/*p,p′*-DDT and *p,p′*-DDT/(*pp′*-DDE+ *pp′*-DDD)

Fig.4 Spatial distribution of PCBs in air. The concentrations are plotted in logarithmic scale

Fig.5 Five days backward air mass trajectory cluster plotted in Kathmandu, Pokhara, Birgunj and Biratnagar by HYSPLIT model.

Table captions

Table 1 Statistical summary of OCPs (pg/m^3)

Table 2 Statistical summary of PCBs (pg/m^3)

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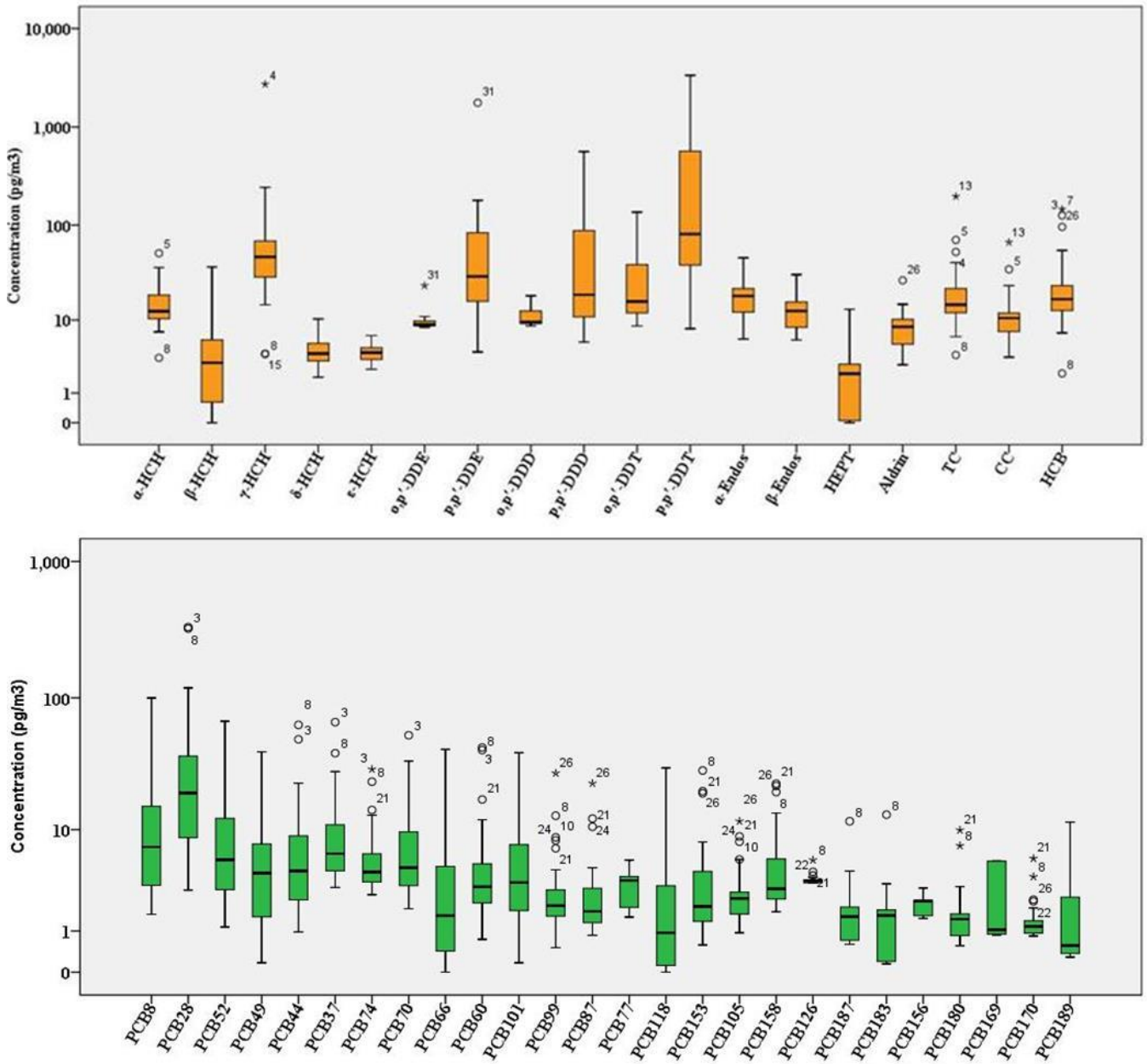


Fig.1 Box plot of OCPs (top) and PCBs (bottom) in air. The concentration in Y-axis is in logarithmic scale.

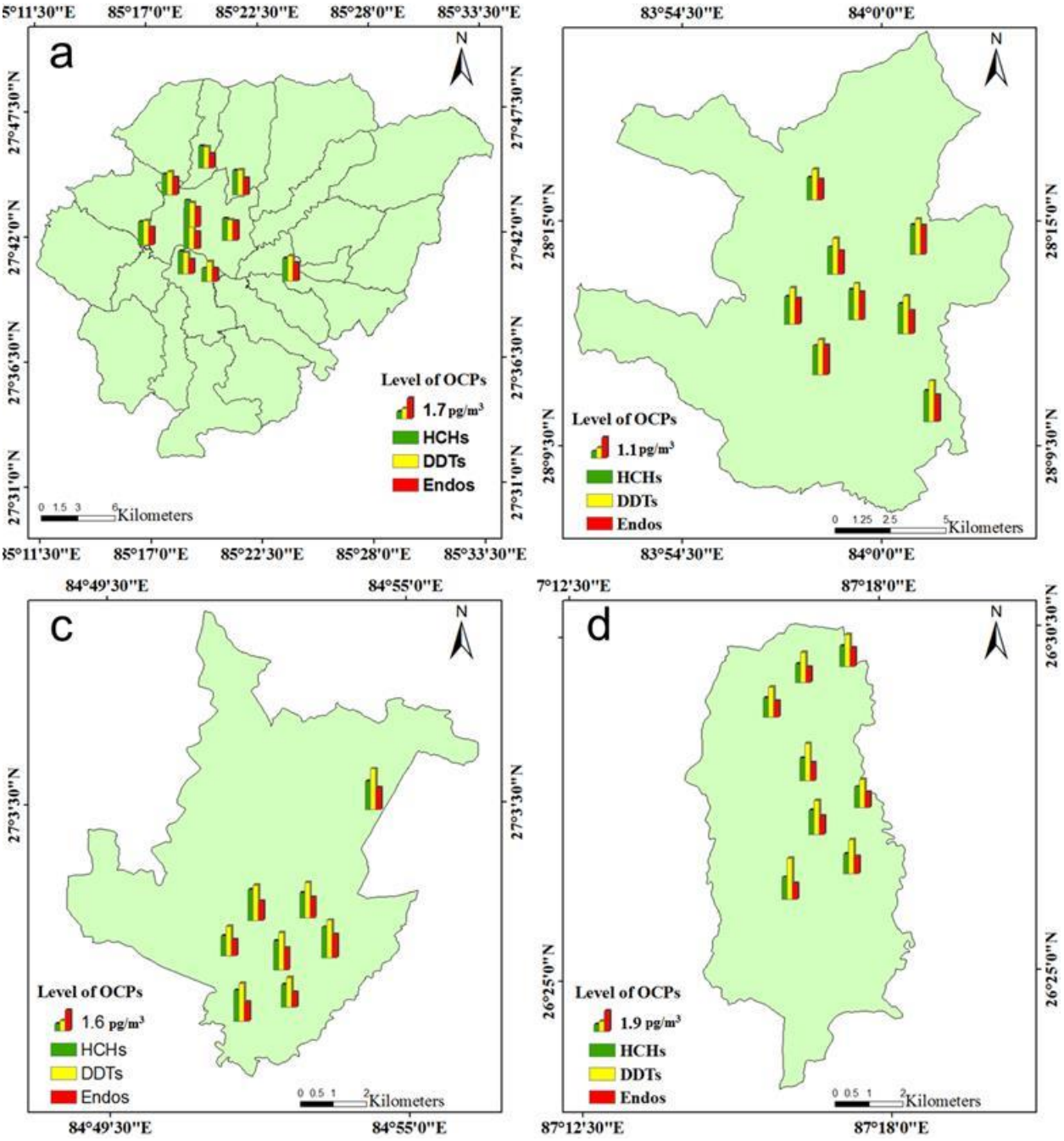
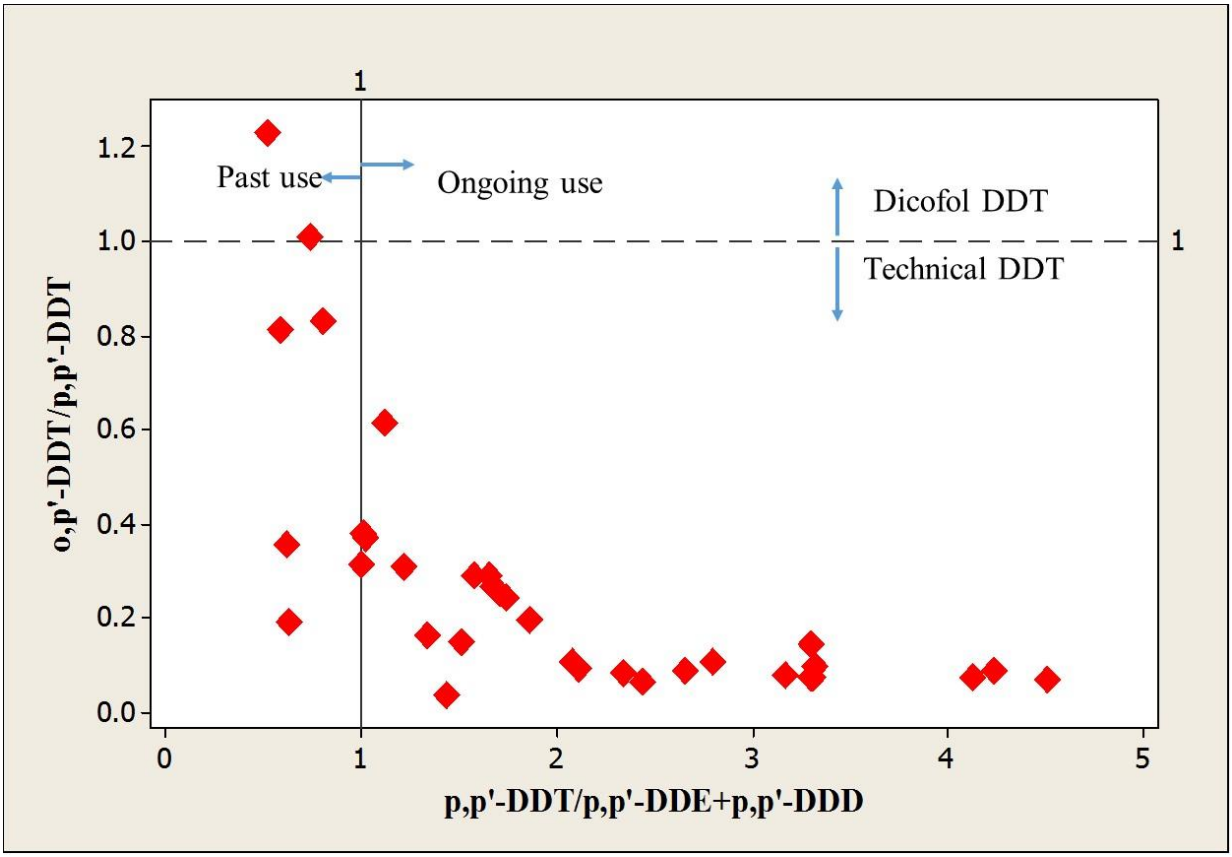


Fig. 2: Spatial distribution of DDTs, HCHs and endosulfans in (a) Kathmandu, (b)Pokhara, (c) Birgunj, and (d) Biratnagar. The concentrations are plotted in logarithmic scale.



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742 Fig. 3 Scattered plots of *o,p′*-DDT/*p,p′*-DDT and *p,p′*-DDT/(*pp′*-DDE+ *pp′*-DDD)

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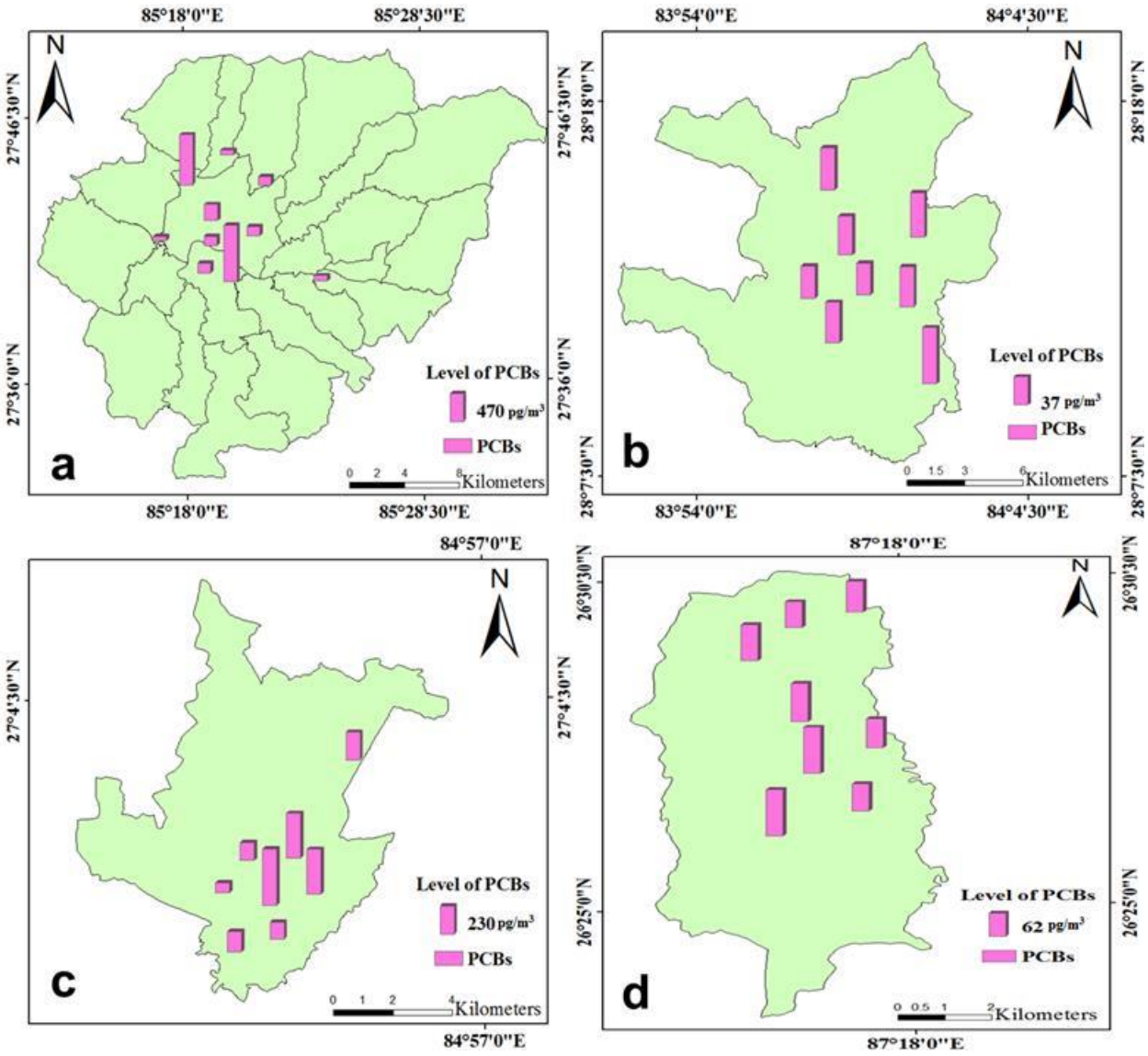

Fig. 4 Spatial distribution of ∑PCBs in (a) Kathmandu, (b)Pokhara, (c) Birgunj, and (d) Biratnagar. The concentrations are plotted in logarithmic scale.

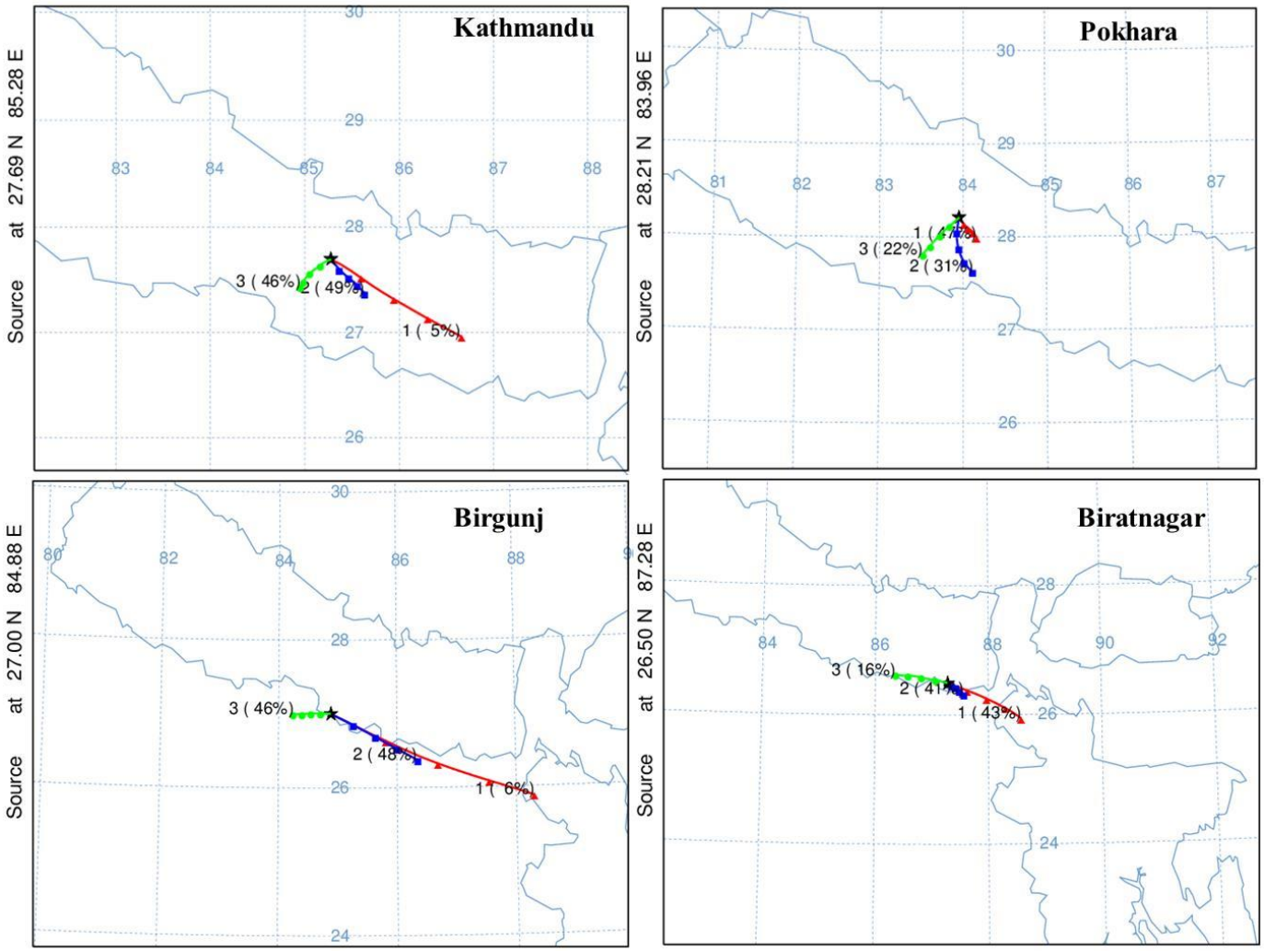

Fig. 5 Five days backward air mass trajectory cluster plotted in Kathmandu, Pokhara, Birgunj and Biratnagar by HYSPLIT model.

773 Table 1 Statistical summary of OCPs (pg/m^3)

OCP (pg/m^3)	Kathmandu				Pokhara				Birgunj				Biratnagar			
	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD
α-HCH	3.50	51.4	7.80	13.0	7.40	13.0	10.9	1.80	8.30	36.5	20.9	9.70	9.40	21.1	12.9	3.60
β-HCH	ND	37.0	6.80	10.9	ND	2.80	1.10	1.20	ND	14.4	5.60	4.60	ND	25.8	6.30	8.40
γ-HCH	4.00	2720	480	843	4.00	35.7	21.1	9.50	24.1	243	130	83.5	33.6	138	62.6	33.2
δ-HCH	2.70	10.3	1.90	2.40	1.90	3.70	3.00	0.70	3.20	6.30	4.60	1.10	3.00	8.20	5.20	1.80
ε-HCH	2.50	6.70	1.20	1.50	2.80	4.30	3.70	0.50	2.60	4.80	3.80	0.80	4.20	6.00	5.10	0.70
∑HCHs	12.7	2830	498	871	16.1	59.5	39.8	13.7	38.2	305	165	99.7	50.2	199	92.1	47.7
o,p′-DDE	8.30	8.90	0.20	0.20	8.50	9.00	8.80	0.20	8.80	10.9	9.80	0.70	9.20	23.6	11.4	5.00
p,p′-DDE	4.20	83.5	15.5	23.5	9.70	25.5	17.8	5.20	40.8	180	116	52.2	29.1	1760	265	604
o,p′-DDD	8.60	9.50	0.30	0.30	9.10	9.40	9.20	0.10	10.3	18.4	13.3	2.60	10.0	15.4	12.4	2.10
p,p′-DDD	5.80	14.8	2.80	3.30	5.60	20.6	13.1	4.70	28.8	174	80.6	45.4	54.7	562	207	171
o,p′-DDT	8.60	15.4	1.90	2.40	9.80	17.6	12.5	2.20	16.5	86.3	41.9	22.2	25.4	136	70.7	41.8
p,p′-DDT	8.50	62.1	18.8	21.6	8.00	120	47.6	33.0	105	1170	487	342	236	3340	1120	1060
∑DDTs	44.0	194	39.5	51.3	50.7	202	109	45.4	210	1640	749	465	364	5840	1690	1880
α-Endo	6.10	30.5	6.50	8.00	8.30	34.0	19.9	9.50	8.30	46.1	24.8	13.6	12.1	19.7	15.7	3.30
β-Endo	6.20	15.5	2.40	3.10	5.90	14.1	8.90	2.60	7.10	25.2	15.3	6.10	13.9	30.8	21.8	7.00
∑Endos	12.3	46.0	8.90	11.1	14.2	48.1	28.8	12.1	15.4	71.3	40.1	19.7	26.0	50.5	37.5	10.3
HEPT	1.50	13.2	2.10	3.50	ND	5.80	1.50	2.00	1.80	6.20	3.30	1.40	ND	2.90	0.70	1.10
t-HEPX	3.20	3.60	0.20	0.20	0.90	7.10	4.40	2.30	3.10	3.90	3.30	0.20	1.40	7.00	3.60	2.80
c-HEPX	8.50	9.90	0.40	0.50	2.00	10.9	6.40	3.20	8.40	9.70	9.10	0.50	3.10	4.80	4.40	0.50
TC	3.90	70.5	15.1	19.8	7.40	197	37.3	64.6	6.50	15.9	11.3	2.80	11.1	25.9	16.2	5.10
CC	3.60	35.0	6.30	8.80	7.30	66.6	17.2	20.0	4.70	9.80	6.90	1.50	9.20	15.4	11.5	2.20
Aldrin	2.90	14.9	3.10	3.80	3.70	10.6	6.70	2.60	3.10	26.8	9.10	7.70	8.30	10.5	9.50	1.00
Isodrin	2.90	6.80	1.10	1.40	3.10	5.00	3.60	0.70	2.80	7.20	4.10	1.50	3.60	6.30	4.40	0.90
Dieldrin	3.80	20.3	3.70	4.90	5.70	11.0	8.20	1.90	5.00	21.9	11.2	5.40	9.30	12.2	11.0	1.00
Endrin	3.00	32.5	6.90	9.00	8.40	20.4	15.1	4.40	6.30	37.0	19.4	10.7	15.2	35.9	25.2	7.30
HCB	2.20	146	37.3	50.1	10.3	16.9	13.5	2.40	7.10	95.9	24.6	29.2	16.1	55.0	24.0	12.8
Methoxychlr	1.10	2.20	0.20	0.30	ND	3.50	1.00	1.30	1.10	2.50	1.70	ND	ND	ND	ND	ND
Mirex	4.70	4.90	0.10	0.10	4.50	4.80	4.70	0.10	4.70	4.90	4.80	0.10	4.50	4.80	4.70	0.10
∑OCP	110	3420	622	1040	134	670	297	177	319	2260	1060	646	523	6270	1930	1970

774 HEPT: heptachlor; t-HEPX: trans heptachlor epoxide; c-HEPX: cis heptachlor epoxide; TC: trans chlordane; CC: cis chlordan

775 Table 2 Statistical summary of PCBs in air (pg/m3)

PCBs (pg/m3)	Kathmandu				Pokhara				Birgunj				Biratnagar			
	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD	Min	Max	Mean	SD
PCB8	4.40	99.9	31.6	36.9	1.70	3.30	2.40	0.6	3.40	33.3	14.7	11.7	2.60	15.3	6.70	4.40
PCB28	14.9	331	92.8	125	3.00	8.50	4.80	2.00	13.6	119	51.6	36.5	8.60	22.0	14.4	5.50
PCB52	4.20	67.2	19.8	21.3	1.10	3.40	1.90	0.80	5.00	29.0	15.7	8.90	2.80	8.00	4.90	2.00
PCB49	1.40	39.7	10.9	14.1	0.20	3.50	1.40	1.10	1.40	15.0	7.40	4.50	2.90	8.10	5.10	2.00
PCB44	3.60	63.1	17.2	21.1	1.00	2.50	1.60	0.60	3.80	23.1	12.1	6.80	1.80	4.90	3.40	1.20
PCB37	6.30	66.2	18.2	19.3	3.20	5.00	4.10	0.70	6.40	28.2	14.4	6.80	4.00	6.20	4.90	0.80
PCB74	3.60	29.4	9.70	9.10	2.70	4.00	3.20	0.40	3.20	14.3	8.80	3.90	3.60	5.20	4.30	0.60
PCB70	3.40	52.8	14.9	16.1	1.90	3.30	2.30	0.50	4.70	24.8	15.2	7.80	2.70	6.70	4.40	1.40
PCB66	0.80	41.5	11.2	16.0	ND	0.40	0.10	0.20	1.00	16.8	8.20	5.40	ND	2.40	1.20	0.90
PCB60	1.20	42.8	11.6	16.2	0.70	3.00	1.80	0.90	2.10	17.3	8.50	4.80	2.70	4.60	3.50	0.70
PCB101	2.00	15.6	7.20	4.10	0.20	3.10	1.30	1.10	4.40	39.2	14.0	11.6	1.00	3.60	2.20	0.90
PCB99	0.50	12.9	3.70	4.00	0.50	2.00	1.30	0.60	1.60	27.4	6.90	8.70	1.80	3.20	2.40	0.50
PCB87	1.30	4.40	2.60	1.00	0.90	1.40	1.10	0.20	1.70	23.0	7.90	7.20	1.10	3.00	1.80	0.70
PCB77	1.50	5.60	2.70	1.50	1.60	4.20	3.00	1.10	2.00	5.60	3.60	1.40	3.60	4.40	3.90	0.20
PCB118	ND	14.8	3.70	5.10	ND	0.30	0.10	0.10	2.10	30.1	9.20	9.90	ND	1.70	0.70	0.70
PCB153	0.60	28.7	4.90	8.50	0.60	2.10	1.20	0.50	2.90	20.0	9.10	6.70	1.60	2.60	2.00	0.40
PCB105	1.30	5.70	2.40	1.70	0.90	2.50	1.90	0.80	1.40	11.7	4.90	3.90	2.50	3.60	2.90	0.50
PCB158	2.40	19.7	5.80	5.50	1.80	3.00	2.30	0.50	5.50	22.9	11.9	7.00	2.10	3.60	2.70	0.60
PCB126	3.60	5.60	3.90	0.60	3.50	3.70	3.60	0.10	3.60	4.40	3.90	0.30	3.50	3.50	3.50	0.00
PCB187	1.50	11.7	2.80	3.10	0.60	1.60	1.00	0.50	1.90	4.50	2.60	0.90	0.60	0.80	0.70	0.10
PCB183	1.60	13.2	2.90	3.60	0.20	1.70	0.70	0.70	1.60	3.40	2.30	0.70	0.20	0.40	0.20	0.10
PCB156	1.50	2.90	1.80	0.50	1.50	2.40	2.00	0.40	1.60	3.10	2.20	0.50	2.30	2.50	2.40	0.10
PCB180	0.70	7.40	1.80	2.10	0.60	1.70	1.10	0.40	0.80	9.90	3.10	2.80	1.40	1.60	1.50	0.10
PCB169	0.90	5.40	1.40	1.40	0.90	5.50	3.80	2.40	0.90	1.40	1.00	0.20	5.50	5.50	5.50	0.00
PCB170	1.10	4.00	1.50	0.90	0.90	1.20	1.00	0.10	1.20	5.80	2.40	1.40	0.80	1.20	1.00	0.10
PCB189	0.40	11.5	1.60	3.50	0.30	2.60	1.70	1.10	0.30	1.80	0.50	0.50	2.50	2.60	2.60	0.00
$\sum_{26}$PCB	64.6	1002	289	342	30	76	51	18	78	534	242	161	62	127	89	24

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