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Concentration and distribution of pesticide residues in soil: Non-dietary human health risk assessment

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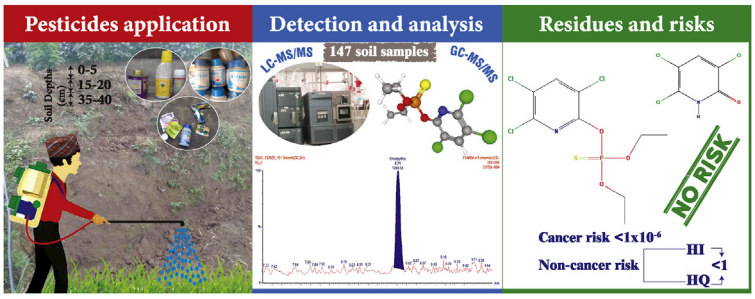
HIGHLIGHTS

- 3,5,6-trichloro-2-pyridinol and chlorantraniliprole were the most frequently detected.
- Chlorpyrifos and p,p'-DDT had the highest concentrations in soil.
- Health risk of soil pollutants to humans was negligible.
- Soils from integrated pest management fields had the lowest concentration of residues

GRAPHICAL ABSTRACT



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ABSTRACT

Soil contamination by pesticide residues is a primary concern because of the high soil persistence of pesticides and their toxicity to humans. We investigated pesticide concentration and distribution at 3 soil depths in 147 soil samples from agricultural land and assessed potential health risks due to non-dietary human exposure to pesticides in Nepal. About sixty percent of the soil samples had pesticides (25% of the soil samples had single residue, 35% of the soil samples had mixtures of 2 or more residues) in 39 different pesticide combinations. Pesticide residues were found more frequently in topsoil. Overall, the concentration of pesticides ranged from 1.0 μg kg^{-1} to 251 μg kg^{-1}, with a mean of 16 μg kg^{-1}. The concentration of the primary group, organophosphates (OPs), ranged from 1.23 μg kg^{-1} to 239 μg kg^{-1}, with a mean of 23 μg kg^{-1}. Chlorpyrifos and 3,5,6-trichloro-2-pyridinol (TCP) were the predominant contaminants in soils. The ionic ratio of DDT and its degradation products suggested a continuing use of DDT in the area. Human health risk assessment of the observed pesticides in soil suggested negligible cancer risks and negligible non-cancer risks based on ingestion as the primary route of exposure. The predicted environmental concentrations (PECs) of pesticides were higher than the values found in the guidance for soil contamination used internationally. Low concentrations of residues in the soils from agricultural farms practicing integrated pest management (IPM) suggest that this farming system could reduce soil pollution in Nepal.

1. Introduction

Chemical pesticides have been used in agriculture for decades in

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the effort to reduce crop loss and to meet the world's growing food demands. About one-third of agricultural commodities are produced using chemical pesticides (Zhang et al., 2011). If farmers worldwide all of a sudden stopped using pesticides, crop losses to pests of fruits, veggies and grains would increase by 78%, 54% and 32%, respectively (Cai, 2008). Global production of pesticides increased by 11% annually, from 0.2 million tons in the 1950s to >5 million tons by 2000 (Carvalho, 2017). In 2012, on average, around 3.8 million tons of chemical pesticides were applied to agricultural land (FAO, 2020). This amounts to a value of >40 billion US dollars (Pimentel, 2009). As a consequence of pesticide use, over two million people, mainly residing in developing nations, are at an elevated health risks (Hicks, 2019). The rate of pesticide use varies across the globe, even within the same region. For instance, the average rate of pesticide use is observed highest in Asia, where 6.5–60 kg ha^{-1} insecticides are used (Carvalho, 2017). However, in the regions of Nepal, pesticide use is relatively low at <400 g ha^{-1} (Sharma, 2015). One hundred and seventeen active ingredients of pesticides were registered and approved in Nepal, with annual imports of 410 tons of active ingredients consisting of 34% insecticides, 40% fungicides, 25% herbicides, 1.6% rodenticides, 0.03% biopesticides and 0.02% other botanical pesticides (PRMD, 2015).

Despite the benefits of using pesticides to improve food safety, intensive and widespread use of chemical pesticides can increase soil pollution, thereby increases environmental and health risks. Soil properties play a crucial role in the fate, behaviour and dispersion of chemical pesticides (Lewis et al., 2016) and has become the repository of pesticides used in agriculture. It adsorbs most pesticides and degradation products, which might negatively affect different food webs. Pesticides can ultimately reach to humans (Zhang et al., 2006), and are thereby subject to bioamplification (Alamdar et al., 2014). Pesticides get washed away from soils by running water and thus find their way into water sources. Pesticides can also be emitted into the atmosphere through volatilization (Sweetman et al., 2005), which adversely affect air (Bidleman and Leone, 2004) and surface water quality (Mekonen et al., 2016). Runoff and flooding are two major pathways of movement of pesticides that may lead to unintentional diffusion and non-target contamination (Wong et al., 2017) that can ultimately negatively affect human health. The concentration of pesticides tend to increase with soil depth (Zhang et al., 2006) and thus, the concentrations found in the bottom soil layer may increase underground water pollution (Sankararamakrishnan et al., 2005). Different levels of pesticides were reported across the globe and have threatened humans and the environment (Sun et al., 2016; Houbraken et al., 2017). Along with analytical approaches, such as GC-MS and LC-MS, concentrations of pesticides are often estimated depending on their predicted environmental concentrations (PECs).

Farmers are exposed with pesticide-contaminated soils via different pathways such as dermal contact, direct ingestion, and inhalation (Li, 2018). The increasing probability of humans to develop cancer and non-cancer diseases as a result of such exposure was calculated by adopting USEPA standard models. Indices such as the hazard quotient (HQ) and the hazard index (HI) are used globally for health risk assessment and are mainly based on the concentration of pesticides (Hu et al., 2014; Sun et al., 2016; Pan et al., 2018). Likewise, risk of pesticides is often estimated based on their predicted environmental concentrations (PECs) (Silva et al., 2019; Vasickova et al., 2019), however, studies on PEC in the risk assessment of pesticide use are limited in literature. Further, monitoring pesticide residues in soils and examining their potential health risks is also scanty in Nepal. The application of hazardous insecticides and fungicides is increasing in the areas of Nepal where vegetables are cultivated for markets (Atreya et al., 2011; Bhandari et al., 2018). More than 80% of the pesticides applied in Nepal are used in vegetable farming (Adhikari, 2017). Vegetable farming is becoming increasingly more popular and is a good income option for farmers in Nepal. Adolescents and adults who work in the vegetable fields can be exposed to pesticides via different pathways such as non-dietary ingestion, dermal exposure and soil particle inhalation. In other parts of the globe, there is a growing evidences of human health risk due to pesticide use in agricultural (Ritz and Yu, 2000; Schreinemachers, 2003; Samsel and Seneff, 2013; Shelton et al., 2014). The dietary risks from pesticide ingestion is at the higher end, and thus considered unacceptable (Bhandari et al., 2019). In Nepal, studies have observed an emergence of higher cancer and non-cancer risks associated with pesticide use (Yadav et al., 2016; PPDB, 2019). Hazardous pesticides that are banned for use in the EU (pesticides use status in bold, Table S1, Supplementary information) are still used in Nepal. In addition, a recent study (Bhandari et al., 2018) showed that persistent and toxic pesticides are frequently applied to vegetable fields. This combined with the fact that farmers in Nepal are less likely to follow safe work practices and do not have access to personal protective equipment, resulting the pesticides exposure risks even larger. This study thus conducted to analyze pesticide residues in the soil of vegetable fields, and to estimate potential health risk for humans due to non-dietary exposure to pesticides in soil.

2. Materials and methods

Supplementary information presents information in support of the materials and methods.

2.1. Study area

Gaidahawa Rural Municipality (GRM) (27° 35.429′ N and 83° 19.215' E) of Nepal was selected for the study (Fig. 1). The area and population of the municipality is 96.79 sq. km and 47,565, respectively. About eighty-one percent (approx. 7900 ha) of the land is agricultural. The mean annual rainfall is about 1400 mm. The land is flat and the soil consists mainly of silt, clay and sand. The temperature may reach 42.4 °C and 8.7 °C in summer and winter, respectively (GRM, 2018).

2.2. Farming practice and selection of pesticides

Bhandari et al. (2018) reported that farmers in the study area practice commercial vegetable farming. They grow many types of vegetables such as eggplant, chilli pepper, coriander, tomato, bean, onion, sponge gourd, pumpkin, broccoli, bitter gourd, fenugreek, cauliflower, spinach, okra, radish, cucumber, cabbage, fennel, bottle gourd, and pea. Application rate of pesticides differs by crops. A few farmers practice integrated pest management (IPM) techniques for controlling pests. IPM techniques included were the use of insect traps, animal dung and urine, ash, alcohol, and tobacco. The existing farming system is heavily dependent on the application of chemical pesticides at significantly higher than recommended doses, however IPM fields were less likely to receive high amount of chemical pesticides. The frequency of applications is also higher than recommended, indicating poor agricultural practices. Farmers use 30 L lever-operated Knapsack sprayers. Of all the pesticides used, the general trend in application rate (kg ha^{-1}) of top five pesticides was: mancozeb > dichlorvos > chlorpyrifos > profenofos > triazophos (Bhandari et al., 2018). Farmers even apply these pesticides as cocktails.

The present study accounted commonly used following

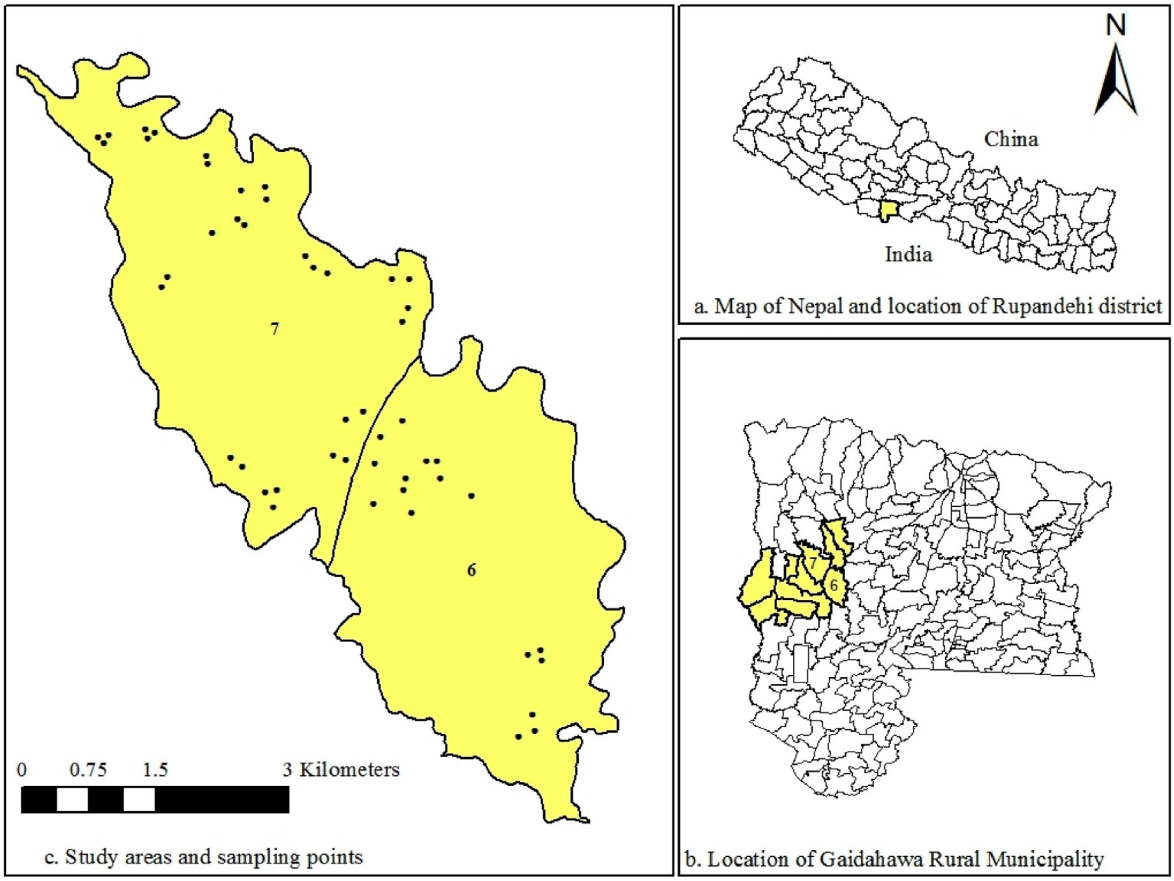

Fig. 1. Map of the study area.

pesticide groups: 7 organophosphates (OP), 1 anthranilic diamide (AD), 1 neonicotinoid (NND), 1 benzimidazole (BD), 1 phenylamide (PA), 1 micro-organism derived (MOD) and 2 unclassified degradation products (UDP). In addition to these compounds, one organophosphate and 8 organochlorines (OC), which are banned in the EU and Nepal due to their high soil persistence and toxicity (Table S1, Supplementary information) were also considered. Due to analytical limits, namely poor recoveries (<70%) as well as logistic and financial limitations, some compounds were excluded.

2.3. The soil samples

At the time of soil sampling, farmers had not sprayed their fields with pesticides for 7 days. Soil samples were collected following the principals laid out in the EU guidelines (Theocharopoulos et al., 2001). The municipality consists of 9 wards, which is the smallest administrative unit of government. Each ward consists of a number of villages. The soil samples were collected from randomly selected villages in the 6th and 7th wards where most farmers were involved in vegetable farming following either IPM or intensive farming practices. The pesticide groups in the areas represented the use and pollution of pesticides in Nepal. Furthermore, the soil sampling areas were the same from where vegetable samples were selected for a study (Bhandari et al., 2019). The soil samples were taken from a total of 49 farmers' standing vegetable fields at 3 depths (0–5 cm, 15–20 cm and 35–40 cm). There were 27 samples from eggplant fields, 36 samples from chilli fields and 84 samples from tomato fields, altogether 147 soil samples were collected during the vegetable growing season in 2017. An auger was used to collect soil samples. Ten samples from each sampling field were collected and mixed thoroughly after foreign materials such as stones, leaves, pebbles, gravel and roots were removed. A composite and representative sample was then collected by quartering and compartmentalization. Furthermore, a 20 g of sample from each sampling field was kept in a separate plastic bag with a zipper and labelled with a unique sample identity. Sterile gloves were used to prevent contamination during the whole process. Final samples were kept in the refrigerator at −20 °C until complete analysis. Collected soil samples were grouped and labelled one of two ways: i) fields where intensive/conventional farming was practiced (n = 114) and ii) fields where IPM was practiced (n = 33).

2.4. Chemical determination and quality control

Soil samples, labelled as clean by Wageningen Food Safety Research and which did not contain any of the 23 tested pesticides examined (list of pesticides including LODs, see Table 1) were considered as blanks. The analytical determination, chemicals and reagents, instrumentation, and quality assurance of the method are described in Bhandari et al. (2019). The chemical analysis and the quality control were performed as per the European Commission guidelines (SANTE/11813/2017) (EC, 2017). In the case where different LODs of the pesticide were observed, the highest LOD of the pesticide was used as the reporting limit and was considered as the final LOD of the pesticide. The calibration curves showed linearity within the range of 70–120%, with a regression coefficient (R^2) > 0.99. Relative standard deviations were <10%, indicating reliability and accuracy of the method.

Table 1
Comparing mean concentrations of pesticides detected in soils from different farming systems during the growing season (μg kg^{-1}). The concentrations < LODs (na) were excluded when calculating the average values. Dichlorvos, dimethoate, omethoate, phorate, α-β-endosulfan and α-γ-HCH had concentrations < LOD (i.e. <1 μg kg^{-1}) and do not appear in the table. Abbreviations of pesticides are shown in parenthesis. na = not applicable.

Chemical group	Pesticides and degradation products	Type of farming						LODs	p-value
		IPM (N = 33)			Conventional (N = 114)				
		NPS (%)	Min-Max	Mean (SD)	NPS (%)	Min-Max	Mean (SD)		
UNC	3,5,6-trichloro-2-pyridinol[m] (TCP)	na	na	na	36(32)	2.63–57.4	10.4(9.98)	2.5	na
OPs	Chlorpyrifos (CHLPY)	na	na	na	11(10)	10.5–177	40.8(49.35)	10	na
	Profenofos	na	na	na	4 (4)	1.09–3.37	1.75(1.09)	1	na
	Quinalphos	na	na	na	3(3)	1.06–2.47	1.59(0.77)	1	na
	Triazophos (TRIZO)	na	na	na	6(5)	1.05–8.12	3.28(2.73)	1	na
OCs	o, p'-DDT!	na	na	na	3(3)	1.60–4.28	2.85(1.35)	1	na
	p, p'-DDD!	na	na	na	3(3)	1.95–11.1	7.11(4.69)	1	na
	p, p'-DDE!	na	na	na	18(16)	1–13.9	3.31(3.16)	1	na
	p, p'-DDT!	na	na	na	10(9)	1.05–78.4	12.1(24.5)	1	na
AD	Chlorantraniliprole (CHLNITR)	na	na	na	35(31)	1.08–14.2	3.17(2.62)	1	na
NND	Imidacloprid (IMDA)	2(6)	1.02–1.17	1.10 (0.11)	26 (23)	1.16–31.6	5.52(6.52)	1	0.354
BD	Carbendazim	na	na	na	18(16)	1.03–6.45	2.12(1.54)	1	na
PA	Metalaxyl (MA)	3(9)	1.22–3.80	2.19 (1.41)	12(11)	1.12–8.97	3.25(2.4)	1	0.482
UNC	*N*-(2,6-dimethylphenyl)-*N*-(methoxyacetyl) alanine[m] (MMB)	1(3)	na	na	2(2)	1.11–1.56	1.34(0.32)	1	0.207
MOD	Emamectin	na	na	na	1(1)	na	na	1	na

Notation.Chemical group: UNC = Unclassified; OPs = organophosphates; OCs = organochlorines; AD = Anthranilic diamide; NND = Neonicotinoid; BD = Benzimidazole; PA = Phenylamide and MOD = Micro-organism derived NPS = Number of positive samples; m = degradation product; ! = banned pesticides.

2.5. Human health risk assessment

The selected pesticides have been found to induce either cancer or non-cancer diseases (PPDB, 2019) (Table S2, Supplementary information). DDT was categorized as a probable carcinogenic for humans by the International Agency for Research on Cancer (IARC). In 2015, the IARC found positive associations between exposure to DDT and diseases such as non-Hodgkin lymphoma, testicular cancer, and liver cancer (IARC, 2015). In order to assess health risks posed to humans, we adopted USEPA models that have been proven successful and adopted worldwide (Liu et al., 2013). The threshold values of the models and the concentrations of pesticides found in our soils were used to assess human health risks (for the threshold values used in the risk assessment, see Table S3, S4 and S5 in Supplementary information, page 5).

To facilitate the process, the risk assessment process for humans is divided into 3 steps: i) hazard identification (HaI), ii) exposure assessment (ExA), and iii) risk characterization (RiC). HaI involves the identification of pesticides in the study area that can have health risks to humans. The persistence and toxicity of pesticides that have risks were identified based on PPDB (2019). ExA was done for pesticides that posed potential health risks. Cancer and non-cancer risks from pesticides exposure were estimated following USEPA models and hazard quotient (HQ) and hazard index (HI) indices, respectively. The relationship between pesticides concentration and the resulting incidence of impacts was based on mathematical models to determine risks to humans via different exposure pathways such as ingestion, inhalation and dermal contact. RiC incorporates information derived from HaI and ExA. It involves the estimation of health risk due to single and multiple pesticides. Total cancer risk (TCR), HQ and HI were used to characterize risks to human health.

2.5.1. Assessment of cancer risk (CR) for OCs

Exposure to DDTs is potentially linked with CR (Band et al., 2011; Wong et al., 2015). Considering the incidental ingestion, inhalation and dermal contact with contaminated soil, (USEPA, 2018), the chronic (average) daily non-dietary intake (CDI, mg kg^{-1} day^{-1}) of compounds (pp'-DDT, pp'-DDE and pp'-DDD) in adolescents and adults can be estimated using the following equations.

$$CDI_{ing} = \frac{C_{soil} \times EF \times ED \times IR_{ing}}{AT \times BW} \times CF \quad (1)$$

$$CDI_{der} = \frac{C_{soil} \times SA \times SAF \times ABS \times EF \times ED}{AT \times BW} \times CF \quad (2)$$

$$CDI_{inh} = \frac{C_{soil} \times EF \times ED \times IR_{inh}}{PEF \times AT \times BW} \quad (3)$$

where CDI_{ing}, CDI_{der} and CDI_{inh} are the average daily doses via ingestion, dermal contact and inhalation (mg kg^{-1} day^{-1}), respectively.

C_{soil} (mg kg^{-1}) = concentration of pesticides in soil

The details of other parameters/exposure factors such as IR_{ing}, EF, ED, BW, CF, AT, SA, SAF, ABS, IR_{inh}, and PEF are listed in Table S3. The incremental lifetime CR denotes the increasing possibility of humans to get cancer during their lifespan via exposure to a carcinogenic compound. In our study, the CR in adolescents and adults over their lifetime exposure to DDT and its degradation products was calculated following USEPA (2001) and Yadav et al. (2016).

$$CR = CDI \times SF \quad (4)$$

where, SF = carcinogenicity slope factor (details in Table S4) CDI = estimated average chronic daily non-dietary intake (Table S6)

If multiple carcinogenic compounds are present, the total CR (TCR) from all of the compounds and possible routes is calculated following Yadav et al. (2016).

$$TCR = \sum_{k=1}^{n} CRi \quad (5)$$

where, i = different exposure pathways.

In general, TCR values between 1×10^{-6} and 1×10^{-4} are considered to be acceptable, while those exceeding 1×10^{-4} are considered to constitute a lifetime carcinogenic risk to humans. A risk factor $<1 \times 10^{-6}$ is regarded as negligible or no risk (USEPA, 1989).

2.5.2. Assessment of non-cancer risk (NCR)

The NCR was calculated following USEPA (2019), which was also adopted in a past study (Pan et al., 2018). The NCR for the pesticides of interest for a specific exposure route can be expressed as Hazard Quotient (HQ). HQ of a pesticide is the ratio of CDI, and RfD (reference dose) of the pesticide.

$$HQ = \frac{CDI}{RfD} \tag{6}$$

The total NCR of pesticides belonging to OPs via two primary routes such as soil ingestion and dermal contact can be denoted as HI, which was estimated by following Equation (7).

$$HI = \sum_{k=1}^{n} HQi \tag{7}$$

where, HQ_i = hazard quotient of exposure pathway i; RfD (mg kg^{-1} day^{-1}) = daily maximum permissible concentration of OPs, including the reference doses for exposures such as ingestion (RfD_{ing}) and dermal contact (RfD_{der}). The RfD_{ing} = RfD and RfD_{der} = RfD x ABS_{GI}. ABS_{GI} is the gastrointestinal absorption factor (dimensionless). Pesticides belonging to OPs have their common mode of action (Table S1), therefore HI was estimated for only the OPs.

The details of parameters such as RfD and ABS_{GI} used in NCR assessment are listed in Table S5. HQ or HI greater than one shows potential NCR, while HQ or HI ≤ 1 means negligible or no risk. Our study could not estimate the potential NCR via inhalation because reference concentration (RfC) values for the pesticides of interest were not available in the PPDB (2019).

2.6. Predicted environmental concentration (PEC)

PEC is an indicator of the expected pesticide concentrations in soil, taking into account the default values (Ockleford et al., 2017). The PEC was estimated with the default values (otherwise stated) using Equations (8)–(12). For multiple applications of chemical pesticides, the maximum time-weighted average (TWA) concentrations for exposure days of 1, 2, 4, 7, 14, 28, 50, and 100 were estimated using a moving time-frame (MTF) Excel spreadsheet. For a given exposure period, the spreadsheet calculates the TWA concentrations for period starting times ranging from day of first application to day of last application (MTF), and scans for the highest value (EC, 2004).

$$PEC_{s,\ ini} = \frac{A(1 - f_{itn})}{100\ DEPTH_{Soil} bd_{Soil}} \tag{8}$$

$$PEC_{s,act} = PEC_{s,ini} e^{-k_{soil} t} \tag{9}$$

$$PEC_{s,twa} = PEC_{s,ini} \frac{(1 - e^{-k_{soil} t})}{k_{soil} t} \tag{10}$$

$$PCEC_{s,ini,n} = \frac{PEC_{s,ini,1}(1 - e^{-nk_{soil} i})}{(1 - e^{-k_{soil} i})} \tag{11}$$

$$PEC_{s,act,n} = PEC_{s,\mathrm{int},n e^{-k_{soil} t}} \tag{12}$$

where, A = application rate (g/ha); f_{int} = fraction intercepted by crop cover; $DEPTH_{soil}$ = depth of soil (cm); bd_{soil} = bulk density of soil (g cm^{-3}); $PEC_{s,ini,1}$ = initial PECs after one application; n = number of applications; i = application interval (d); k_{soil} = degradation rate in soil (d^{-1}) = ln(2)/half-life.

PECs of pesticides are estimated for the different farms at three different time points (after pesticide application) for each pesticide prediction: the initial PECs (immediately), the short-term PECs (1–4 days) and the long-term PECs (7–100 days). Since the farmers in our study hadn't applied pesticides for a week prior to our first sample measurement, predicted concentrations ($PEC_{s,act,7\ days\ after\ pesticide\ application}$) of pesticides would be suitable for making possible comparisons with their measured environmental concentrations (MECs). Likewise, since farmers followed poor agricultural practices, we used the initial PECs of pesticides in order to compare measurements with the international guidelines of soil quality standard. All the tested pesticides had a 90% degradation time under a year, thus the background concentrations and the PECs that accumulated were not considered in this study. Human health risk (HR) was evaluated by comparing the PECs of the pesticides in soils with international guidelines of soil quality such as pesticide soil regulatory guidance values (PSRGVs), the maximum concentrations of pesticides present in soil without hampering the environmental balance (Li and Jennings, 2017) and Chinese Soil Quality Standard (GB 15618–2018) (MoEP, 2018). Past studies (Wang, 2007; Qu et al., 2015) also followed similar methods for the evaluation of the health risk.

2.7. Statistical analysis

The data analysis was performed using Canoco 5 and SPSS 23. To avoid underestimating soil concentrations of pesticides, only pesticide residue concentrations $\geq$ LODs were considered in data analysis. Concentrations below the LOD were not included in the analysis (Sun et al., 2016). The normality of the data was tested by the Kolmogorov–Smirnov test. In the cases of normal distribution, a one-way ANOVA was used to compare the number of pesticide residues and the total pesticide concentrations in soil between conventional and IPM farming practices, vegetable farms (eggplant, chilli and tomato), and three depths of soil. In case of significant effects at the 95% confidence level ($p < 0.05$), the Bonferroni post hoc test was conducted. Principal component analysis (PCA) and Pearson correlation coefficients were used to study relationships between the pesticide concentrations and the pesticide properties.

3. Results

3.1. Number of pesticides in soil

Pesticide residues analysis in soils revealed the presence of a variety of pesticide combinations. Thirty-nine pesticide combinations were detected in soils. One single pesticide residue was detected in 25% of the soil samples, while multiple residues were present in 35% of the soil samples (Fig. 2A). The number of residues varied significantly with soil depths, vegetable fields, and farming practices (Fig. 2B). Pesticide residues were found less frequently in the depth 35–40 cm and more frequently in the top soil (0–5 cm) ($p = 0.001$). A large number of pesticide residues (up to 7 residues) was detected in 2% of the tested top soils.

The number of pesticide residues detected in soils from eggplant fields was significantly lower than tomato fields ($p = 0.025$; Fig. 2B). Seventy-three percent of soil samples from tomato fields and half of the soil samples from chilli fields contained detectable pesticide residues.

Soils from IPM fields had a significantly smaller number of observed pesticide residues > LOD than the soils from conventional fields ($p < 0.01$; Fig. 2B and LODs in Table 1). Only fifteen percent of the soil samples from the IPM fields contained detectable pesticide residues. On the other hand, about seventy-three percent of soil

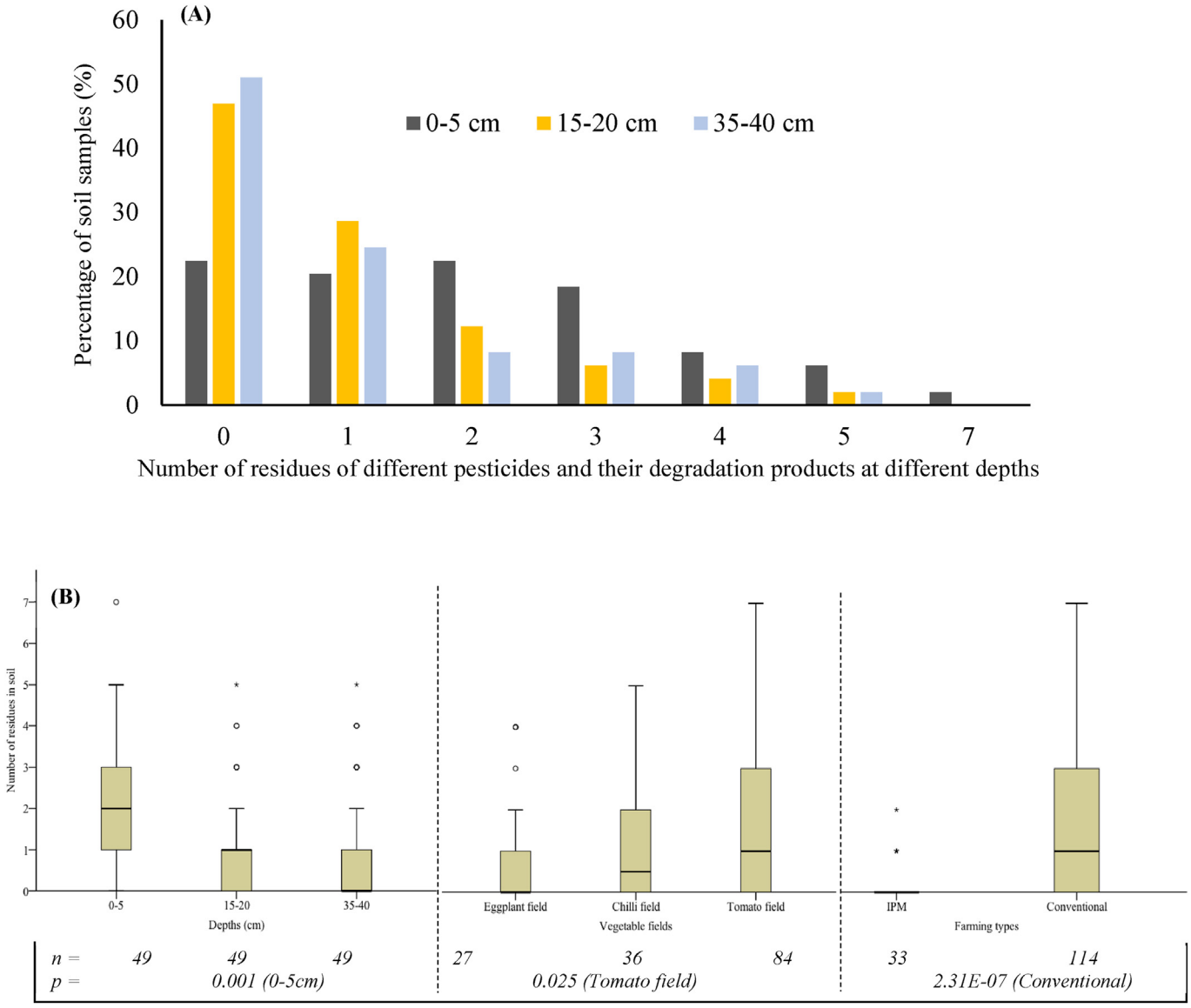

Fig. 2. Number of residues of different pesticides and their degradation products at different soil depths and the percentage of soil samples (A); and box plots for comparison of mean number of residues at different soil depths detected in different vegetable fields of two farming types and the p-values (B).

samples from the conventional fields contained detectable residues of pesticides.

The concentrations of residues of dichlorvos, dimethoate, omethoate, phorate, α-β-endosulfan and α-γ-HCH were < LOD and do not appear in Fig. 3A.

3.2. Types of pesticide residues and their combinations

Fifteen different pesticide residues (approximately 65% of the tested pesticides) were observed in the soils (Table 1). Residues of dichlorvos, dimethoate, omethoate, phorate, α-γ-HCH and α-β-endosulfan were below LOD (<1 μg kg^{-1}). The six most common pesticide mixtures (for abbreviations of pesticides, see Table 1) detected in soils were:

(i) CHLNITR + IMDA
(ii) CHLNITR + CHLPY + TCP + TRIZO
(iii) MA + TCP
(iv) CHLNITR + p,p'-DDE + p,p'-DDT
(v) IMDA + p,p'-DDE and
(vi) o,p'-p,p'-DDTs + p,p'-DDE + p,p'-DDD

Chlorantraniliprole and imidacloprid residues were the most prominent with 11% and 5% of the total soil samples with detectable residue levels, respectively, and found frequently at the soil depth 15–20 cm. The most common pesticide, CHLNITR, was found in 17% of the soil samples from tomato fields. In IPM fields, most soil samples (85%) were residues free however, residues of MA, IMDA, and MA + MMB were detected.

Of all the detected pesticides in our samples, about 60% of the pesticides were non-persistent or moderately persistent compounds. Persistent and very persistent compounds represented about 13% and 27% of the detected pesticides, respectively. Four of the compounds such as o,p'-p,p'-DDTs; p,p'-DDE and p,p'-DDD, detected in soils were degradation products of active substances that are currently banned in Nepal. Overall, eight percent of the soils contained quantifiable residues of DDT and its degradation products.

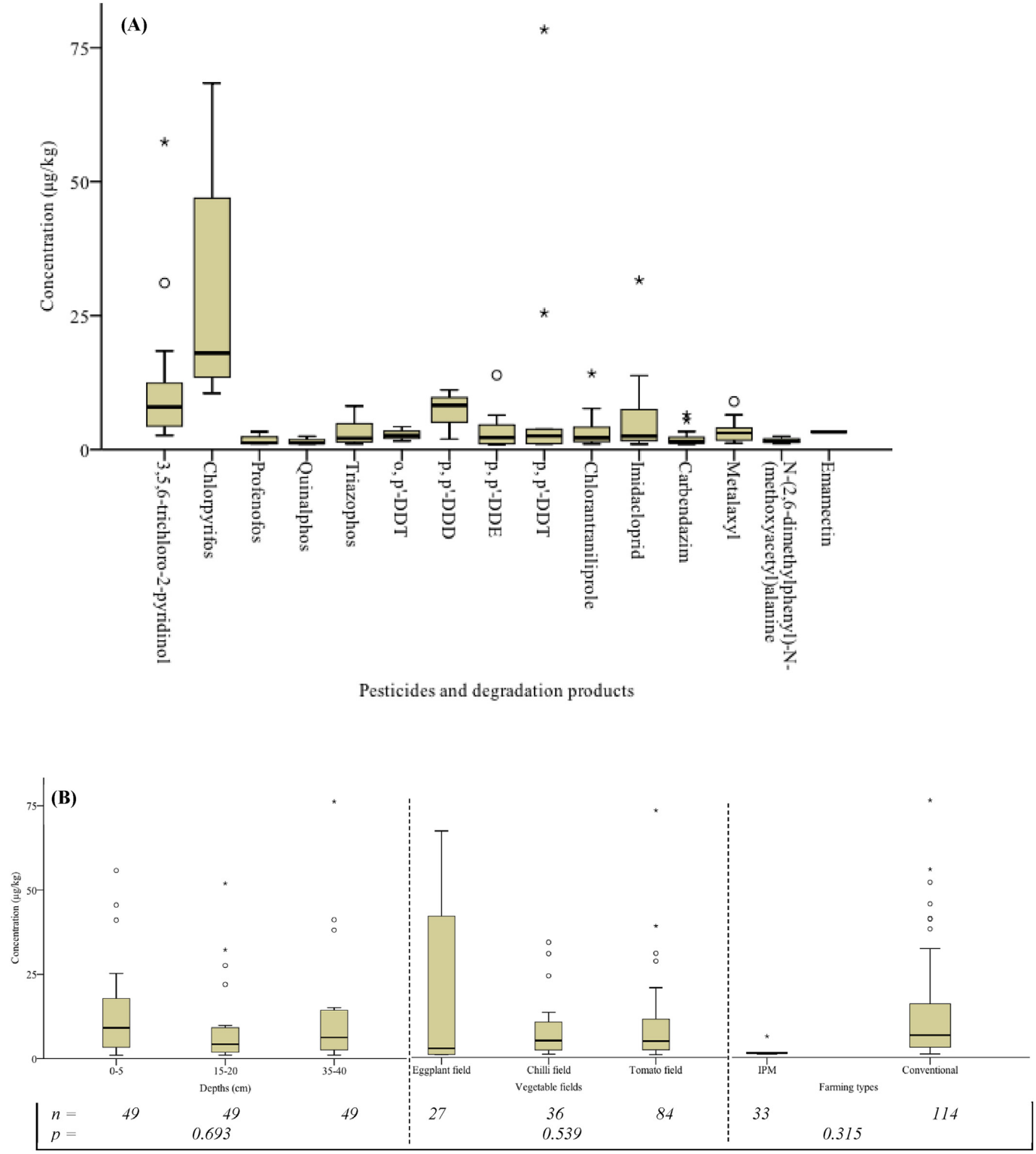

Fig. 3. Distribution of pesticide concentrations and pesticide degradation products in soils (A); and box plots for comparison of mean concentration of pesticide residues at different soil depths detected in different vegetable fields of two farming types and the p-values (B).

3.3. Hazard identification

Our study found 4 organophosphates (chlorpyrifos, profenofos, quinalphos and triazophos), 4 organochlorines (o,p'-DDT, p,p'-DDT, p,p'-DDD and p,p'-DDE), 1 anthranilic diamide (chlorantraniliprole), 1 neonicotinoid (imidacloprid), 1 benzimidazole (carbendazim), 1 phenylamide (metalaxyl), 1 micro-organism derived (emamectin) and 2 unclassified degradation products

[TCP and *N*-(2,6-dimethylphenyl)-*N*-(methoxyacetyl)alanine] in soil samples (Table 1). The mean concentration of pesticides in soil samples was 16.05 μg kg^{-1} with a range of 1.02–251.28 μg kg^{-1}. Total pesticide concentrations in soils differed according to the farming practices, vegetables cultivated, and soil depths as seen in Table 1 and Fig. 3B. A significant correlation existed between total pesticide concentration and farming practices, vegetables cultivated, and soil depth ($p < 0.05$). The identified hazards are the most commonly detected pesticides in soils such as carbendazim, imidacloprid, chlorantraniliprole, p,p'-DDT, p,p'-DDE, metalaxyl, chlorpyrifos, and TCP (Table 1). Except chlorantraniliprole, concentrations of the other pesticides in soils showed relationship with each other, indicating positive or negative correlation (Table 2).

The average concentration of the pesticides was weakly correlated with their properties such as H_2O solubility, DT50, adsorption and mobility, vapour pressure, GUS index, and BCF; while the frequency of detection of the pesticides had a significant positive correlation with GUS index (Table 3). Principal Component Analysis (PCA) was performed to make the results of the table more informative and easily interpretable (Fig. 4). Except p,p'-DDE, all the DDTs made a similar contributions to the OCs and suggested a similar source of origin.

TCP and chlorantraniliprole contributed the most to the total frequency of detection, while chlorpyrifos and p,p'-DDT had the highest pesticide concentration in soils with a maximum concentration of 177 and 78.4 μg kg^{-1}, respectively. These two compounds also pose the highest health risks. The compounds that had lower

Table 2
Correlations between the most common pesticides and degradation products detected in soils (positively correlated in bold font).

Pesticides and degradation products	N	CARBE	IMDA	CHLNITR	p,p'-DDT	p,p'-DDE	MA	CHLPY	TCP
CARBE	18	1							
IMDA	28	−0.527	1						
CHLNITR	35	−0.049	0.422	1					
p,p'-DDT	10	na	−1.000**	0.202	1				
p,p'-DDE	18	**1.000****	0.073	0.038	0.319	1			
MA	15	−0.284	−0.971	0.906	na	na	1		
CHLPY	11	**0.987***	−1.000**	−0.946	na	na	na	1	
TCP	36	**0.949****	0.075	0.667	na	0.41	**0.723***	**0.889****	1

Notation. CARBE = Carbendazim, IMDA = Imidacloprid, CHLNITR = Chlorantraniliprole, MA = Metalaxyl, CHLPY = Chlorpyrifos and TCP = 3,5,6-trichloro-2-pyridinol. "*" and "**" represented significant correlation at the levels 0.05 and 0.01 (both 2-tailed), respectively. "N" = Number of positive soil samples, "na" = Cannot be calculated because at least one of the variables is constant.

Table 3
Correlations between the parameters, frequency and concentration of pesticides detected in soils (positively correlated in bold font).

Parameters of pesticides	N	WS	DT_{50}	K_d	K_{oc}	VP	GUS	BCF	FREQ	AVGC
WS (mg L^{-1})	13	1								
DT_{50}	11	−0.126	1							
K_d	3	−0.985	**0.999***	1						
K_{oc}	12	−0.191	−0.313	**1.000****	1					
VP (mPa)	11	−0.099	0.054	**1.000****	−0.178	1				
GUS index	13	0.143	0.587	−1.000**	−0.921**	0.414	1			
BCF	10	−0.261	**0.654***	0.686	−0.242	−0.21	−0.456	1		
FREQ	15	0.04	0.091	−0.236	−0.387	0.536	**0.593***	−0.348	1	
AVGC	15	−0.133	−0.114	−0.495	−0.11	0.09	0.115	0.389	0.059	1

Notation.WS (mg L^{-1}) = Water solubility, DT_{50} = Half-life, K_d = Soil distribution Coefficients, K_{oc} = Soil adsorption Coefficient, VP (mPa) = Vapour pressure, GUS = Groundwater Ubiquity Score, BCF = Bio-concentration factor, FREQ = Frequency, and AVGC = Average concentration "N" = Number of pesticides corresponding to the parameters "*" and "**" represented significant correlation at the levels 0.05 and 0.01 (both 2-tailed), respectively.

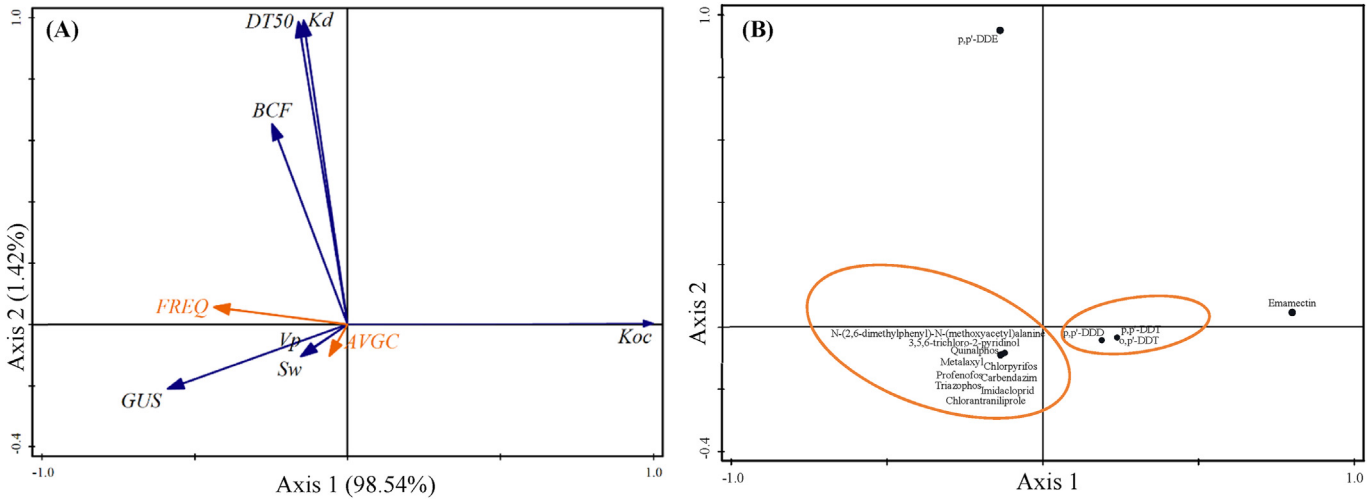

Fig. 4. Loading plots of PCA. Loading plot of PCA (A) showed loadings of the frequency of detection (FREQ) and average concentration of detected pesticides (AVGC) in soil with the pesticide properties such as DT50-soil half-life time (days); K_d-soil distribution coefficient (mL g^{-1}); Koc-organic carbon-water partition coefficient (mL g^{-1}); Sw-H_2O solubility at 20 °C (mg L^{-1}); Vp-vapour pressure at 25 °C (mPa); GUS-leaching potential index; BCF-bio-concentration factor (l kg^{-1}). Loading plot of PCA (B) showed the distribution of sources of different pesticides observed in soil (N = 15).

concentrations such as carbendazim and p,p'-DDE were comparable to those of metalaxyl and chlorantraniliprole, respectively. For all the DDTs, the concentration of p,p'-DDT (40 μg kg^{-1}) and p,p'-DDD (11.1 μg kg^{-1}) was higher in the depth 35–40 cm. Likewise, the concentration of p,p'-DDE (5.70 μg kg^{-1}) and o,p'-DDT (4.28 μg kg^{-1}) was higher in the depth 15–20 cm.

Predicted concentrations ($PEC_{s,act,7\ days\ after\ pesticide\ application}$) of pesticides in soils from tomato, eggplant and chilli farms are presented in the Supplementary information, Table S7. $PEC_{s,act,7}$ of most of the pesticides were much higher than their measured environmental concentrations (MECs). Carbendazim, dichlorvos, imidacloprid and profenofos showed higher initial PECs of pesticides than their global pesticide soil regulatory guidance maximum values, indicating that farmers might be at greater risks from the pesticides (Table S7 and Table S8).

3.4. Exposure assessment for farm workers

The non-dietary chronic daily intake from exposure to p,p'-DDE, p,p'-DDT and p,p'-DDD in soils via non-dietary ingestion, dermal contact, and inhalation are presented in Table S9. Similarly, the total average cancer risks (CR) resulting from exposure to OCs are presented in Table 4. The CR posed by p,p'-DDE, p,p'-DDT, and p,p'-DDD in soils for adults were 1.30E-09, 4.75E-09, and 1.97E-09, respectively, which were slightly lower than those for adolescents (1.38E-09, 5.03E-09, and 2.09E-09, respectively). Likewise, the carcinogenic effects of p,p'-DDT in adults and adolescents was comparable and was higher than that of p,p'-DDE and p,p'-DDD in adolescents and adults, respectively.

3.5. Risk characterization for farm workers

The estimated CR of three OCs such as p,p′-DDE, p,p′-DDT and p,p′-DDD in adolescents and adults via different pathways were found below 1×10^{-6} (Table 4) and showed no CR due to the exposure to the pesticides in soils. Even after considering the TCR as cumulative, the CR for adolescents and adults were below the USEPA bench mark (1×10^{-6}).

The average HI for OPs was <1 and posed negligible NCR. The HI (mean ± SD) estimated for adolescents via dermal contact exposure routes and soil ingestion was 8.02E-05 ± 1.04E-04 and 1.10E-04 ± 1.43E-04, respectively. Likewise, the HI (mean ± SD) estimated for adults via dermal contact exposure routes and soil ingestion was 2.95E-05 ± 3.83E-05 and 5.69E-05 ± 7.38E-05, respectively. The total HIs for adolescents and adults were 1.90E-04 and 8.64E-05, respectively, showing a negligible NCR.

The non-cancer risks of the OPs to farmers via dermal contact pathways and soil ingestion are presented in Fig. S1 and Fig. S2. The HQ and HI (of the total OPs) were found <1 for both adolescents and adults, indicating negligible NCR.

4. Discussion

4.1. Pollution assessment

In general, top soils (0–5 cm) contained higher concentrations and numbers of pesticide residues. However, DDT and its degradation products were less frequently found in the top soils. According to the Chinese standard (GB 15618–2018), risk screening value for the total DDT is 100 μg kg^{-1}. Likewise, Ma et al. (2016) classified soil into (i) negligible contamination, with DDT concentration <50 μg kg^{-1}; (ii) lower contamination, with DDT concentration 50–500 μg kg^{-1}; (iii) medium contamination, with DDT concentration 500–1000 μg kg^{-1}, and (iv) higher contamination, with DDT concentration >1000 μg kg^{-1}. Although DDT has been banned in Nepal since 2001, we found DDT concentration <50 μg kg^{-1} in 99% of the total soil samples.

Farmers applied most of the pesticides with higher application rates, greater numbers of applications and shorter application intervals (Bhandari et al., 2018). This might have contributed to higher predicted environmental concentrations (PECs) of pesticides (see Table S7 in Supplementary information). The PECs after 7 days of pesticide application ($PEC_{s,act,7}$) for most pesticides, except for dichlorvos and emamectin, were much higher than their measured concentrations (Table 1 and Table S7, Supplementary information). In the same study, they applied dimethoate and dichlorvos at higher than their recommended levels. However, soils were free from pesticide residues such as dimethoate and its degradation product (omethoate), dichlorvos, phorate, α-β-endosulfan and α-γ-HCH (<LOD). Dimethoate, omethoate and dichlorvos have higher water solubility (Table S1) and phorate, α-β-endosulfan and α-γ-HCH have been banned for many years which may explain why their residues were absent in the soils. Soils from conventional systems had significantly higher numbers of pesticide residues than the soils from IPM farming. In the IPM farms, about 85% of the soil samples were clean, and the remaining samples had pesticide concentrations close to their corresponding LODs (Table 1). In an another study, we observed higher residues of pesticides in vegetables from conventional farms than that of IPM farms (Bhandari et al., 2019).

The fate of pesticides in soils is determined by their various factors: mobility, persistence, and volatility. Furthermore, other pesticide properties such as phosphorus and nitrogen levels, organic carbon content, and soil pH affect distribution and occurrence (Gong et al., 2004; Pan et al., 2018). Few of the pesticides detected in soils have lower soil organic carbon-water partitioning coefficients (Koc) and thus, moderate leaching potential which suggests a risk of ground water pollution. The sorption of chemical pesticides was the highest for the soils with greatest OC content (Zbytniewski and Buszewski, 2002).

Conventional fields contain less OC than IPM fields and this might enhance the mobility of pesticides and could thus increase groundwater pollution (Sánchez-González et al., 2013). About ninety percent of total inhabitants in the study area drink water from tube wells adjacent to their vegetable fields (GRM, 2018) which could increase their HR. Pesticides may contaminated groundwater and make it unsuitable for drinking, which is the case for Nigeria (Sosan et al., 2008) and the Philippines (Castaneda and Bhuiyan, 1996). The mean concentrations of the pesticides in our soil samples are at lower end in comparison to other countries (Table 5).

Depending on the date of pesticides application, the $PEC_{s,act,7}$ were correct for only dichlorvos and emamectin. However, the PECs of other pesticides in different fields (see Table S7 in

Table 4
The cancer risks (CR) of adolescents (adol) and adults (adul) resulting from the pesticides exposure in soils.

Pesticides	CR- (adol- ing)	CR- (adol- der)	CR- (adol- inh)	CR- (adul- ing)	CR- (adul- der)	CR- (adul- inh)	TCR(adol)	TCR(adul)
p,p′-DDD	1.02E-09	1.06E-09	1.33E-13	1.13E-09	8.38E-10	1.46E-13	2.09E-09	1.97E-09
p,p′-DDE	6.75E-10	7.03E-10	8.79E-14	7.47E-10	5.54E-10	9.61E-14	1.38E-09	1.30E-09
p,p′-DDT	2.46E-09	2.56E-09	3.21E-13	2.73E-09	2.02E-09	3.51E-13	5.03E-09	4.75E-09

Table 5
Comparison of pesticide levels in soils from conventional farming in this study with past studies across the globe. To find the relevant literature, Web of Science database was considered by using the search phrase pesticide and soil and *concentration*. Hyphens indicate that no information was available. Pesticides concentration in μg kg^{-1}. The mean concentration of the most pesticides in this study is lower than the other studies abroad.

Pesticides	Abroad		Nepalese agricultural soil
	Place, country/land use/mean concentration	Reference	Mean concentration (this study)
3,5,6-trichloro−2-pyridinol	–	–	10.4
Chlorpyrifos	Okara, Pakistan/cotton, wheat/1393	Rafique et al. (2016)	40.8
	China/persimmons and jujubes/17.15	Liu et al. (2016)	
	China/nuts/42.2	Han et al. (2017)	
	Dormaa West, Ghana/cocoa/30	(Fosu-Mensah et al., 2016)	
Profenofos	Okara, Pakistan/cotton, wheat/89.79	Rafique et al. (2016)	1.75
	Dormaa West, Ghana/cocoa/30	(Fosu-Mensah et al., 2016)	
Quinalphos	–	–	1.59
Triazophos	Okara, Pakistan/cotton, wheat/99.74	Rafique et al. (2016)	3.28
o,p′-DDT	Nagaon, India/paddy fields, tea gardens and others/150	Mishra et al. (2012)	2.85
	Hong Kong/different types of land use/0.05	Zhang et al. (2006)	
	Shanghai, China/agriculture/1.66	Jiang et al. (2009)	
	Beijing, China/school yards/42.38	Wang et al. (2008)	
	Moldavia, Romania/forest/0.7	Tarcau et al. (2013)	
p,p′-DDD	Limuru, Kenya/rural and semi urban areas/1.71	Sun et al. (2016)	7.11
	Nagaon, India/paddy fields, tea gardens and others/73	Mishra et al. (2012)	
	Hong Kong/farmland/0.05	Zhang et al. (2006)	
	Shanghai, China/agriculture/4.56	Jiang et al. (2009)	
	Beijing, China/school yards/6.47	Wang et al. (2008)	
	Moldavia, Romania/forest/1.2	Tarcau et al. (2013)	
p,p′-DDE	Limuru, Kenya/rural and semi urban areas/0.97	Sun et al. (2016)	3.31
	Nagaon, India/paddy fields, tea gardens and others/276	Mishra et al. (2012)	
	Hong Kong/farmland/1.73	Zhang et al. (2006)	
	Shanghai, China/agriculture/16.14	Jiang et al. (2009)	
	Beijing, China/school yards/27.29	Wang et al. (2008)	
	Moldavia, Romania/forest/10	Tarcau et al. (2013)	
p,p′-DDT	Limuru, Kenya/rural and semi urban areas/11.76	Sun et al. (2016)	12.1
	Nagaon, India/paddy fields, tea gardens and others/351	Mishra et al. (2012)	
	Hong Kong/farmland/0.02	Zhang et al. (2006)	
	Shanghai, China/agriculture/3.26	Jiang et al. (2009)	
	Beijing, China/school yards/17.54	Wang et al. (2008)	
	Moldavia, Romania/forest/8.1	Tarcau et al. (2013)	
Chlorantraniliprole	–	–	3.17
Imidacloprid	Okara, Pakistan/cotton, wheat/548.7	Rafique et al. (2016)	5.52
Carbendazim	Basrah, Iraq/agricultural soil/1259	Raheem et al. (2017)	2.12
Metalaxyl	Spain/agricultural areas/3.82	Sánchez-González et al. (2013)	3.25
N-(2,6-dimethylphenyl)-*N*-(methoxyacetyl)alanine	-	-	1.34
Emamectin	–	–	–

Supplementary information) were much higher than their MECs (measured environmental concentrations or the mean concentration) (Table 1) and pesticide soil regulatory guidance values (Table S8, Supplementary information). The farmers' pesticide use behaviours such as the application rates were self-reported and observed higher than recommended (Bhandari et al., 2018). The differences between MECs and PECs are several orders of magnitude that might be due to the estimation of PECs from the realistic worst-case scenario. Since PSRGVs might be risk-based, the values could more accurately reflect the potential environmental and health risks which are worth consideration. In addition, immediate predicted environmental concentrations ($PEC_{s,act,0}$) of contaminated soils and their effects on human health should not be neglected.

Our predicted environmental concentration in soil (PEC_s) for dimethoate on tomato after multiple applications was much higher than their respective values in the draft assessment report (DAR) of the European Commission (EC, 2004). The possible reason for higher PEC_s for dimethoate might be due to the fact that the pesticide was applied at level higher than its recommended dose (Bhandari et al., 2018). Likewise, the PEC_s for other pesticides from eggplant and chilli farms could not be compared due to unavailability of their DARs.

4.2. Source identification of DDT

DDT is a mixture of its degradation products: 15% o,p'-DDT and 85% p,p'-DDT (Zheng et al., 2009), and the half-life has been estimated >15 years in the environment. Parent DDT disintegrates to DDE and DDD, more stable compounds than their parent. The ratio o,p'-DDT/p,p'-DDT is used to differentiate dicofol from DDT. The ratio between 0.2 and 0.3 corresponds to the occurrence of technical DDT, while the ratio between 1.9 and 9.3 or higher corresponds to the presence of dicofol (Qiu et al., 2005). In our study, the ratio ranged between 0.03 and 0.17, except in one sample, signifying the application of technical DDT. One sample showed the ratio comparatively higher which corresponds with dicofol use in the area. The ratio (p,p'-DDE + p,p'-DDD)/p,p'-DDT assesses the time and degree of disintegration of p,p'-DDT in soil (Qiu et al., 2004). Ratios greater than one indicate aged mixtures, while ratios <1 indicate fresh applications of the parent DDT in soil. In our study, the ratios ranged from 0.21 to 2.20. The ratio was less than one in two samples, specifying the ongoing use of DDT and the ratio was >1 in another sample, indicating its historical use (Dhimal et al., 2014). The current use of DDT might be due to conventional farming and/or expansion of diseases such as malaria fever and dengue (Shah et al., 2012; Awasthi et al., 2017). Similar findings have been reported from other regions of Nepal (Yadav et al., 2016;

Yadav et al., 2017). Potential source analysis indicated that DDT and related compounds mainly originated from a recently applied DDT, possibly due to: (i) the illegal entry due to the porous India-Nepal border; (ii) inadequate execution of the ban and/or (iii) application of DDT for dengue control.

DDT disintegrates to DDD from anaerobic degradations while it changes to DDE from aerobic degradations. The ratio DDD/DDE indicates whether DDT is degraded aerobically or anaerobically. In our study, the ratio DDD/DDE ranged from 0.30 to 2. The ratio was less than one in a higher number of samples, indicating higher percentages of DDE than DDD and thus, DDT was aerobically degraded. Our results were different from those in soils from China (Ma et al., 2016), where the ratio was >1 in a higher number of samples, indicating higher percentages of DDD than DDE and thus, DDT was anaerobically degraded. The disintegration of DDT-DDE-DDD can occur directly or indirectly (Wenzel et al., 2002). The ratios of DDE:DDT, DDD:DDE, and DDD:DDT decide dechlorination paths in soils. DDT to DDE was the major disintegration route, as the ratios were: DDE:DDT (1.73) > DDD:DDE (0.96) > DDD:DDT (0.33). These results coincide with Zhang et al. (2006), but differed with Ma et al. (2016). This differences can be explained by dissimilarities in the precipitation, temperature, humidity, soil moisture, soil texture, microbes, CEC, and OM, which affect the conversion of DDT into DDE-DDD (Aislabie et al., 2010; Chattopadhyay and Chattopadhyay, 2015).

PCA estimates the source and disintegration behaviour of pesticides (Yang et al., 2012). In our study, pesticides belonging to the groups such as organophosphates, anthranilic diamide, neonicotinoid, benzimidazole, phenylamide and unclassified degradation products were aligned together indicating similar source and degradation behaviour, while OCs were separated, suggesting different source and fate of the pesticides.

4.3. Carcinogenic and non-carcinogenic risk

TCR and CR via dermal and ingestion pathways of exposure to DDTs for adolescents and adults were below 1×10^{-6}, indicating negligible cancer risk (Table 4). Adolescent and adult exposure to single non-carcinogenic pesticides (HQ) and multiple pesticides (HI) was <1, suggesting no appreciable non-cancer health risk. Likewise, HIs of pesticides via ingestion and dermal exposure for adolescents and adults were also negligible (<1) (Fig. S2 and Fig. S3). However, other pathways of exposure such as inhalation could still exist in Nepal and cannot be excluded for a non-cancer risk assessment. The risk via inhalation was not considered in this study because essential parameters were unavailable. Furthermore, metabolism and excretion of pesticides in humans were excluded from this study. All soil samples came from farmers' fields close to their houses thus, children may have had direct contact with these soils on a daily basis.

Overall, the cancer and non-cancer risks of pesticides for adolescents were relatively higher than those for adults. Previous studies (Landrigan and Goldman, 2011; Pan et al., 2018) also indicated relatively higher risks for children than adults. The possible reason for higher risks for adolescents might be due to their higher exposure to given doses of OCs and OPs. The soil ingestion was the main pathway of OP exposure and added to 58% and 66% of the total risks in adolescents and adults, respectively [Fig. S2 (b) and Fig. S3 (b), Supplementary information]. Even though the soil samples of GRM were contaminated with pesticides, a negligible health risk from the exposure to the pesticide contaminated soil was observed in this study.

This study considered the worst case scenario (only positive samples and their total average concentrations): replacing the non-detects with 0 (Yadav et al., 2016) would even further decrease the CR, HQ and HI values. However, children are more likely to unintentionally ingest significant amounts of contaminated soil because of their childish behaviour such as putting contaminated hand or fingers in their mouths (Rasmussen et al., 2001). Henceforth, this study warrants further research to investigate the implications of exposure for children through all of the possible pathways.

4.4. Limitations and future recommendations

Pesticide residues could move from neighbouring fields via water and wind and be deposited in surrounding environments (Silva et al., 2018) and accumulate in higher concentrations on the topsoil (1–2 cm) than deeper soil (Yang et al., 2015). Future research should consider soil samples and the distribution of residues in the topmost surface layer. We used conservative risk assessment methods that are generally used for risk assessment of contaminated sites and their applications in farmland needs further research. The PECs of pesticides in soils were based on information related to the pesticide application history in our earlier study (Bhandari et al., 2018) and thus, the results may not be representative of other areas and the latest pesticides use statistics. The twenty-three prioritized pesticides and degradation products assessed in Nepalese soil correspond to <20% of the active ingredients imported for use, indicating that the total pesticides in soils might even be higher than detected in our study and the pesticide mixtures may even be more complicated. Reference concentrations (RfC) of the pesticides and degradation products for the estimation of inhalation exposure were not available thus, risks due to the exposure to pesticides could not be estimated. Worldwide PSRGVs (Table S8) were not calculated comprehensively in humans (Li and Jennings, 2017) and comparison of the PECs with the global values for pesticides in soil may be inadequate for the assessment of HR. Further, whether or not the PECs are reasonable to evaluate the risk of pesticides in the area compared with the PSRGVs could not be answered. Despite such limitations, we have used widely accepted models and indices for the risk assessment.

5. Conclusion

Pesticides applied to vegetables farming in Nepal pollutes soils. Adoption of IPM techniques could reduce pesticide pollution in soils, as this study showed a notably smaller number of pesticide residues and their minimum concentration in the soil samples collected from IPM fields, compared to conventional farming. OCs concentration were sufficiently low in most soil samples (<LOD). However, DDTs were detected with p,p'-DDE being the predominant compound. There is no appreciable health risk from pesticides residues in soils, based on direct dermal contact and/or ingestion in adults or adolescents. The focus should be placed on DDT pollution and the recommendations from the United Nations treaty, the Stockholm Convention should be implemented. A few pesticides detected in soils have a potential of leaching thus, there is a risk of ground water pollution.

Predicted environmental concentrations (PEC_s) for most of the frequently applied pesticides used on vegetables in Nepal did not appear in the European Commission (EC) draft assessment reports thus, the estimated PECs is of minimal use. The PECs 7 ($PEC_{s,act,7\ days\ after\ pesticide\ application}$) for almost all of the pesticides were much higher than their measured environmental concentrations (MECs). The initial PECs of carbendazim, dichlorvos, imidacloprid and profenofos were much higher than their guidance values in soil. The PECs scenario based on the poor agricultural practices is insufficient to claim an increasing health risk of farm workers which warrants future research on PECs and health risk from pesticides in soils from other locations.

Declaration of competing interest

None.

CRediT authorship contribution statement

Govinda Bhandari: Conceptualization, Methodology, Software, Writing - original draft. **Kishor Atreya:** Writing - review & editing, Supervision, Conceptualization. **Paul T.J. Scheepers:** Writing - review & editing. **Violette Geissen:** Writing - review & editing, Supervision.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.chemosphere.2020.126594.

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