Environmental Pollution 159 (2011) 3344–3350

Contents lists available at SciVerse ScienceDirect

Environmental Pollution

journal homepage: www.elsevier.com/locate/envpol

Pesticide pollution in agricultural areas of Northern Vietnam: Case study in Hoang Liet and Minh Dai communes

Pham Manh Hoai[a], Zita Sebesvari[b], Tu Binh Minh[a], Pham Hung Viet[a], Fabrice G. Renaud[b,*]

[a] Research Center for Environmental Technology and Sustainable Development, Hanoi University of Science, 334 Nguyen Trai Str., Hanoi, Viet Nam
[b] United Nations University, Institute for Environment and Human Security (UNU-EHS), Hermann-Ehlers-Str. 10, D-5311, Bonn, Germany

ARTICLE INFO

Article history:
Received 31 May 2011
Received in revised form
19 August 2011
Accepted 20 August 2011

Keywords:
Pesticide residues
Pollution
Water
Biota
Vietnam

ABSTRACT

Soils and agricultural products from the Red River basin in Northern Vietnam were reported to be contaminated by agrichemicals. To assess potential exposure of local farmers and consumers to these contaminants, pesticide use and management practices of local farmers were surveyed and residue concentrations were determined for recently used as well as for banned pesticides in water, soil, vegetables, and fish samples in two communes of Northern Vietnam. DDTs, HCHs, and Drin compounds still persist at relatively high concentrations in soil and occur in vegetable and fish samples. Recently used pesticides, such as fenobucarb, trichlorfon, cyfluthrin, and cypermethrin were detected in vegetable and fish samples. Thresholds for acceptable daily intake levels (ADI) were frequently reached in the analyzed food products pointing to the fact that current pesticide management practices do not only result in a pollution of the environment but also pose threats to human health.

© 2011 Elsevier Ltd. All rights reserved.

1. Introduction

A large number of chemicals have extensively been used to maintain high agricultural yields and eradicate vector borne diseases in Vietnam in the last decades. As a result of the economic transformation process (Doi Moi) starting in 1986 in Vietnam, pesticide use has nearly doubled in the 1990s by reaching over 40,000 tons of pesticide active ingredients in 1998 (Meisner, 2005). Some of these compounds are toxic and/or persistent and thus potentially threaten the environment. Since 1995, some organochlorine pesticides such as dichlorodiphenyltrichloroethane (DDTs), hexachlorobenzene (HCB) and hexachlorocyclohexanes (HCHs) are banned in Vietnam (Sinh et al., 1999). Most of these banned pesticides can still be found in relatively high concentrations in the environment throughout Vietnam as reported by various authors (Viet, 2002; Minh et al., 2006, 2007a,b; Kishida et al., 2007; Toan et al., 2007; Carvalho et al., 2008; Hoai et al., 2010) and reviewed by Minh et al. (2007c). Carvalho et al. (2008) for example monitored the residues of more than 70 pesticides in the Mekong Delta and found sediment concentrations of ΣDDT ranging from 0.45 to 67.5 ng/g dry weight and concentration in the soft tissues of bivalve molluscs ranging from 5.5 to 123.0 ng/g dry weight. Carvalho's study represents one of the few that monitored also for some more recently used pesticides. Although organophosphates, carbamates, pyrethroids, and nicotinoids have largely replaced the organochlorines, little information exists on their current usage (Berg, 2001; UNU-EHS, 2010) and almost no information is available on their residue concentrations in the environment and food products and resulting potential impacts on human health. Recent data published by Lamers et al. (2011) showed that there is a considerable loss of organophosphates, carbamates, and nicotinoids applied in paddy fields of Northern Vietnam to receiving streams as well as to groundwater.

The Vietnamese government has put considerable effort into the promotion of different campaigns (e.g. "Ba Giam, Ba Tang" (3 Reductions, 3 Gains), "Mot Phai, Nam Giam" (1 Must Do, 5 Reductions), and "Bon Dung" (4 Rights)) aiming to reduce chemical inputs in agricultural production. However, unpublished field survey results of the authors show that pesticide overuse/pollution is still a major issue.

In order to determine the extent of the pesticide pollution and potential health consequences, the objectives of this study were to (1) identify currently used pesticides and common pesticide management practices via questionnaire surveys and (2) assess residue concentrations of selected currently used pesticides and some formerly used persistent organochlorine pesticides in the environment (water, soil) and biota (vegetables and fish) at two study sites in Northern Vietnam. The questionnaire survey and the samples for residue monitoring were collected at the same study

* Corresponding author.
E-mail address: renaud@ehs.unu.edu (F.G. Renaud).

0269-7491/$ – see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envpol.2011.08.044

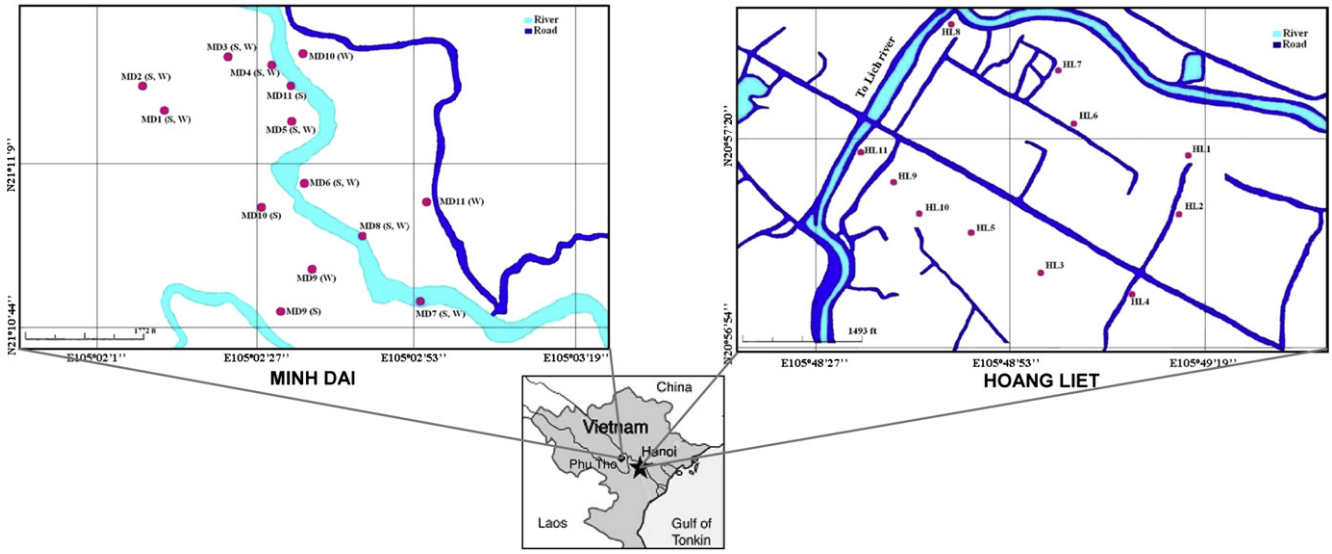

Fig. 1. Hoang Liet and Minh Dai sampling sites.

sites in order to guide the selection of pesticides for the monitoring, to gain information on pesticide management practices and to investigate if general patterns of pesticide use observed at the scale of the study site are reflected in residue concentrations found in the environment.

2. Materials and methods

2.1. Study area

Two communes in Northern Vietnam, Hoang Liet (20° 57′ N, 105° 49′ E) and Minh Dai (21° 11′ N, 105° 2′ E), were selected as study sites (Fig. 1). Both sites have an annual average temperature of ca. 24 °C and an annual average precipitation of ca. 1700 mm.

Hoang Liet commune is located in one of five suburban districts of Hanoi City and represent in this study suburban agricultural sites (Fig. 1). Suburban areas in Vietnam play an important role in the supply of the cities with fresh vegetables and fruits. Vegetables from Hoang Liet have been reported to be contaminated by heavy metals and pesticide residues recently (Hai, 2006; MONRE, 2006).

The second study site, Minh Dai commune, is located in Thanh Son district – a southwest mountainous district in Phu Tho province, ca. 140 km northwest from Hanoi (Fig. 1). Tea is the main crop in Minh Dai with a cultivation area of 3500 ha, occupying one third of the total area of the district. Thus, the commune represents mountainous tea producing areas, which have a growing importance in Northern Vietnam. Additionally, there was a pesticide stockpile located in this commune until the end of 1990s. Up to 30 μg/g total DDT was reported in the soil at this site in 2005 (Vietnam Environmental Protection Agency (VEPA), personal communication).

2.2. Samples and sampling sites

2.2.1. Water and soil samples

A total of twenty-two water samples and twenty-two soil samples (11 samples from each study sites) were collected from Hoang Liet and Minh Dai communes in August 2007 (Fig. 1).

Water samples were collected from ponds, ditches, and canals 20 cm below the water surface into 2-L glass bottles with PTFE sealed caps.

Soil samples were collected from fields covered by different crops (rice, vegetables, tea), from irrigation ditches and residential areas. Samples were taken by stainless steel corer at depths from 0 to 20 cm, at five points with an area of approximately 25 m^2 and combined to make a composite sample. Each soil sample consisted of a composite of ten sub-samples taken from a 5 × 5 m square plot (two rows of 5 sub-samples collected at 1 m intervals; rows were spaced 5 m apart).

2.2.2. Vegetable samples

In Hoang Liet commune, composite samples (stem and leaves, 1 kg each) of five different types of vegetables, cultivated at the time of sampling, were collected in June 2008. The five vegetables were mugwort (*Artemisia vulgaris*), heartleaf (*Houttuynis cordata*), water spinach (*Ipomoea aquatica*), water mimosa (*Neptunia oleracea*), and katuk or star gooseberry (*Sauropus androgynus*). In Minh Dai commune, five composite samples of tealeaves (1 kg each) were collected in February 2008. Composite samples of tea and vegetables were prepared by combining five sub-samples from the same field.

2.2.3. Fish samples

Ten fish samples, including Mozambique tilapia (*Oreochrimis mossambicus*), black pacu (*Colossoma brachypomum*), catfish (*Siluriformers*), carp (*Cyprinus carpio carpio*), mud carp (*Cirrhina molitorella*), and channa maculates (*Ophiocephalus maculates*) were collected from ten fishponds around vegetable or tea fields in February 2008.

Samples were transported to the laboratory on ice and stored either at 4 °C (water and vegetable samples) or at −20 °C (soil and fish samples) for no longer than one week before analysis.

2.3. Chemical analysis

Water and soil samples were analyzed for 21 organochlorinated pesticides, including DDTs (o,p′-DDE, p,p′-DDE, o,p′-DDD, p,p′-DDD, o,p′-DDT, p,p′-DDT), hexachlorocyclohexanes (HCHs: α-HCH; β-HCH; γ-HCH; δ-HCH; ε-HCH), chlordane compounds (CHLs: trans-chlordane, cis-chlordane), drin compounds (aldrin, dieldrin, isodrin), hexachlorbenzene (HCB), heptachlor, cis-heptachloroepoxide, methoxychlor, and mirex. Fish, vegetable and tea samples were analyzed for the above-mentioned 21 organochlorine pesticides and additionally for fenobucarb (carbamate pesticide), trichlorfon (organophosphorous pesticide), cyfluthrin and cypermethrin (pyrethroid pesticides). A summary of sample sites, samples and analyzed compounds is shown in Table 1.

2.3.1. Water and soil samples

Organochlorine pesticides in water and soil samples were determined by applying EPA 3620B and EPA 8081A methods (EPA, 1996a,b) with small

Table 1
Sampling sites, samples and analyzed pesticide compounds.

Sample matrix	Hoang Liet	Minh Dai	Analyzed compounds
Water	11 samples	11 samples	21 organochlorines
Soil	11 samples	11 samples	21 organochlorines
Vegetable	5 composite samples	–	21 organochlorines, fenobucarb, trichlorfon, cyfluthrin, cypermethrin
Tea	–	5 composite samples	21 organochlorines, fenobucarb, trichlorfon, cyfluthrin, cypermethrin
Fish	10 individual samples	10 individual samples	21 organochlorines, fenobucarb, trichlorfon, cyfluthrin, cypermethrin

modifications. Briefly, for water, 30 g NaCl and 20 μl of 5 μg/ml surrogate compound (p,p′-DDT-^{13}C) was added to the 1 L water sample which was then liquid–liquid extracted by 50 ml of n-hexane. The extract was dried over anhydrous Na_2SO_4 and then concentrated to approximately 1 ml. The cleanup step was carried out by solid phase extraction using florisil cartridges (1 g, 6 ml). Pesticides were eluted by 12 ml of 2% (v/v) acetone in n-hexane. The eluent was then concentrated, spiked with 20 μl of 5 μg/ml internal standards, and filled up to 1 ml by hexane.

For soil samples, 10 g of air-dried sample was mixed with 40 ml of acetone, 5 g of Na_2SO_4 and 20 μl of 5 μg/ml surrogate standard (p,p′-DDT-^{13}C) in a 100 ml centrifuge tube. The sample was ultrasonicated for 5 min, shaken for 2 h and then centrifuged for 5 min at 3000 rpm. The extract was then concentrated to 1 ml. Activated copper slices were added to remove sulfurous compounds before the solution was cleaned up by a florisil cartridge (1 g, 6 ml). After elution with 7 ml of hexane/acetone (9:1, v/v) the eluate was concentrated to less than 1 ml, spiked with internal standards, and filled up to 1 ml by hexane.

2.3.2. Fish samples

Organochlorine pesticides, trichlorfon, fenobucarb, cyfluthrin, and cypermethrin in fish samples were simultaneously determined using a method developed in the framework of United Nation University program (UNU, 2007) with small modifications. Briefly, 50 ml acetonitrile, 20 μl of 5 μg/ml surrogate standards (p, p′-DDT-^{13}C and diazinon-d_{10}) were combined in a homogenizer cup with 2 g of minced edible parts of the fish and was homogenized for 5 min. The extract was filtered and concentrated to 1 ml before cleanup with C18 (1 g, 6 ml) and R-NH_2 cartridges (2 g, 12 ml). The pesticides were sequentially eluted by acetonitrile. The eluate was concentrated to less than 1 ml, spiked with internal standards, and filled up to 1 ml by acetonitrile.

2.3.3. Vegetable and tea samples

Organochlorine pesticides, trichlorfon, fenobucarb, cyfluthrin, and cypermethrin in vegetable samples were simultaneously determined following the method reported by Ueno et al. (2004) with necessary modifications. Briefly, 5 g of minced edible parts of vegetables or minced leaf parts of tea samples were ultrasonically extracted using 40 ml of ethyl acetate for 10 min. The extract was filtered and rinsed twice with 10 ml of ethyl acetate into a 250 ml round bottom-flask and then concentrated to around 1 ml. The cleanup step was conducted with activated carbon packed column (2 g) and florisil cartridge (1 g, 6 ml). The pesticides were eluted by 40 ml of ethyl acetate. The eluent was then concentrated to less than 1 ml, spiked with internal standards, and filled up to 1 ml by ethyl acetate.

Pesticides were quantified by injecting 2 μl of the final extracts in GC–MS (GCMS-QP2010, Shimadzu, Co. Ltd., Japan) at following conditions: injector block temperature: 280 °C; DB 5 capillary column 30 m × 0.25 mm ID × 0.25 μm film thickness, oven temperature was ramped from 70 °C up to 290 °C. The quantification of the analytes was carried out by using internal standards (phenanthrene-d_{10} and chrysense-d_{12}).

2.4. Quality assurance and quality control

Laboratory blank samples were extracted and analyzed on a regular basis. All samples and blanks were spiked with surrogate standards (p,p′-DDT-^{13}C and diazinon-d_{10}) prior to extraction to monitor for extraction quality. Surrogate recovery was evaluated by internal standards (phenanthrene-d_{10} and chrysense-d_{12}) spiked to the sample after the extraction. With the exception of one fish sample (HLF05) surrogate standard recovery in samples ranged between 70 and 130%, which was considered as an acceptable range.

The performance of the extraction was assessed by recovery experiments. Uncontaminated matrices were spiked with a mixture of pesticides (30 ng/L for water, 10 ng/g dry wt. for soil, 25 ng/g wet wt. for fish and 10 ng/g wet wt. for tea), extracted and analyzed as described above. While the method performance was good for water, soil, and tea samples (recoveries ranged from 68 to 98%, 85–112%, and 83–101%, respectively), recovery performance was lower for fish samples (56–98%). Since the different compounds with different polarities were extracted in one step, high recovery rates could not be successfully obtained for all compounds in fish samples with high content of fatty acids, fatty esters, diglycerides, triglycerides etc. Among the organochlorines detected in fish samples in this study, the following failed to reach 70% recovery rate: aldrin (64%), dieldrin (65%), isodrin (69%), o,p′-DDE (64%), o,p′-DDT (64%), and ε-HCH (69%). However, the extraction method performed well for trichlorfon, fenobucarb, cyfluthrin, and cypermethrin, which were predominant in fish samples. Therefore, the method was not changed. Consequently, the results reported in this study for the above-mentioned six compounds in fish samples will be lower than the actual residue concentrations.

The precision of the analysis was tested by repeated injections ($n = 5$) of the same sample containing 5 ng/ml of each pesticides. Deviation from the expected value for all pesticides ranged between 1.1 and 9.0% with an average of 3.7%.

2.5. Questionnaire survey

A questionnaire survey was conducted with 30 and 24 randomly selected farming households, accounting for 25% and 11% of farming households in Hoang Liet and Minh Dai, respectively. A brief questionnaire was prepared in order to understand farmer's pesticide management by surveying the type and amount of pesticides used, factors influencing farmer's decision in the selection of pesticides, preferred suppliers, usage of protective equipment when spraying, and treatment of contaminated waste. The outcome of the survey wad used to select target pesticides for the analysis as well as to evaluate farmer's pesticide management practices.

3. Results and discussion

3.1. Pesticide use and management

3.1.1. Hoang Liet commune

At the time of the interview, a total of 80 ha were used for agriculture in Hoang Liet commune. The main vegetables grown were water spinach, water mimosa, watercress, mugwort, katuk, and heartleaf. All interviewed farming households cultivated exclusively vegetables on their fields. Cypermethrin, trichlorfon, fenobucarb, fenitrothion, validamicin, and cartap (in order of frequency) were the most frequently used active ingredients at this site. 40% of the farming households selected pesticides without consulting extension workers, agriculture cooperatives, other farmers or experts. The main selection criteria for a pesticide were its expected effectiveness (90% of respondents), its price (40%) and safety for humans and animals (30%). Only 3.3% of the farming households considered environmental safety while selecting the pesticides. 73% of the framing households did not have any strategy to deal with empty pesticide containers. Annual amount of active ingredients used at Hoang Liet was in the range of 250–500 kg from various trademarks (VEPA, personal communication).

3.1.2. Minh Dai commune

The majority of the 24 interviewed farming households cultivated rice and/or tea (47% rice and tea, 18% rice and tea with some additional vegetables and/or maize, 18% only tea, 13% only rice, 4% other crop mixture). Imidacloprid, cartap, cyfluthrin, cypermethrin, trichlorfon and fenitrothion (in order of frequency) were the most frequently used active ingredients at this site. 33% of the farming households selected pesticides without consulting extension workers, agriculture cooperatives, other farmers or experts. Main selection criteria for a pesticide were its effectiveness (92%) and its safety for humans and animals (38%). Only 3% of the farmers considered environmental safety in their decision. 50% of the farming households did not have any strategy to deal with pesticide contaminated waste (empty bottles, cans etc.). Annual amount of active ingredients used at Minh Dai ranges from 1120 to 1900 kg (VEPA, personal communication).

Comparing the two communes, farmers in Hoang Liet tended to have more advanced management skills in terms of pesticide use. This finding could be related to the more homogenous crop pattern in Hoang Liet (only vegetables) and to better access to pesticide related knowledge, as the commune is located in a suburban area of Hanoi. Farmers in Hoang Liet used more protective equipment while spraying then farmers in Minh Dai, reflecting higher level of awareness for pesticide related health issues.

3.2. Pesticide residue concentrations

3.2.1. Water samples

Concentrations of organochlorinated pesticides in water samples were all below the detection limit except for sample MDW01 (ΣDDT: 16 ng/l), which was collected from a rice field at Minh Dai commune. The predominance of p,p′-DDT (85%) compared to other metabolites (DDE and DDD) at this sample hints to a recent input of this pesticide, indicating a possible illegal use of DDT in this rice field. The limits of quantification ranged from 0.11 ng/l (p,p′-DDE) to 0.90 ng/l (δ-HCH).

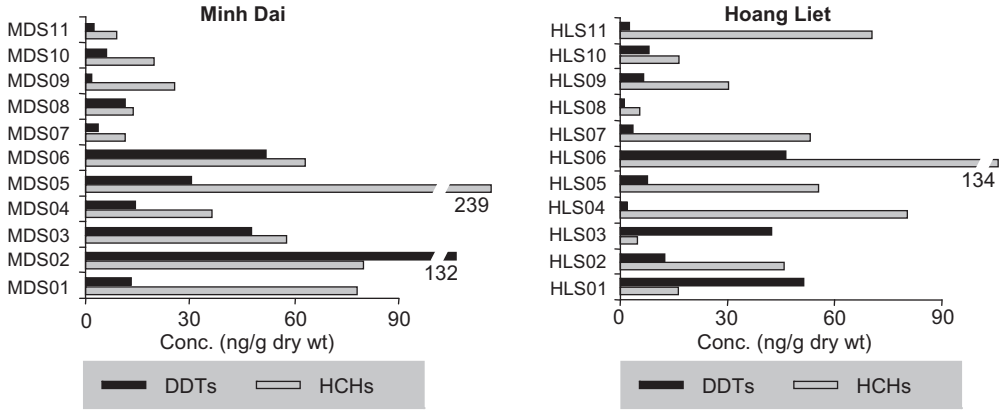

Fig. 2. Concentration of DDTs and HCHs in soil samples.

3.2.2. Soil samples

Cis-chlordane, trans-chlordane, heptachlor-epoxide, methoxychlor, and mirex were not detected in any of the collected soil samples. However, other organochlorine pesticides such as HCHs, Drin compounds, and DDTs were detected in most of the samples analyzed. In general, the contamination pattern at both study sites was as follows: HCHs > DDTs > Drins. The concentrations of HCHs, DDTs, and Drins in soils from Hoang Liet commune varied from 4.8 to 134 ng/g dry wt. (average 47 ng/g), 1.0 to 51 ng/g dry wt. (average 17 ng/g), and <0.20 to 27 ng/g dry wt. (average 6.7 ng/g), respectively. In Minh Dai commune, the concentrations of HCHs, DDTs, and Drins in soil ranged from 9.1 to 239 ng/g dry wt. (average 58 ng/g), 1.8 to 132 ng/g dry wt. (average 29 ng/g), and <0.20 to 29 ng/g dry wt. (average 6.8 ng/g), respectively (Fig. 2). γ-HCH (lindane, 41%) and α-HCH (41%), a byproduct of lindane production, were predominant in soil at Hoang Liet, while α-HCH was the predominant isomer at Minh Dai (69%) (data not shown).

The share of DDT and its metabolites was different between Hoang Liet and Minh Dai communes. p,p′-DDT contributed from 31% (MDS01) to 86% (MDS02) to the sum of DDTs in the samples from Minh Dai commune (median 56%) and from 0% (HMS01, 03, 04, and 06) to 86% (HMS05) in samples from Hoang Liet commune (median 19%). p,p′-DDE contributed from 1.0% (MDS02) to 50% (MDS07) to the sum of DDTs in the samples from Minh Dai commune (median 17%) and 0% (HLS02) to 100% (HLS01) (median 81%) in the samples from Hoang Liet commune (Fig. 3). High proportion of DDE versus DDT in most samples in Hoang Liet indicates the biodegradation of DDT to DDE. However, the sample site HLS05 displays a different pattern, which might have been caused by a recent DDT input at this site. In Minh Dai commune, a high proportion of DDT versus low proportion of DDE was observed in 45% of the samples (MDS02, MDS03, MDS04, MDS06 and MDS10). At these sites, the predominance of DDT compared to DDE may imply a recent input of DDT (Strandberg et al., 1998). In Minh Dai commune, the concentrations of HCHs and DDTs at the sampling sites MDS10 (20 and 5.9 ng/g dry wt., respectively) and MDS11 (9.1 and 2.3 ng/g dry wt., respectively), which are near to the old pesticide stockpile, were lower than concentration observed at other sampling sites indicating that the old pesticide stockpile is not the main source for the observed HCHs and DDTs. It seems that farmers at Minh Dai commune might still use some banned pesticides.

3.2.3. Fish samples

The concentrations of organochlorine pesticides in fish samples are shown in Fig. 4. Similar to soil samples, HCHs (12–78 ng/g wet wt., averge 31 ng/g) were the predominant compounds followed by DDTs (5.1–43 ng/g wet wt., average 18 ng/g), and Drin compounds (5.2–14 ng/g wet wt., average 8.3 ng/g) in fish samples collected at Minh Dai commune. The concentrations in fish samples from Hoang Liet commune were in order of HCHs (8.3–66 ng/g wet wt., average 27 ng/g) > Drins (5.4–46 ng/g dry wt., average 17 ng/g) > DDTs (4.0–8.1 ng/g wet wt., average 5.2 ng/g). Similar to the soil samples, concentrations of DDTs and HCHs in fish samples in Minh Dai commune were generally higher than those in Hoang Liet. α-HCH was predominant among the five monitored HCH isomers in Minh Dai (average 48% share) with the exception of a carp sample

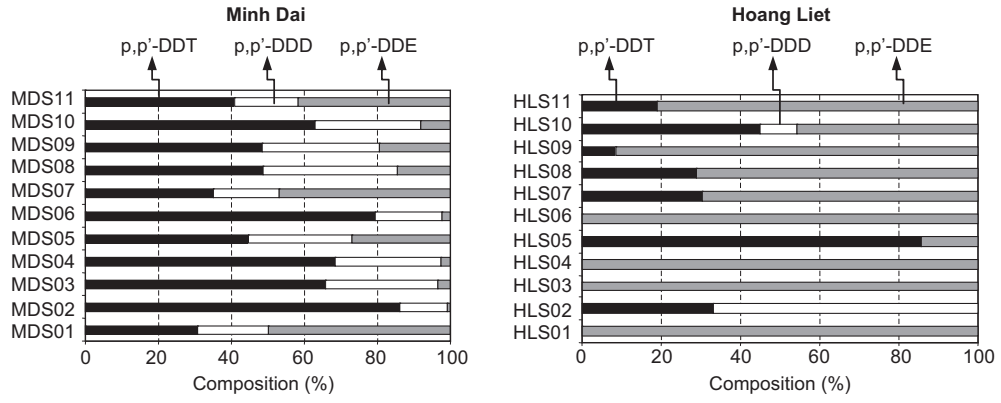

Fig. 3. Composition of p,p′-DDTs in soil samples in both communes.

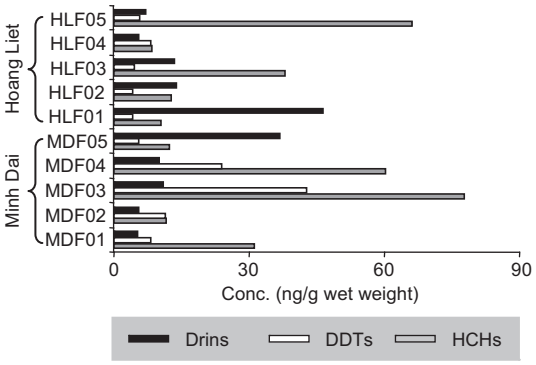

Fig. 4. Distribution of DDTs, Drins and HCHs in fish samples.

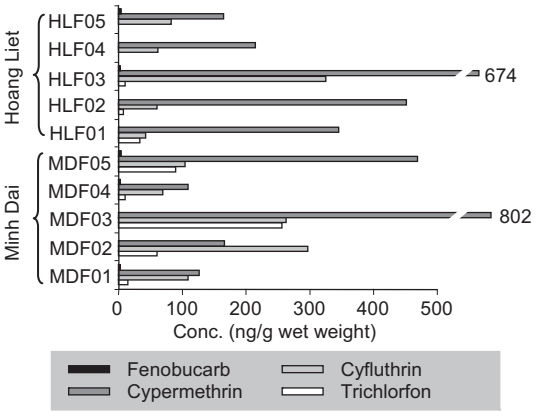

Fig. 6. Distribution of trichlorfon, fenobucarb, cyfluthrin, and cypermethrin in fish samples.

(no predominant isomer, γ, δ, ε-HCH together 81% share) while the pattern was more diverse in Hoang Liet: ε-HCH dominated in three fish samples (cultivated carps and wild channa, average share 60%) and α-HCH dominated in two of the samples (cultivated tilapia and wild channa, average share 55%) (data not shown). In general, the proportion of p,p′-DDE was higher than p,p'-DDD and p,p'-DDT (Fig. 5); however the high relative abundance of p,p′-DDT (in half of the samples it was higher than 20%), especially in two samples collected at Minh Dai (MDF02 cultivated black pacu and MDF04, wild catfisch) and in one sample from Hoang Liet (HLF03, cultivated tilapia), also indicated a possible recent input of DDT to the environment. HCB, heptachlor, heptachlor-epoxide, trans-chlordane, cis-chlordane, methoxychlor and mirex were not detected in any of the fish samples. As described earlier the extraction method had shortcomings for aldrin, dieldrin, isodrin, o,p′-DDE, o,p′-DDT, and ε-HCH. For these compounds, the recovery rates were between 64 and 69%. The concentrations reported here might therefore underestimate the residue concentrations in fish samples.

All four investigated non-organochlorine pesticides (trichlorfon, cyfluthrin, fenobucarb and cypermethrin) were detected in fish samples (Fig. 6). In general, the concentration of these four pesticides were in the order of cypermethrin (164–675 ng/g wet wt., average 370 ng/g) followed by cyfluthrin (43–325 ng/g wet wt., average 114 ng/g), trichlorfon (<0.17–34 ng/g wet wt., average 11 ng/g) and fenobucarb (0.7–4.3 ng/g wet wt., average 2.0) in Hoang Liet; and of cypermethrin (109–802 ng/g wet wt., average 335 ng/g) followed by cyfluthrin (69–297 ng/g wet wt., average 169 ng/g), trichlorfon (9.8–255 ng/g wet wt., average 86 ng/g), and fenobucarb (<0.40–4.0 ng/g wet wt., average 2.4 ng/g) in Minh Dai. Comparing average and median values (data not shown), trichlorfon and cyfluthrin residue concentrations tended to be higher in Minh Dai than in Hoang Liet, while the opposite was the case for cypermethrin.

3.2.4. Tea and vegetable samples

With the exception of HCB, heptachlor, methoxychlor, and mirex, other organochlorinated pesticides were detected in all collected samples (Fig. 7). Similar to those in soil and fish samples, HCHs, Drins and DDTs were predominant. The contamination pattern was consistently as follows: HCHs > Drins > DDTs. In Hoang Liet, the concentrations of HCHs in three samples HLV01 (144 ng/g wet wt.), HLV02 (80 ng/g wet wt.) and HLV04 (122 ng/g wet wt.) were much higher than those of DDTs and Drins. Among five monitored HCH isomers, α-HCH was predominant in samples collected at Hoang Liet whereas β- and δ-HCH were predominant in samples collected at Minh Dai (data not shown).

Fenobucarb, trichlorfon, cyfluthrin, and cypermethrin were detected in a large range of concentrations. The concentration of cyfluthrin tended to be the highest, especially for samples collected at Minh Dai. The highest concentration of cyfluthrin was 162 ng/g wet wt. in sample HLV01 (Fig. 8). In general, the concentrations of fenobucarb, cypermethrin, and cyfluthrin in tea samples at Minh Dai were higher than those in vegetables at Hoang Liet, except sample HLV01. The relatively high concentrations of fenobucarb, trichlorfon, cyfluthrin and cypermethrin in fish, tea, and vegetable samples indicated that these pesticides have been used recently at both sites which corresponds well with the survey results.

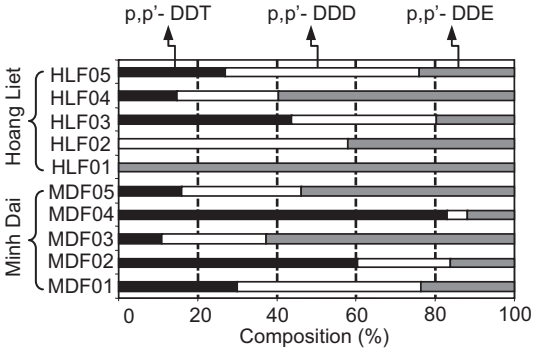

Fig. 5. Composition of p,p′-DDTs in fish samples.

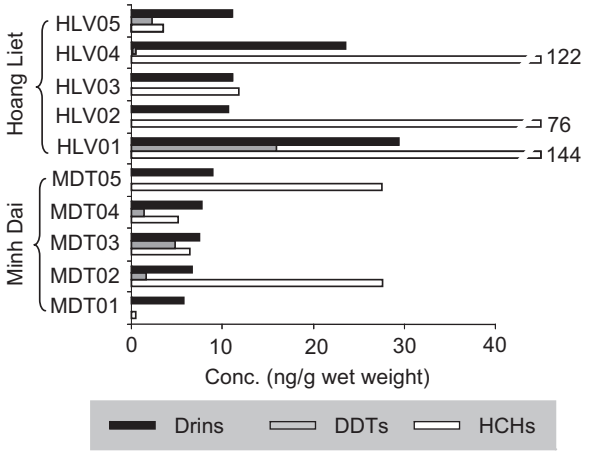

Fig. 7. Distribution of DDTs, Drins and HCHs in tea and vegetable samples.

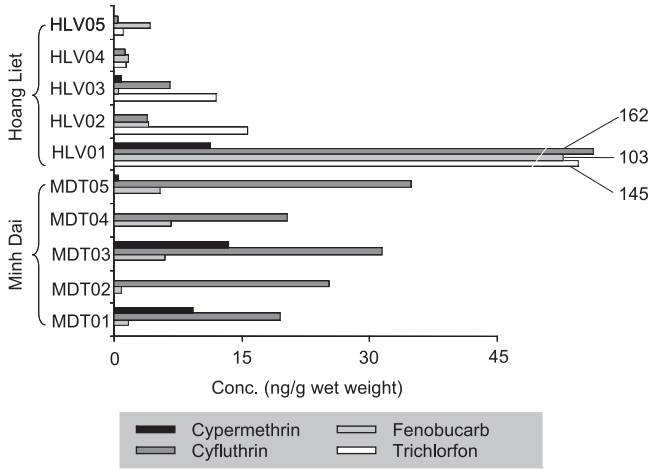

Fig. 8. Concentration of trichlorfon, fenobucarb, cyfluthrin, and cypermethrin in tea and vegetable samples.

3.3. Implication of the results

3.3.1. Recently used pesticides

The results were compared with maximum residue levels (MRLs) as set by the European Commission (EC NO 396/2005, 2005) to reflect the highest amount of pesticide residue expected in food when pesticides are applied correctly. MRLs are established for tealeaves and for different types of vegetables on dry weight basis. As the results reported here are based on wet weight, 70% moisture content of the tealeaves was estimated to calculate dry weight based pesticide concentrations (Mizukami et al., 2006). According to these calculations, the MRL for cypermethrin (0.5 μg/g) and trichlorfon (0.1 μg/g) was not exceeded in any of the samples. The MRL for cyfluthrin (0.1 μg/g) was exceeded in one sample from Minh Dai (MDT03).

The vegetables under investigation do not have established MRLs. However, comparing the reported concentration with MRLs for other vegetables in general and in case of mugwort specifically with other herbs, the mugwort sample from Hoang Liet seems to have elevated concentrations of cyfluthrin (MRL at 0.02 μg/g).

Since the MRL does not account for health risk, the reported concentrations were compared with the acceptable daily intake (ADI) values established by the European Food Safety Authority (EFSA) and the WHO/FAO Joint Expert Committee on Food Additives (JECFA). The ADI is a measure for the toxicity of a substance by long-term and repeated ingestion. Among the studied pesticides ADI was established for cypermethrin (0.02 mg/kg bw.), cyfluthrin (0.04 mg/kg bw) and trichlorfon (0.002 mg/kg bw) (all ADIs derived from WHO, 2009). A daily intake of 50 g of any of the fish samples would reach the ADI for all above-mentioned compounds (calculation for a person with 50 kg body weight; exception: one fish sample for trichlorfon). In case of cypermethrin a daily intake of 10 g fish would imply that the respective ADI would be exceeded. Tea can be considered as largely safe for use since – assuming again 70% moisture content of the leaves – a consumption of ca. 19 g dry tealeaves on a daily basis would be necessary to exceed the ADI for at least one analyzed pesticide (tea bags usually have 2.5–5 g filling weight). In case of mugwort, heartleaf and water spinach trichlorfon exceeds the ADI by a consumption of 10 g of the vegetables per day.

3.3.2. Organochlorine pesticides

DDTs, Drins and HCHs were detected in tea, vegetable and fish samples. ADI is established for DDT (PTDT, provisional tolerable daily intake, 0.01 mg/kg bw), lindane (0.005 mg/kg bw) and aldrin/dieldrin (PTDT, provisional tolerable daily intake, 0.0001 mg/kg bw) (all ADIs derived from WHO, 2009). Considering again a person with 50 kg body weight a daily intake of 50 g fish would cause that the ADI is exceeded for all samples for aldrin/dieldrin, in 60% of the samples for lindane and in 30% of the samples for DDT. In case of aldrin a daily intake from 3 g fish would cause the ADI to be exceeded. In order to not to exceed the ADI for DDT and lindane only 3 g of the tea sample MDT05 could be consumed daily (dry weight basis, calculated assuming 70% moisture content). All vegetable samples contain aldrin in a concentration which does not allow for a consumption of more than 3 g of the respective vegetable per day. ADI was exceeded for lindane in 60% of the samples by a consumption of 10 g per day.

4. Conclusions

Currently, management practices of farmers are not sustainable from the human and environmental health point of view. Increased awareness when selecting and applying pesticides and improved waste management could help to reduce pesticide concentrations in environment and food products. Differences in pesticide management practices between the two studied communes indicated that better access to information and a more homogenous production patterns lead to reduced pesticide pollution.

Cypermethrin, trichlorfon, fenobucarb, cyfluthrin, cartap, and imidachlorprid, were the most frequently used active ingredients in vegetable and tea production. Four of these currently used pesticides (cypermethrin, trichlorfon, fenobucarb, and cyfluthrin) were frequently found in the environment and biota at the two study sites. The number of samples in this study does not allow for a detailed statement regarding species or site-specific pollution patterns. However, to the best of our knowledge, these are the first residue concentrations reported for recently used pesticides in Vietnam in these food products. Recently used pesticides' environmental fate and concentration in food products deserve more attention and should be monitored regularly. Since acceptable daily intake (ADI) levels were repeatedly exceeded in fish and vegetable samples, the improvement of pesticide management practices would be crucial from a human health perspective, particularly in terms of reducing exposure.

In addition, some of the banned organochlorine pesticides still persist or where even recently added to the environment and accumulate in biomass and fish. Since most of the substances are banned or restricted, their residue concentration – provided effective control mechanisms are put in place – will continue to decline in the future as it was observed already in the past two decades (Minh et al., 2007c).

References

Berg, H., 2001. Pesticide use in rice and rice–fish farms in the Mekong Delta, Vietnam. Crop Protection 20, 897–905.

Carvalho, F.P., Villeneuve, J.P., Cattini, C., Tolosa, I., Thuan, D.D., Nhan, D.D., 2008. Agrochemical and polychlorobyphenyl (PCB) residues in the Mekong River delta, Vietnam. Marine Pollution Bulletin 56, 1476–1485.

EC NO 396/2005 of the European Parliament and of the Council of 23 February 2005 on Maximum Residue Levels of Pesticides in or on Food and Feed of Plant and Animal Origin and Amending Council Directive 91/414/EEC. http://ec.europa.eu/sanco_pesticides/public/index.cfm (accessed 22.09.07).

EPA, U.S. Environmental Protection Agency, 1996a. Florisil Cleanup EPA 3620B.

EPA, U.S. Environmental Protection Agency, 1996b. Organochlorine Pesticides by Gas Chromatography EPA 8081A.

Hai, N.X., 2006. Initial Research on Environmental Pollution in Vegetable Trade Village of Hoang Liet, Hoang Mai, Hanoi. http://english.vista.gov.vn/english/st_documents_abstract/200502185483126039/200604181693285957/200609118915316084/200609115850783994/ (accessed 23.09.07).

Hoai, P.M., Ngoc, N.T., Minh, N.H., Viet, P.H., Berg, M., Alder, A.C., Giger, W., 2010. Recent levels of organochlorine pesticides and polychlorinated biphenyls in sediments of the sewer system in Hanoi, Vietnam. Environmental Pollution 158, 913–920.

Kishida, M., Imamura, K., Maeda, Y., Lan, T.T.N., Thao, N.T.P., Viet, P.H., 2007. Distribution of persistent organic pollutants and polycyclic aromatic hydrocarbons in sediment samples from Vietnam. Journal of Health Science 53, 291–301.

Lamers, M., Anyusheva, M., La, N., Vien, N.V., Streck, T., 2011. Pesticide pollution in surface- and groundwater by paddy rice cultivation: a case study from northern Vietnam. Clean – Soil, Air, Water 39, 356–361.

Meisner, C., 2005. Poverty Environment Report: Pesticide Use and Poverty in Vietnam. World Bank, Washington. http://siteresources.worldbank.org/NIPRINT/Resources/PEN_Report_PesticideUse_in_Vietnam_Mar2005.doc (accessed 20.08.10).

Minh, N.H., Minh, T.B., Kajiwara, N., Kunisue, T., Subramanian, A., Iwata, H., Tana, T.S., Baburajendran, R., Karuppiah, S., Viet, P.H., Tuyen, B.C., Tanabe, S., 2006. Contamination by persistent organic pollutants in dumping sites of Asian developing countries: implication of emerging pollution sources. Archives of Environmental Contamination and Toxicology 50, 474–481.

Minh, N.H., Minh, T.B., Iwata, H., Kajiwara, N., Kunisue, T., Takahashi, S., Viet, P.H., Tuyen, B.C., Tanabe, S., 2007a. Persistent organic pollutants in sediments from Sai Gon-Dong Nai River basin, Vietnam: levels and temporal trends. Archives of Environmental Contamination and Toxicology 52, 458–465.

Minh, N.H., Minh, T.B., Kajiwara, N., Kunisue, T., Iwata, H., Viet, P.H., Tu, N.P.C., Tuyen, B.C., Tanabe, S., 2007b. Pollution sources and occurrences of selected persistent organic pollutants (POPs) in sediments of the Mekong River delta, South Vietnam. Chemosphere 67, 1794–1801.

Minh, T.B., Minh, N.H., Iwata, H., Takahashi, Shin, Viet, P.H., Tuyen, B.C., Tanabe, S., 2007c. Persistent organic pollutants in Vietnam: levels, patterns, trends, and human health implications. Developments in Environmental Sciences 7, 515–555.

Mizukami, Y., Sawai, Y., Yamaguchi, Y., 2006. Moisture content measurement of tea leaves by electrical impedance and capacitance. Biosystems Engineering 93, 293–299.

MONRE, Ministry of Natural Resources and Environment, 2006. http://www.monre.gov.vn/MONRENET/default.aspx?tabid=181&ItemID=13348 (accessed 24.09.07).

Sinh, N.N., Thuy, L.T.B., Kinh, N.K., Thang, L.B., 1999. The persistent organic pollutants and their management in Vietnam. In: Proceedings of the Regional Workshop on the Management of Persistent Organic Pollutant, POPs, United Nations Environment Programme, March 16–19, 1999, Hanoi, Vietnam, 385–406.

Strandberg, B., Van Bavel, B., Bergqvist, P.-A., Broman, D., Ishaq, R., Näf, C., Pettersen, H., Rappe, C., 1998. Occurrence, sedimentation, and spatial variations of organochlorine contaminants in settling particulate matter and sediments in the northern part of the Baltic Sea. Environmental Science and Technology 32, 1754–1759.

Toan, V.D., Thao, V.D., Walder, J., Schmutz, H.-R., Ha, C.T., 2007. Contamination by selected organochlorine pesticides (OCPs) in surface soils in Hanoi, Vietnam. Bulletin of Environmental Contamination and Toxicology 78, 195–200.

Ueno, E., Oshima, H., Saito, I., Matsumoto, H., Hiroshi, Y., Nakazawa, H., 2004. Multiresidue analysis of pesticides in vegetables and fruits by gas chromatography/mass spectrometry after gel permeation chromatography and graphitized carbon column cleanup. Journal of AOAC International 87, 1003–1015.

UNU [United Nations University], 2007. Environmental Monitoring and Governance in the Asian Hydrosphere: Manual for Sample Collection and Analysis. United Nations University.

UNU-EHS [United Nations University, Institute for Environment and Human Security], 2010. Final Report of the contribution of UNU-EHS to the first phase of the BMBF-funded WISDOM Project. Report to the German Federal Ministry of Education and Research.

Viet, P.H., 2002. Persistent organic pollutants in Vietnam – an overview. In: Proceedings of the Workshop on Environmental monitoring of Persistent Organic Pollutants (POPs) in East Asian Countries, Dec. 2–4, 2002, Tokyo/Tsukuba, Japan, 113-121.

WHO, 2009. Inventory of IPCS and Other WHO Pesticide Evaluations and Summary of Toxicological Evaluations Performed by the Joint Meeting on Pesticide Residues (JMPR) through 2009. http://www.who.int/ipcs/publications/jmpr/pesticide_inventory_edition10.pdf (accessed 24.09.07).