SJSS
SPANISH JOURNAL OF SOIL SCIENCE

A preliminary study of the content and distribution of pesticide residues in soil samples from the Kathmandu valley, Nepal

Estudio preliminar sobre el contenido y distribución de residuos de pesticidas en suelos del valle de Katmandú, Nepal

Estudo preliminar sobre o teor e distribuição de resíduos de pesticidas em amostras de solos do vale de Kathmandu, Nepal

AUTHORS

Giri N. [@ 1]
nir_ita@hotmail.com

Blum W. E. H. [1]

Sieghardt M. [2]

Lesueur C. [3]

Mentler A. [1]

[@] Corresponding Author

[1] Institute of Soil Research, University of Natural Resources and Life Sciences (BOKU), Peter Jordan Strasse 82. 1190, Vienna Austria.

[2] Institute of Forest Ecology, University of Natural Resources and Life Sciences (BOKU), Peter Jordan Strasse 82. 1190, Vienna, Austria.

[3] Lebensmittel Vertrauen Analysen (LVA) GmbH, Magdeburggasse 10. 3400 Klosterneuburg, Austria.

Received: 13.08.2012 | **Revised:** 23.10.2012 | **Accepted:** 26.10.2012

ABSTRACT

The increasing use of pesticides for agricultural production is causing soil pollution problems in different parts of Nepal. Uncontaminated agricultural soils are of great importance as they have a direct impact on food security and human health. The objective of this study was to investigate the quality and quantity of pesticides in soil samples from districts near the capital city of Kathmandu, from where fruit and vegetables are brought to the city for consumption. A questionnaire survey was carried out in four districts around Kathmandu city to investigate the types of pesticides that are most commonly used in these districts. A total of 15 soil samples were taken at a depth of 10 cm and four complete soil profiles were sampled at three different depths (10 cm, 30 cm and 50 cm) on the farms of those who were interviewed. A total of four replicates of each soil sample were extracted and analyzed. The pH, soil texture and organic carbon content of the soil samples were analyzed to understand the general soil characteristics. The QuEChERS method used for the analysis of food samples was modified and applied to the soil samples. An HPLC-MS/MS was used for the qualification and quantification of the pesticide residues in the soil samples. The questionnaire survey revealed that carbendazim, chlorpyrifos-methyl, parathion-methyl, imidacloprid, metalaxyl, dimethoate, omethoate and dichlorvos were the most commonly used pesticides in the area studied. The chemical analysis showed that soil samples from all the districts except Kathmandu city were contaminated with various pesticides. The soil samples collected at the depth of 10 cm were found to be contaminated with the fungicide carbendazim and the insecticide chlorpyrifos-methyl at rates of up to 0.038 mg kg^{-1} and the systemic insecticide imidacloprid was found at up to 0.016 mg kg^{-1}. The study of soil samples taken at different depths (10 cm, 30 cm and 50 cm) showed that pesticides were homogenously distributed with soil depth. The recovery between 77.5-112%, linearity between 0.01 mg kg^{-1} - 2 mg kg^{-1} with correlation factors R^2 higher than 0.99 and LOQ between 0.2 µg kg^{-1}- 6.25 µg kg^{-1} were found. Some samples were contaminated with parathion-methyl, a highly carcinogenic organophosphorous insecticide, even though these pesticides had already been banned. For the first time, this study provides information about soil contamination levels due to pesticides in Central Nepal and shows that further research and information campaigns for farmers are necessary.

RESUMEN

El uso creciente de pesticidas para la producción agrícola está ocasionando problemas de contaminación de suelos en diferentes partes de Nepal. Los suelos agrícolas no contaminados son de gran importancia puesto que tienen un impacto directo sobre la seguridad alimentaria y la salud humana. El objetivo de este estudio fue investigar la calidad y cantidad de pesticidas en muestras de suelo de distintos distritos cercanos a la capital de Nepal, Katmandú, que abastecen de fruta y verdura a esta ciudad. Se llevó a cabo un cuestionario en cuatro distritos de los alrededores de

DOI: 10.3232/SJSS.2012.V2.N3.03

Katmandú para evaluar los tipos de pesticidas de uso más común en ellos. Se tomaron 15 muestras de suelo a una profundidad de 10 cm y se muestrearon cuatro perfiles de suelos a tres profundidades distintas (10, 30 y 50 cm) en las granjas de las personas encuestadas. Se tomaron cuatro réplicas de cada muestra de suelo, analizando pH, textura y contenido en carbono orgánico. Para el análisis de las muestras de suelos se utilizó una modificación del método QuEChERS aplicado a alimentos y la caracterización y cuantificación de los residuos de pesticidas en suelos se llevó a cabo por HPLC-MS/MS. El cuestionario realizado reveló que los pesticidas más comunes utilizados en el área de estudio eran carbendazim, chlorpyrifos-methyl, parathion-methyl, imidacloprid, metalaxyl, dimethoate, omethoate y dichlorvos. El análisis químico mostró que todas las muestras de suelo de todos los distritos estaban contaminadas con varios pesticidas excepto las procedentes de la ciudad de Katmandú. Las muestras de suelo a 10 cm estaban contaminadas con el fungicida carbendazim y el insecticida chlorpyrifos-methyl en valores de hasta 0,038 mg kg^{-1}. El insecticida sistemático imidacloprid se encontró en concentraciones de hasta 0,016 mg kg^{-1}. El estudio realizado con suelos muestreados a varias profundidades (10, 30 y 50 cm) mostró que los pesticidas analizados estaban distribuidos homogéneamente a lo largo de todo el perfil. Se obtuvo una recuperación entre el 77,5% y 112%, con una linearidad entre 0,01 y 2 mg kg^{-1} con factores de correlación R^2 superiores a 0,99 y un LOQ entre 0.2 μg kg^{-1} y 6.25 μg kg^{-1}. Algunas muestras de suelo estaban contaminadas con parathion-methyl, un insecticida organofosforado con alto poder cancerígeno, a pesar de que su uso está prohibido. Este trabajo representa el primer estudio realizado sobre niveles de contaminación en suelos debidos a pesticidas en la zona de Nepal Central y muestra que son necesarios futuros estudios y campañas informativas para los agricultores de estas áreas.

KEY WORDS
Agricultural soils, contamination, QuEChERS method, LC-MS/MS, food security

PALABRAS CLAVE
Suelos agrícolas, contaminación, método QuEChERS, LC-MS/MS, seguridad alimentaria

PALAVRAS-CHAVE
Solos agrícolas, contaminação, método QuEChERS, LC-MS/MS, segurança alimentar

RESUMO

A utilização crescente do uso de pesticidas em produção agricola tem conduzido a situações de poluição em diferentes zonas do Nepal. Os solos agrícolas não contaminados desempenham um importante papel uma vez que têm um impacto direto na segurança alimentar e saúde humana. O objetivo deste trabalho foi investigar a qualidade e quantidade de pesticidas em amostras de solo dos distritos vizinhos da capital, cidade de Kathmandu, locais onde têm origem as frutas e legumes consumidos na capital. Foi realizado um inquérito em quatro distritos em redor da cidade de Kathmandu para investigar os tipos de pesticidas que são mais comumente usados. Recolheu-se um total de 15 amostras de solo com uma profundidade de 10 cm e amostraram-se nas fazendas dos agricultores entrevistados quatro perfis de solo completos a três profundidades diferentes (10 cm, 30 cm e 50 cm). Extraíu-se e analisou-se um total de quatro repetições de cada amostra de solo. Determinou-se o pH, textura do solo e teor de carbono orgânico nas amostras de solo analisadas para uma melhor compreensão das características gerais do solo. O método QuEChERS utilizado para a análise de amostras de alimentos foi modificado e aplicado para a análise dos solos. Para a qualificação e quantificação dos resíduos de pesticidas nas amostras de solo usou-se a HPLC-MS/MS. O questionário revelou que o carbendazim, cloropirifos-metilo, paratião-metilo, imidacloprid, metalaxil, dimetoato, ometoato e diclorvos eram os pesticidas mais utilizados na área estudada. A análise química demonstrou que as amostras de solo de todos os distritos, exceto as da cidade de Kathmandu, estavam contaminadas com vários pesticidas. As amostras de solo recolhidas a uma profundidade de 10 cm apresentavam contaminação com o fungicida carbendazim e com o insecticida clorpirifos-metilo até valores de 0,038 mg kg^{-1} e com o inseticida sistémico imidaclopride até 0,016 mg kg^{-1}. O estudo de amostras de solo a diferentes profundidades (10 cm, 30 cm e 50 cm) demonstrou que ocorreu uma distribuição homogénea dos pesticidas com a profundidade de solo. Foi encontrada uma recuperação entre 77,5-112%, linearidade entre 0,01 mg kg^{-1} - 2mg kg^{-1} com fatores de correlação R^2 superiores a 0,99 e LOQ entre 0,2 μg kg^{-1}-6,25 μg kg^{-1}. Algumas amostras foram contaminadas com paratião-metilo, um inseticida organofosforado altamente cancerígeno, embora estes pesticidas já tenham sido retirados do mercado e proíbido o seu uso. Este estudo forneceu pela primeira vez informações sobre os níveis de contaminação do solo devido a pesticidas no Nepal Central e mostra a necessidade de uma investigação mais profunda bem como de realização de campanhas de informação junto dos agricultores.

1. Introduction

The increasing production of food to fulfill the demands of a growing population throughout the world has resulted in the widespread use of pesticides. Some pesticides are highly persistent; they can last for many years before breaking down. These persistent substances can be highly mobile and are capable of bioaccumulation. They circulate globally: although they are released in one region, they can be easily transported through the atmosphere and to regions far away from the original source by repeated processes of evaporation and deposition (Williams 2000).

Organophosphorous insecticides like chlorpyrifos and chlorpyrifos-methyl are the pesticides that are most commonly used throughout the world – organophosphorous pesticides and their metabolites are still detected in the environment although several of them have been banned for several years (Kreuger et al. 2006). About 900 chemical pesticides are still used worldwide, both legally and illegally, with various food products and for the treatment of crops and soils (Thurman et al. 2008). The level of pesticide application for agricultural production is even more severe in developing countries due to efforts to eradicate insect borne diseases, to protect farms and forests and to produce adequate food (Schumann 2005). For several years, organophosphorous pesticides and pyrethroids have been widely used in agriculture, especially in developing countries. Although the use of organophosphorous pesticides and pyrethroids increases crop production, their usage has a severe negative impact on the environment (Wang et al. 2008).

Like any other country in the world, Nepal is confronted with the problems of extensive pesticide use and food security (Baker and Gyawali 1994; Palikhe 2002; Upadhyaya 2002). According to Dahal (1995), chemical pesticides were introduced into this country as early as 1955 when Paris Green, Gamaxone, and nicotine sulfates were imported from the United States of America (USA) for malaria control. Dichlordiphenyltrichlorethane (DDT) made its first impact in Nepal in 1956. This was soon followed by a variety of other organochlorine pesticides in the 1950s, organophosphorous pesticides in the 1960s, carbamates in the 1970s, and synthetic pyrethroids in the 1980s. The most commonly used pesticides in Nepal are malathion, chlorpyrifos-methyl, cypermethrin, deltametrin, mancozeb, parathion-methyl, fenvelarate, dichlorvos, endosulfan sulphate, dimethoate and carbendazim (Palikhe 2001). Many misuses have been reported generally from farmers who do not realize the extent to which pesticides are poisonous and hazardous to humans and the environment. Farmers and retailers of pesticides do not have adequate knowledge regarding pesticide use and health safety (Giri 1998; Baker and Gyawali 1994; Dahal 1995). Furthermore there are no strict government control mechanisms to control the purchase, trading, import and export of pesticides. A significant proportion - ranging from 20 to 70%- of an applied pesticide or its associated degradation products may remain in the soil as a persistent residue bound to soil colloids (Miglioranza et al. 2003). The presence of contaminants in agricultural soils above a certain level entails several negative consequences for food production and for the agricultural ecosystem. Through food chain and other pathways, like inhalation, pollutants can be accumulated within the human body and have an adverse impact on human health (Tang et al. 2010). There is a potential risk of pesticide contamination of ground waters and river systems due to their haphazard use. Chemicals seeping into shallow wells may quickly appear, but could possibly be cleared away within couple of years of treatment. Even so, in deep wells, they may still appear after a very long time because of the percolation time in soils. Such contamination cannot be easily treated and it can take many years for the chemicals to degrade if they remain distant from the active microbial zone (Khanal 2012). Areas with sandy soils face a higher risk of contamination than clayey soils due to easier water percolation (Engle et al. 1993). Carelessness during tank filling, mixing, spraying, re-filling, hand washing, tank emptying and cleaning, the disposal of pesticides near ground water sources like dug wells and tube wells, and/or deep boring, etc., may pose a serious threat.

Due to the lack of technical, financial and trained human resources required for the monitoring and analysis of pesticide residues in agricultural soils and food products, there is no up- to- date database on the pesticide concentrations in different parts of Nepal. Occasional tests for pesticides carried out by the Central Food Research Laboratory affiliated to the Nepalese Government showed contaminated fruit and vegetables brought to Kathmandu markets (Poudel 2011). This means that city dwellers depending on food products grown in peri-urban and rural parts all over the country unknowingly consume pesticides. A systematic study of agricultural soil pollution in different parts of the country is essential in order to elucidate the extent of contamination due to the use of haphazard pesticides and to assess potential risks for the health of local residents and the security of the agricultural products. In this regard, it was of utmost importance to conduct a study in these areas to investigate the types of pesticides used for agricultural production and their residue levels in soil samples. The main objective of this study was therefore to investigate the quality and quantity of pesticides found in soil samples taken from districts neighboring the capital city of Kathmandu, from where fruit and vegetables are brought to market and thereby to quantify the risk with both a questionnaire survey and sample analyses. This study could be considered a first survey conducted on a pilot scale in order to detect the magnitude of pesticide pollution in the soils of the Kathmandu valley.

2. Materials and Methods

2.1. Questionnaire survey

A total of four districts, Kathmandu (Kirtipur I & II and Kathmandu I & II), Lalitpur (Lalitpur I & II and Godavari), Bhaktapur (Thimi I & II and Bhaktapur) and Dhading (Chattradeurali I, II & III and Dhading besi I & II) were selected as study sites (**Figure 1**). From these districts, fruits and vegetables are brought to Kathmandu markets every morning. A multiple choice questionnaire survey was carried out with thirty farmers from the selected sites at the end of September 2008. The main objective of the questionnaire survey was to gather information about the types of pesticides used by farmers to be used as baseline data for the purposive soil sample collection and laboratory analysis.

2.2. Sample collection

The soil samples collected on the farms of interviewed farmers were randomized across the field. A total of fifteen soil samples were collected at a depth of 10 cm. In order to determine the distribution of pesticide residues at different soil depths, a total of four soil profiles from Godavari, Lalitpur district were collected from the depths of 10 cm, 30 cm and 50 cm. About 2 kg of each soil sample was collected and then packed in a clean plastic bag and stored at -18 ^{0}C until analysis. The extraction and analyses of soil samples were carried out at the Institute of Soil Research, University of Natural Resources and Life Sciences (BOKU), Vienna, Austria.

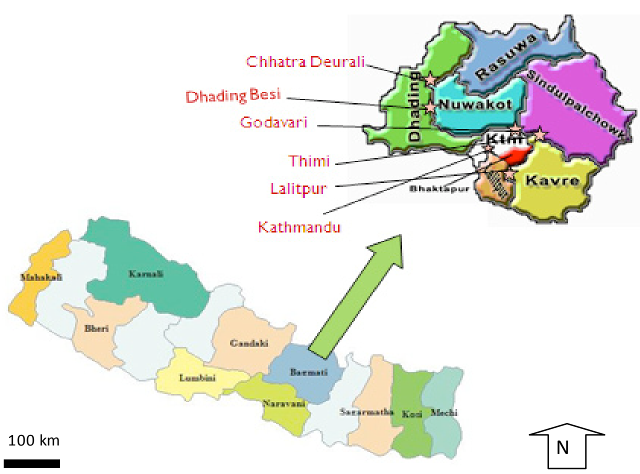

Figure 1. The map of Nepal showing the sampling points in the studied sites.

2.3. General soil characteristics

The soil texture, pH and Organic Carbon (*Corg*) were analyzed according to the methods developed by Austrian Standards Institute (OENORM L 1061, OENORM L 1083 and OENORM L 1080). The pH was analyzed after extraction with water in the ratio of 1:5 for 2 hours. The C_{org} was analyzed by using Carlo Erba NA 1500 (CNS) analyzer.

2.4. Reagents, standards and apparatus

Magnesium sulfate anhydrous ($MgSO_4$), sodium chloride (NaCl), ultra residue reagent acetonitrile (ACN), HPLC/MS grade methanol (MeOH), ultra HPLC/MS grade water (H_2O) and HPLC/MS grade formic acid (HCOOH) were purchased from J.T. Baker. Bondesil-PSA 40 m was from Varian. Pesticide standards and triphenylphosphate (TPP) as an internal standard were purchased either from Dr. Ehrenstorfer or from Sigma-Aldrich with the highest available purity. The pesticides analyzed in this study were: chlorpyrifos-methyl, carbendazim, dichlorvos, dimethoate, imidacloprid, omethoate, metalaxyl and parathion-methyl. Stock solutions (1000 mg l^{-1}) of each pesticide were prepared in acetonitrile. From each of the single stock standards, a mix standard of 10 mg l^{-1} was prepared and used for the preparation of calibration standards with subsequent dilution with acetonitrile. The single and mix standards were stored at -4 ^{0}C while the calibration standards were prepared fresh on the day of analysis.

The HPLC-MS/MS analyses were performed using an Agilent 1200 liquid chromatographic system interfaced to an Agilent 6440 triple quadrupole mass spectrometer (Agilent Technologies, Waldbronn). Mass Hunter software was used for data acquisition and data processing.

2.5. Soil sample preparation and extraction

The soil samples were freeze dried for 24 h, gently crushed in a ceramic mortar and sieved (< 2 mm). A total of four replicates of each soil sample were made for extraction followed by the laboratory analysis. The QuEChERS method reported by Anastassiades et al. (2003) and developed for the extraction of pesticides from fruits and vegetables was modified and applied to the extraction of soil samples as follows: 10 g of homogenized soil sample was weighed in a 50 ml teflon tube and 10 ml of ultrapure water was added. The mixture was vigorously shaken for 1 min using a vortex, and then 10 ml of ACN was added and the suspension sonificated with an ultrasound system for 1 min. Four g $MgSO_4$ and 1 g NaCl were added and the sample shaken again for 1 min. Fifty µl of 10 mg l^{-1} TPP as internal standard was added and the tube shaken for 30 s followed by centrifugation at 3452 g for 30 min. Six ml of an aliquot of the supernatant was transferred to a teflon centrifuge tube (15 ml), and 0.90 g $MgSO_4$ and 0.15 g PSA were added. The tube was shaken for 30 s and centrifuged at 3452 g for 15 min. A 1.5 ml aliquot of the supernatant was filtered through a Whatmann syringe filtering vial (0.45 µm) and transferred to a LC vial. The extract was evaporated to dryness in a nitrogen stream and the residue was redissolved with ACN to make up 1.5 ml, and was used for further qualification and quantification in the LC-MS/MS system.

2.6. Recovery experiments

The recovery was determined using 3 replicates at 8 spiking concentrations from 0.01 mg kg^{-1} to 2 mg kg-1, with eight calibration solutions prepared in acetonitrile. The linearity was tested with eight concentrations between 0.01 mg l^{-1} to 2 mg l^{-1} and correlation factors (R^2) were calculated.

2.7. Chromatography and mass spectrometry

The chromatographic separation was achieved using a Zorbax SB-C-18 column 2.1×100 mm, 1.7 µm particle size from Agilent Technologies at a flow rate of 0.6 ml min^{-1}. The column was thermostated at 40 ^{0}C and the injection volume was 5 µl. The mobile phases consisted of A: H_2O-MeOH, 90% - 9.95% (v/v) with 0.05% HCOOH and B: H_2O-MeOH, 9.95%-90% (v/v) with 0.05% HCOOH. The solvent gradient used is given in Table 1.

The MS/MS was equipped with an electrospray ionization (ESI) interface operated in positive mode. The nebulizer gas (nitrogen) pressure was 25 psi, the drying gas flow rate was 8 ml min^{-1} and the drying gas temperature was 300 ^0C. Collision cell energy and fragmentor voltage were optimized in the dynamic Multiple Reaction Monitoring mode (MRM) for each pesticide and are listed in Table 2.

Table 1. Solvent gradient used for the separation of the pesticides

Time (min)	Solvent B (%)
0	10
28	98
30	100
31	100
31.5	10
40	10

Table 2. Optimized LC-MS/MS parameters for the selected pesticides

Compound name	Retention time (min)	Precursor ion (m/z)	Product ion (m/z)	Fragmentor voltage (v)	Collision cell enegry (V)
Chlorpyrifos-methyl (Qualifier)	27.1	321.9	289.6	80	12
Chlorpyrifos-methyl	27.1	321.9	124.8	80	20
Metalaxyl (Qualifier)	20.4	280.2	191.9	90	12
Metalaxyl	20.4	280.2	219.9	90	8
Parathion-methyl (Qualifier)	22.2	264	231.7	110	12
Parathion-methyl	22.2	264	124.8	110	16
Dimethoate (Qualifier)	15.4	230	170.8	70	8
Dimethoate	15.4	230	198.7	70	4
Dichlorvos (Qualifier)	18.7	221	144.8	130	12
Dichlorvos	18.7	221	108.9	130	16
Imidacloprid (Qualifier)	14.8	256	179	60	12
Imidacloprid	14.8	256	209	60	4
Carbendazim (Qualifier)	6.6	192	160	95	40
Carbendazim	6.6	192	132	95	18
Omethoate (Qualifier)	11.7	214	154.8	80	20
Omethoate	11.7	214	182.8	80	4

All the above mentioned compounds were analyzed in positive polarity mode with a dwell time of 30 ms.

3. Results and Discussion

3.1. Questionnaire survey

The questionnaire survey revealed that the farmers were using various pesticides for agricultural production: carbendazim (fungicide), chlorpyrifos-methyl (organophosphorous insecticide), cypermethrin (insecticide-synthetic pyrethroid), parathion-methyl (organophosphorous insecticide), imidacloprid (systemic insecticide), metalaxyl (systemic fungicide), dimethoate (organophosphorous insecticide), omethoate (organophosphorous insecticide) and dichlorvos (organophosphorous insecticide). According to a survey carried out by Dahal (1995) in some parts of eastern, western and central Nepal organochlorine insecticides like aldrin, endosulfan and BHC (benzene hexachloride) dust were found to be used in larger amounts by 95% of interviewed farmers. The study also concluded that farmers used chemical pesticides not only to control pests on crops but also to store the food grains, lentils, vegetables and fruits (Dahal 1995). However, in the present study, out of a total of thirty farmers interviewed, none of them were using organochlorine pesticides. The reason could be that the number of interviewed farmers was not big enough and was restricted only to a few districts around Kathmandu city. It might also be that organochlorine pesticides are not purchased anymore and have been substituted by persistent organophosphorous pesticides.

3.2. Recovery experiments

The recovery of the analyzed compounds was found to be 77.5-112% (standard deviation between 1.78% and 4.01%). All the compounds under study were found to be linear over a concentration range between 0.01 mg kg^{-1} - 2 mg kg^{-1} with correlation factors R^2 between 0.996-0.999. The LOQ (limit of quantitation) of the analyzed compounds were between 0.2 µg kg^{-1} and 6.25 µg kg^{-1}. The highest value of LOQ was found to be 6.25 µg kg^{-1} for parathion-methyl while the lowest value of 0.20 µg kg^{-1} was found for metalaxyl. The recoveries, relative standard deviation, correlation factors and LOQ are represented in **Table 3**.

Table 3. The recoveries, relative standard deviation, correlation factor and LOQ (limit of quantitation) of the studied pesticides. The measurements were made for eight calibration solutions with three replicates of each

Compound name	Recovery (%)	Standard deviation (%)	Correlation factor (R^2)	LOQ (µg kg^{-1})
Chlorpyrifos-methyl	98	1.78	0.998	1.81
Metalaxyl	78	3.38	0.997	0.20
Parathion-methyl	77.5	4.01	0.996	6.25
Dimethoate	112	2.27	0.997	0.22
Dichlorvos	97	2.04	0.999	0.65
Imidacloprid	108	2.04	0.998	0.61
Carbendazim	78	3.27	0.998	0.58
Omethoate	83	3.86	0.998	0.62

3.3. Results of the soil analyses

The soil texture, pH and organic carbon content are presented in **Table 4**. The sites Kathmandu, Godavari, Thimi and Chattradeurali had loamy sand, Kirtipur and Bhaktapur had silty sand, and Lalitpur and Dhading Besi had clay sand and sandy loam respectively. Generally, sandy soils tend to have a low organic matter content. The lesser the organic matter content in soil, the lesser the microbial activity, which increases the probability of pesticide contamination on the soil surface (Kerle et al. 2007). The soils from Kathmandu and Thimi were strongly acidic, from Kirtipur, from Godavari and Bhaktapur very strongly acidic and from Chattradeurali and Dhading Besi moderately acidic when the values were compared with Bruce and Rayment (1982). The organic carbon content of the soil from Thimi was higher than that of the other sites with a value of 3.7%. Soils that have an organic layer, such as crop residues or thatch in turf grass, may strongly sorb pesticides and reduce their mobility (Kerle et al. 2007).

Table 4. Analytical results of soil characteristics (10 cm depth) from studied sites (n = number of samples)

S.N.	Soil sampling sites	Soil Texture	pH (H_2O)	Organic carbon content [w/w%]
1	Kathmandu (n=2)	loamy sand	5.4	1.74
2	Kirtipur (n=2)	silty sand	4.5	1.05
3	Lalitpur (n=2)	clay sand	5.8	1.55
4	Godavari (n=1)	loamy sand	4.7	1.30
5	Thimi (n=2)	loamy sand	5.2	3.70
6	Bhaktapur (n=1)	silty sand	4.5	1.52
7	Chattradeurali (n=3)	loamy sand	5.9	1.30
8	Dhading Besi (n=2)	sandy loam	5.6	1.63

The soil samples at 10 cm from all the selected sites except that from Kathmandu (Kathmandu and Kirtipur) were contaminated by various pesticides (**Figure 2**). The soil samples were contaminated with the fungicide carbendazim up to 0.038 mg kg^{-1}, the insecticide chlorpyrifos-methyl up to 0.038 mg kg^{-1} and the systemic insecticide imidacloprid up to 0.016 mg kg^{-1}. A study carried out by the Soil Entomology Division of Nepal Agricultural Research Council (NARC) revealed that the analytical results of soil samples from the vegetable growing area of Kavre, Dhading, Chitwan and Bhaktapur showed only a few isomers of γ and α–BHC (γ-BHC -0.001 mg kg^{-1} and α–BHC 0.003 mg kg^{-1}) in some samples (Soil Entomology Division 1998).

The pesticide amounts in the soil samples of the present study were lower than the reported amount of applied pesticides according to the field survey. As far as we know, target values for pesticide residues in soil have not yet been developed, and therefore the present data could not be compared with any of the internationally authorized target values. However, according to environmental quality standards for soil (Wang et al. 2008), three soil contamination degrees are depicted: slightly polluted soils containing 0.05-0.5 mg kg^{-1}, moderately polluted soils with 0.5 to 1 mg kg^{-1} and heavily polluted soils with > 1 mg kg^{-1}. In the present study, the soil samples collected at the depth of 10 cm were found to be contaminated with the fungicide carbendazim and

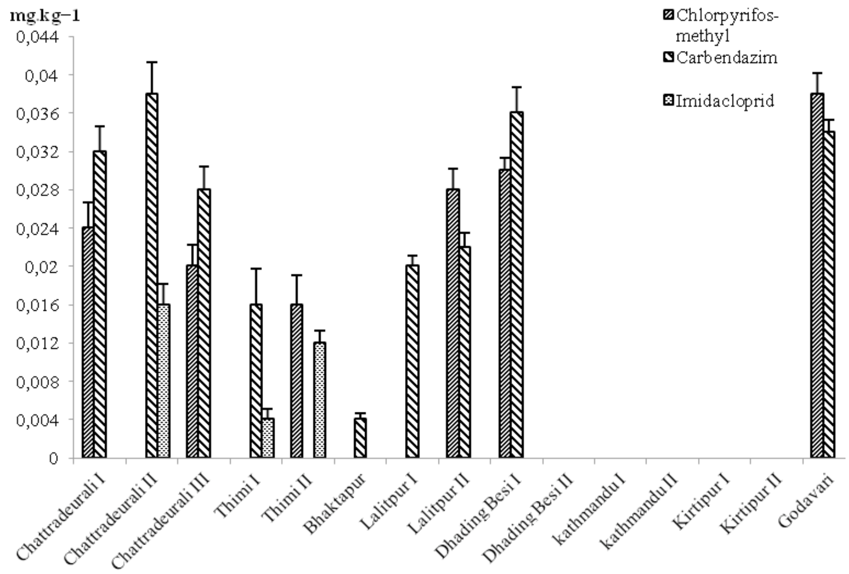

Figure 2. Levels of pesticide residues in the soil samples at 10 cm depth.

the insecticide chlorpyrifos-methyl up to 0.038 mg kg^{-1} and the systemic insecticide imidacloprid up to 0.016 mg kg^{-1}. When the pesticide residue levels in the soil samples of this study are compared with these target values, each soil sample of this study can be defined as having little pollution. The reason could be due to different soil properties and climatic conditions (Herrmann et al. 2002). An investigation into the degradation of pesticide in soil samples contaminated with malathion, dimethoate, fenvalarate and metalaxyl at different moisture contents and temperatures under defined laboratory conditions showed that the disappearance of pesticides was mainly dependent on moisture content and incubation temperature (Vinke et al. 2002). Other factors responsible for the concentration of pesticide residues in soil are the chemical properties of soil, soil use and type, persistence of the pesticide, the technique and rate of application, the frequency and timing of precipitation, soil organic carbon, the tillage system etc. (Redondo et al. 1994; Wang et al. 2006; Fabietti et al. 2009; Ling et al. 2010). The soil samples in the present study were collected during monsoon season at the end of September 2008, which could be one of the reasons for the lower than expected pesticide concentration when the values are compared with the standard values as described in Wang et al. (2008).

The distribution pattern of pesticide residues at different soil depths (10 cm, 30 cm and 50 cm) of the four collected soil profiles from Godavari (Lalitpur district) showed that they were contaminated with dimethoate, omethoate, dichlorvos, parathion-methyl and metalaxyl at different concentrations. The soil profile survey revealed a homogenous distribution of the pesticides in all the studied depths (**Figures 3a** to **3e**). As all the samples of this study were agricultural soils, the tillage activity could have churned and homogenized the concentration of the pesticide residues in the studied soil depths. A similar study carried out to investigate the pesticide residue level at different soil depths (surface (0-30 cm) and subsurface (30-60 cm)) showed that the distribution in surface layer was higher than in the sub surface layer (Al-Wabel et al. 2011). The movement of pesticides through a soil profile depends mainly on solubility, adsorption and desorption of the compounds. Most pesticides and their biologically active metabolites only penetrate to deeper soil layers in very special circumstances, although penetration in the deeper soil layers has been reported by some researchers (Manandhar 2005).

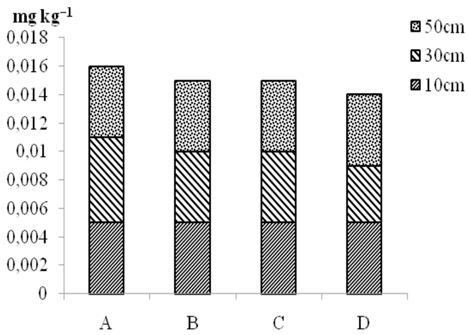

Fig. 3a Omethoate

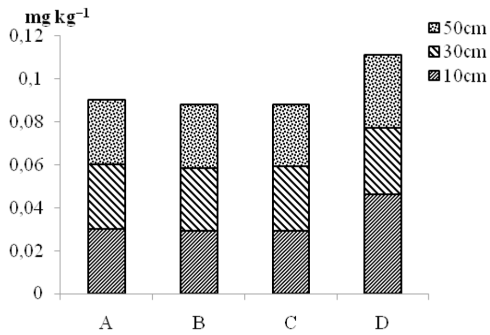

Fig. 3b Dimethoate

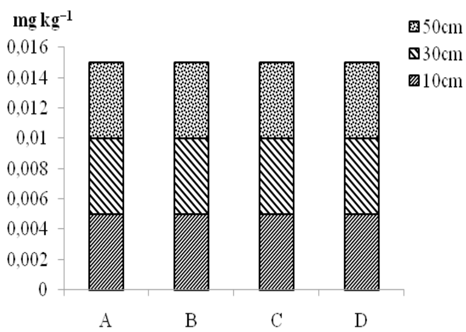

Fig. 3c Dichlorvos

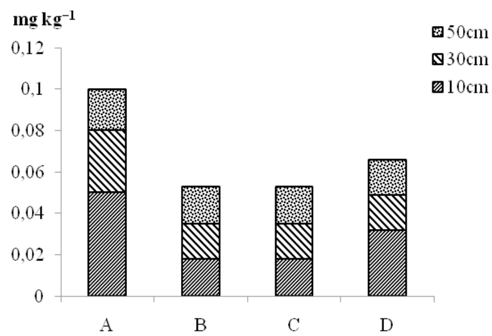

Fig. 3d Metalaxyl

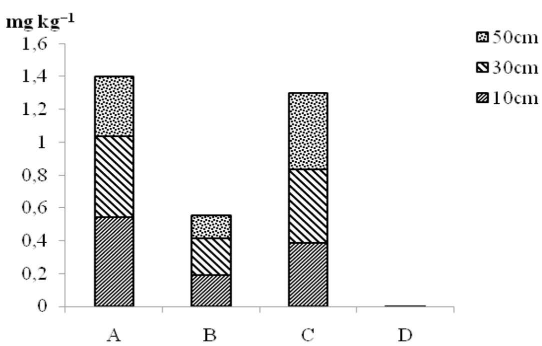

Fig. 3e Parathion-methyl

Figure 3. Distribution of pesticide residues (**a**) omethoate (**b**) dimethoate (**c**) dichlorvos (**d**) metalaxyl (**e**) parathion-methyl in the soil samples at 10 cm, 30 cm and 50 cm. The sites A to D indicate different sampling points of Godavari, Lalitpur district.

4. Conclusions

The field study revealed that various organophosphorous pesticides, insecticides, systemic fungicides and synthetic pyrethroids were detectable in the soils. The soils were acidic and sandy with an organic carbon content in the range of 1.5% for all the samples except those from Thimi. The soil samples collected at 10 cm depth were found to be contaminated with pesticides at various concentrations. Maximum concentrations of 0.038 mg kg^{-1} for carbendazim and 0.038 mg kg^{-1} for chlorpyrifos-methyl were found in the Chattradeurali II of Dhading and Godavari of Lalitpur districts respectively. However, the concentration of pesticide residues in the analyzed samples was found to be less than that observed in the field application. Another test carried out at different soil depths (10 cm, 30 cm, and 50 cm) showed that the pesticides were distributed homogenously through the profile. An “old” insecticide parathion-methyl, which has been banned for use in many countries, was still applied and found in some samples. The findings of the present study are based on a small sample size, which decreases the probability of finding contaminated samples. However, this study is the first to quantify the magnitude and type of soil contamination due to pesticide use in central Nepal, and by doing so, reveal the pesticide use and safety among farmer communities in the region. It could act as a base for a comprehensive study covering all the ecological divisions of Nepal (Terai, Hills and Mountain), which would result in a better understanding and more meaningful conclusions to be drawn for the whole country. This study also shows that an information campaign regarding pesticide use and food safety for farmers and consumers is essential.

5. Acknowledgments

This work was financially supported by the Commission for Development Studies (KEF Project No. 162)/OEAD GmbH, Vienna, Austria. The authors would like to acknowledge the Institute of Soil Research, Institute of Forest Ecology and Institute of Analytical Chemistry at the University of Natural Resources and Life Sciences (BOKU), Vienna. Special thanks to LVA GmbH, Vienna, and Environment and Public Health Organization (ENPHO), Nepal.

REFERENCES

- Al-Wabel MI, El-Saeid MH, Al-Turki AM, Abdel Nasser G. 2011. Monitoring of pesticide residue in Saudi Arabia Agricultural Soils. J Environ Sci. 5:269-278.
- Anastassiades M, Lehotay SJ, Stajnbaher D, Schenck FJ. 2003. Fast and easy multiresidue method employing extraction/partitioning and dispersive solid phase extraction for the determination of pesticide residues in produce. J AOAC Int. 86:412-431.
- Baker SL, Gyawali BK. 1994. Promoting Proper Pesticide Use in Nepal. Ministry of Agriculture (Nepal)/Winrock Int. Report Ser. 28, Kathmandu, Nepal.
- Bruce RC, Rayment GE. 1984. Soil Testing and some soil test interpretations used by the Queensland Department of Primary Industries. Agricultural Chemistry Branch, Brisbane: Queensland Department of Primary Industries; Available from: http://www.southwestnrm.org.au/sites/default/files/uploads/ihub/soil-testing-and-some-soil-test-interpretations-used-queensland-department-primary-industries.pdf. ISSN 0727-6273.
- Dahal L. 1995. A Study on Pesticide Pollution in Nepal, International Union for Conservation of Nature (IUCN), Nepal.
- Engle CF, Cogger CG, Stevans RG. 1993. Role of soil in groundwater protection. Cru.cahe.wsu.edu/CEP Publications/eb 1633.html/ access date 16.10.2012.
- Fabietti G, Biasioli M, Barberis R, Ajmone- Marsan F. 2009. Soil contamination by organic and inorganic pollutants at the regional scale: the case of Piedmont, Italy. J Soils Sediments 10:290-300.
- Giri NP. 1998. Pesticide pollution in vegetable crop in Kathmandu valley. Dissertation. Tribhuwan University, Nepal.
- Hermann A, Schumann S. 2002. Collaborative project on environmental risks of pesticides and sustainable development of Integrated Pesticide Management System (IPMS) in Nepal considering socio-economic conditions. Landschaftsökologie und Umweltforschung, Braunschweig 38:38-48.
- Kerle EA, Jenkins JJ, Vogue PA. 2007. Understanding pesticide persistence and mobility for groundwater and surfacewater protection. Extension.oregonstate.edu/catalog/pdf/. Access date 16.10.2012.
- Khanal G. 2012. Pesticide use in agriculture, our health and risk on underground water in Nepal. Agriculturecornerblog.blogspot.co.at/2012/07/. Access date 16.10.2012.
- Kreuger J, Adielsson S. 2006. In: 4th European conference on pesticides and related organic micro pollutants in the environment. Almería, Spain.

• Ling W, Zeng Y, Gao Y, Dang H, Zhu X. 2010. Availability of polycyclic aromatic hydrocarbons in aging soils. J Soils Sediments 10:799-807.

• Manandhar D. 2005. Implementation of the Stockholm Convention on Persistence Organic pollutants (POPs) enabling activities in Nepal. Proceedings of Inception Workshop. Ministry of Population and Environment (MOPE)/Unite Nations International development Organization (UNIDO). p. 104-110.

• Miglioranza KSB, Julia E, Moreno A, Moreno V. 2003. Trends in Soil Science: Organochlorine pesticides in Argentinean Soils. J Soils Sediments 4:264-265.

• OENORM L 1083. 2006. Chemical Analysis of soils-Determination of pH value.

• OENORM L 1080. 1999. Chemical Analysis of soils-Determination of organic carbon by dry combustion.

• OENORM L 1061. 2002. Chemical Analysis of soils-Determination of soil texture.

• Palikhe BR. 2001. Pesticides pollution management in Nepal: In harmony with nature. Agriculture and Environment communication issue. Ministry of Agriculture and Cooperative. Kathmandu, Nepal.

• Palikhe BR. 2002. Challenges and options of pesticide use: in the context of Nepal. Proceedings Int Workshop on Environmental Risk Assessment of pesticides and Integrated Pesticide Management in Developing Countries. Landschaftsökologie und Umweltforschung, Braunschweig 38:132-143.

• Poudel A. 2011. Pesticide residue found in food across the country. archives.myrepublica.com/portal/. Access date 20.10.2012.

• Redondo M, Ruiz M, Boluda R, Font G. 1994. Persistence of pesticide residues in orchard soil. Sci Total Environ. 58:199-205.

• Schumann S. 2005. Environmental Risk Assessment of Pesticides in Nepal and Hindukush-Himalayan Region. Earth and Environ Sci. 38:83-98.

• Soil Entomology Division. 1998. Annual Technical Report. Entomology Division, Nepal Agricultural Research Council (NARC). Khumaltar, Lalitpur, Nepal.

• Tang X, Shen C, Chen L, Xiao X, Wu J, Khan MI, Dou C, Chen Y. 2010. Inorganic and organic pollution in agricultural soil from an emerging e-waste recycling town in Taizhou area, China. J Soils Sediments 10:895-906.

• Thurman EM, Fany Y, Zavitsanos P, Zweigenbaum JA. 2008. Multiresidue analysis of 100 pesticides in food samples by LC/Triple Quadrupole Mass Spectrometry, Agilent Technologies Publication. p. 5989-5469.

• Upadhyaya NS. 2002. Experiences of community Integrated Pest Management in Nepal. Proceedings Int Workshop on Environmental Risk Assessment of pesticides and Integrated Pesticide Management in Developing Countries. Landschaftsökologie und Umweltforschung, Braunschweig 38:256-264.

• Vinke C, Kreuzig R, Bahadir, M. 2002. Analytical investigations on the fate of pesticides in soils of the Jhikhu Khola catchment. Proceedings Int Workshop on Environmental Risk Assessment of pesticides and Integrated Pesticide Management in Developing Countries. Landschaftsökologie und Umweltforschung, Braunschweig 38:265-275.

• Wang F, YR Bian, Jiang X, Gao HJ, Yu GF, Deng JC. 2006. Residual characteristics of organochlorine pesticides in Lou soils with different fertilization modes. Pedosphere 16:161-168.

• Wang X, Wang D, Qin X, Xu X. 2008. Residues of organochlorine pesticides in surface soils from college school yards in Beijing, China. J Environ Sci. 20:1090-1096.

• Williams M. 2000. Nutrition and the Environment. ACC SCN 21 Report. Geneva, Switzerland.