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Short Communication

Pesticide Pollution in Surface- and Groundwater by Paddy Rice Cultivation: A Case Study from Northern Vietnam

This study was designed to examine the environmental exposure of surface- and groundwater pollution in remote mountainous regions of northern Vietnam. In 2008, we monitored the loss of four commonly applied pesticides (imidacloprid, fenitrothion, fenobucarb, dichlorvos) from paddy rice farming systems to a receiving stream on the watershed scale and quantified groundwater pollution. For the entire monitoring period, runoff loss of pesticides from the watershed was estimated to range between 0.4% (dichlorvos) and 16% (fenitrothion) of the total applied mass. These losses were correlated well with the octanol–water partition coefficient and water solubility of pesticides ($r^2 = 0.78$–0.99). In the groundwater collected from eight wells, all target pesticides were frequently detected. Maximum measured concentrations were 0.47, 0.22, 0.17, and 0.07 $\mu g\,L^{-1}$ for fenitrothion, imidacloprid, fenobucarb, and dichlorvos, respectively. Our results strongly indicate that under the current management practice pesticide use in paddy fields poses a serious environmental problem in mountainous regions of northern Vietnam.

1 Introduction

During the last decade, high population growth and export-oriented economics in Vietnam have led to a major intensification of rice production. In Vietnam, more than 60% of the total farmed area is currently cultivated with rice fields; the total rice production was 35.6 million tonnes in 2007 (FAO 2008, www.faostat.fao.org). Increasing production rates in the past were enabled by the introduction of high-yield, short-duration rice varieties in tight connection to increasing applications of agrochemicals. Consequently, after the land reforms in 1988, the amount of applied pesticides doubled in the subsequent decades, reaching almost 50 000 tons per annum in 2004 (FAO 2004, www.faostat.fao.org). In Vietnam, there is concern that lowland and upland paddy rice production systems are the major non-point sources of pesticide pollution of surface- and groundwater, both of which are used for domestic purposes. Pesticides are toxic by design, calling for monitoring their impact on human health and environmental quality. Against this background, quantifying and predicting pesticide losses to rivers or wells from paddy rice fields is a crucial task. It is also a prerequisite for assessing the potential environmental exposure of water pollution across vulnerable landscapes in Vietnam.

Numerous monitoring studies from Europe and Japan have provided evidence that paddy rice cultivation is responsible for surface- and groundwater contamination, with pesticide concentrations exceeding 0.1 $\mu g\,L^{-1}$, the European drinking water quality standard [1–5]. Capri et al. [6], e.g., reviewed most of the published data from pesticide-monitoring in paddy areas of Italy. The data showed a diffuse pollution of both surface- and groundwater, with pesticide concentrations ranging from 0.1 to 30 $\mu g\,L^{-1}$. Similar results have been reported for the main rice cultivation areas in Greece [7] and Portugal [8].

In Vietnam, only few studies have focused on the environmental occurrence of pesticides in surface water [9–11], surface soil [12], or water sediments [13–15]. Moreover, these studies are limited to the large rice-growing areas of the Mekong and the Red River delta. Carvalho et al. [10], e.g., analyzed water, sediment, and bivalve mollusc samples for 70 polar and non-polar pesticide residues in the Mekong River delta. Amongst all compounds searched for, the authors detected diazinon, fenitrothion, nonylphenol, and endosulfan in most of the water samples, with concentrations ranging from 0.003 to 0.043 $\mu g\,L^{-1}$. Hung and Thiemann [9] screened water samples from the Red River, the Duong River, and from various lakes, irrigation channels and wells located in the Hanoi district for fifteen insecticides. Most of the measured concentrations were limited to the range of ng L^{-1}, except dichlorodiphenyltrichloroethane (DDT), which showed concentrations up to 0.32 $\mu g\,L^{-1}$. Pesticide contamination was highest and lowest in the rivers and wells, respectively.

No attempts, however, have been made to examine the environmental exposure of surface- and groundwater pollution in the

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remote mountainous regions of south-east Asia. The present study contributes to closing this gap. In 2008, we monitored the loss of pesticides from paddy rice farming systems to receiving streams on the watershed scale and quantified groundwater pollution in a remote catchment in northern Vietnam.

2 Material and methods

2.1 Study area

The present study was conducted in the Chieng Khoi watershed (186 hectare (ha)), Son La province, northern Vietnam (20°37′0 N, 106°4′60 E). In this mountainous area, with elevations ranging from 300 to 1000 m above see level, subsistence-oriented agriculture systems dominate. The flat plateau is exclusively used for paddy rice (*Oryza sativa* L.) cultivation, mainly for self-consumption, while the peripheral upland fields on the steep hill slopes are planted with various annual crops, often on maize intercropped with cassava. The application of pesticides is almost exclusively restricted to paddy rice fields. The tropical monsoon climate entails a rainy season from May to October and relatively dry, cold winters from November to April. The annual average temperature and rainfall is 21°C and 1200 mm, respectively. Regarding both topography and land use system, the watershed is typical for the wider area.

A reservoir was built in 1962–1968 to provide domestic and irrigation water (Fig. 1). A common irrigation system pipelines water from the main concrete channel through rice paddies into a natural stream. The irrigation system guarantees two rice growing seasons per year, from February to June ("spring season") and from July to November ("summer season").

In the spring season, crops are irrigated, whereas in the summer season they are mainly rain-fed.

2.2 Field survey

In 2008, we conducted a field survey focusing on six villages in the study area. From each village, 20 farmers were randomly selected and extensively interviewed about current pesticide use and application practices. According to this survey, insecticides were identified to constitute the main group of pesticides used in rice. We expected to find mainly the neonicotinoid imidacloprid (*N*-[1-(6-chloro-3-pyridyl)methyl]-4,5-dihydroimidazol-2-yl]nitramide), the carbamate fenobucarb (2-butan-2-ylphenyl) *N*-methylcarbamate), and the organophosphates fenitrothion ((dimethoxy-(3-methyl-4-nitrophenoxy)-thioxophosphorane) and trichlorfon (*RS*)-2,2,2-trichloro-1-dimethoxyphosphoryl-ethanol). The chemicals are active ingredients of the commercial rice insecticides Conphai 15 WP, Bassa 50EC, and Ofatox 400EC^{TM}, respectively. The former two are applied against white (Metcalfa pruinosa) and brown planthopper (*Nilaparvata lugens*), the latter against rice stem borer (*Scirpophaga incertulas*), and rice leaffolder (*Cnaphalocrocis medinalis*). Key physical–chemical properties of the pesticides are presented in Tab. 1.
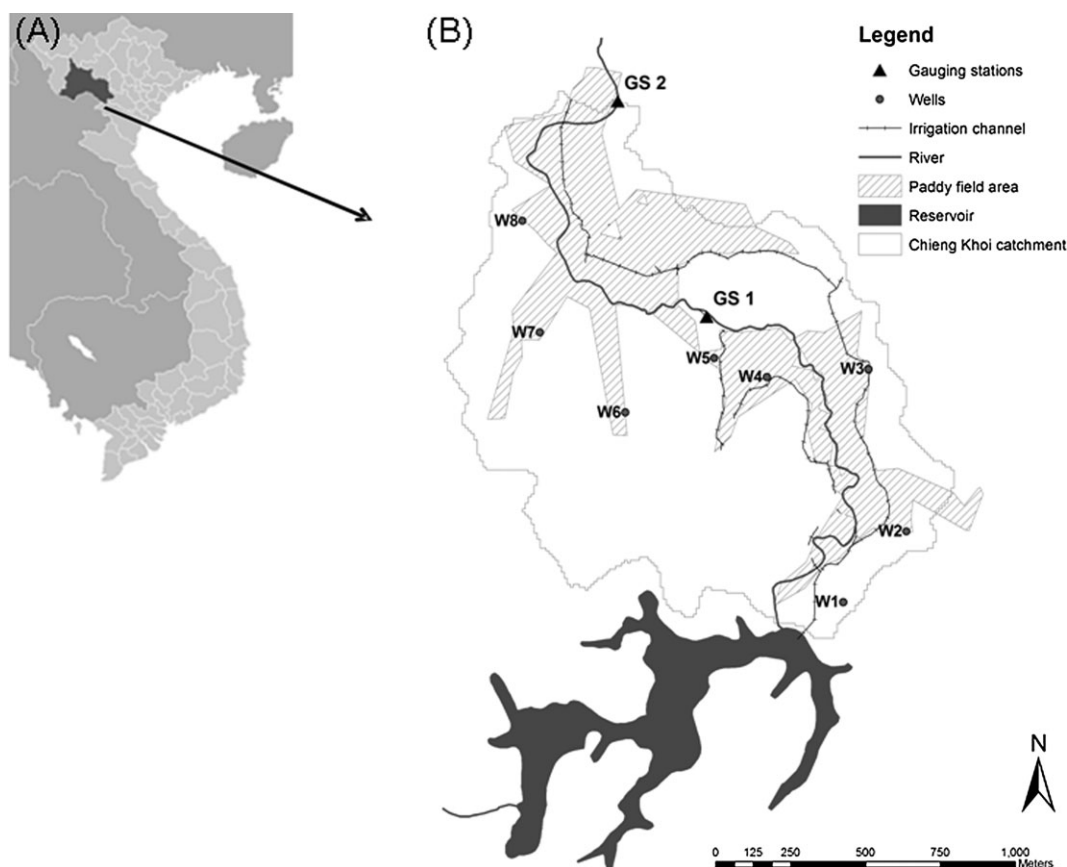
Figure 1. Location of Son La province in northern Vietnam (A) and overview of the Chieng Khoi catchment (B).

Table 1. Key physical–chemical properties of selected pesticides according to [16]

No.	Substance	Chemical group	S (mg L^{-})[a]	VP (mPa)[b]	logP_{ow}[c]	DT$_h$50[d] (d^{-1})
1	Imidacloprid	Neonicotinoid	610	9×10^{-7}	0.57	30[e]
2	Fenitrothion	Organophosphate	14	18	3.43	84 (pH 7)
3	Fenubocarb	Carbamate	420	13	2.79	17 (pH 9)[f]
4	Dichlorvos	Organophosphate	18 000	2100	1.9	2.9 (pH 7)

[a] Solubility in water.
[b] Vapor pressure.
[c] Logarithm of octanol/water partitioning coefficient.
[d] Hydrolysis half-life.
[e] Stable to hydrolysis at pH 5−11.
[f] Hydrolyzed by acids and alkalis.

2.3 Monitoring programme

A weather station (Campbell Scientific, Inc., USA) was installed within the upper part of the watershed. The weather station provides data on air temperature, relative humidity, solar radiation, wind speed, and precipitation. To monitor water fluxes and pesticide loads on the watershed scale, two gauging stations (hereinafter referred to as GS1 and GS2) were installed at the midstream and the outlet position of the watershed. The locations of the gauging stations are depicted in Fig. 1. At each gauging station, water heads were continuously measured using automatic pressure sensors (ecoTech, Bonn, Germany) and converted to flow rates using stage−discharge relationships determined by means of the velocity−area method [17]. The total rice-cropped area covered by gauging stations GS1 and GS2 is 25.1 and 64.1 ha, respectively (Fig. 1).

To assess the pesticide load, stream water was manually sampled about every 10 days starting on 17 April 2008. On each sampling day, 2 L of stream water were retrained from each position, cooled, and directly transported to the field laboratory in Yen Chau, where the samples were split. One fraction was send to the Northern Pesticide Control Centre (NPCC) in Hanoi, where it was analyzed for the insecticide imidacloprid (single measurement). The other fraction was concentrated as replicates by solid phase extraction (SPE) [18] and shipped to the University of Hohenheim, Germany, for analysis of the pesticides fenitrothion, dichlorvos, and fenobucarb.

To calculate the total mass of lost pesticides in the surface water, pesticide concentrations were interpolated linearly between data points on daily time steps. Pesticide loads were then calculated for each day by multiplying pesticide concentrations by the corresponding mean daily runoff. Subsequently, the cumulative pesticide loads were calculated by adding the daily loads for the period under study.

To assess pesticide concentrations in groundwater, eight wells located within the watershed were sampled three times in 2008 during rice cropping seasons (April, August, and September). The water samples were treated as mentioned above but without replicates. The locations of the wells are shown in Fig. 1. All selected wells had been constructed between 1995 and 2001 for drinking water and domestic purposes.

2.4 Pesticide analysis

All pesticides other than imidacloprid were analyzed at the State Institute of Agricultural Chemistry in Hohenheim (certified laboratory) using a gas chromatograph system (HP 6890, Hewlett Packard, USA) equipped with a nitrogen−phosphorus detector (Agilent Technologies, USA), an application-specific column for organophos-

phorus pesticides (Rtx-OPPPesticides (Fused Silica), 30 m × 0.25 mm; Restek, USA) and an autosampler (7683 Series Auto Sampler, Hewlett Packard, USA). However, trichlorfon was not analyzed due to shortcomings of the analytical procedure as reported elsewhere [19]. Therefore, the organophosphate dichlorvos as a degradation product of trichlorfon was added to the catalogue of target pesticides. The detection limits were calculated based on the signal-to-noise ratio approach ($S/N = 3$). Using a 400-fold concentration factor and corresponding instrumental detection limits for each analyte, the method detection limits for imidacloprid, fenitrothion, fenobucarb, and dichlorvos were estimated to be 0.001, 0.008, 0.013, and 0.008 µg L^{-1}, respectively. More detailed information about the analytical procedure and analytical quality assurance can be found in [20].

3 Results and discussion

3.1 Pesticide application

Our field survey revealed that 95, 85, and 5% of all interviewed farmers applied the commercial rice insecticides Conphai 15WP, Ofatox 400EC, and Bassa 50EC, respectively. During the spring rice crop season 2008, pesticides were mostly applied between 10 and 15 April using a backyard sprayer. Several farmers repeated pesticide application on 5 and 6 May. During the summer season, pesticides were applied between 10 and 15 August. On average the farmers applied 0.12, 0.46, 0.53, and 0.53 kg ha^{-1} imidacloprid, fenobucarb, fenitrothion, and trichlorfon, respectively, on the paddy fields. A total amount of 2.9 kg (7.4 kg), 13.4 kg (34.1 kg), and 11.6 kg (29.5 kg) of imidacloprid, fenitrothion (trichlorfon), and fenobucarb was applied on rice fields of the rice area drained by GS1 (GS2) during spring season 2008. Note that the average amount of pesticides applied by the farmers is nearly twice as high as the upper dosage recommended by the distributing companies.

3.2 Surface water

During the monitoring period (April–June) no pesticides were detected in river and irrigation water samples taken at locations close to the reservoir outlet (Fig. 1), indicating that the target pesticides were applied exclusively on rice fields located downstream. Pesticide concentrations in the surface water and the cumulative pesticide loads are depicted in Fig. 2. After the first application, all tested pesticides were detected in the river water at least once at both gauging stations. Only imidacloprid was detected above the detection limit at all six sampling dates at both gauging stations. By
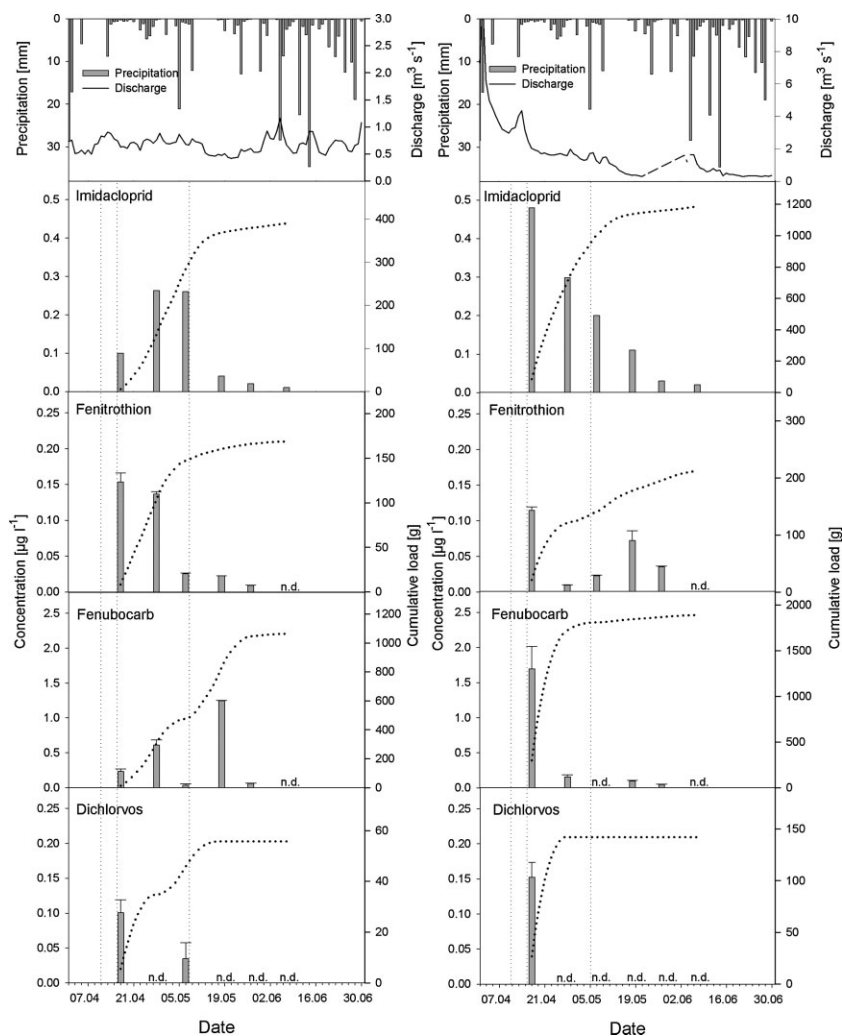
Figure 2. Concentration and cumulative load of the insecticides measured at a midstream position (left, GS1) and at the main outlet (right, GS2). Error bars represent standard deviations. The vertical dotted lines represent the days of pesticide application. The dashed discharge line indicates measurement gap filled by linear interpolation (n.d.: not detectable).

contrast, dichlorvos was detected only at the beginning of the measurement period, that is the first and third and the first sampling date at GS 1 and GS 2, respectively.

The mean measured concentrations at GS1 and GS2 could be clearly ranked in the following order: fenobucarb (0.36 and 0.33 µg L^{-1}), imidacloprid (0.12 and 0.19 µg L^{-1}), fenitrothion (0.06 and 0.04 µg L^{-1}), and dichlorvos (0.02 and 0.03 µg L^{-1}). Concentration values below the detection limit (nd) were set to zero. Among all pesticides, fenobucarb exhibited the highest measured concentrations with 1.25 (18 May) at GS1 and 1.70 µg L^{-1} (17 April) at GS 2 (Fig. 2).

During the entire monitoring period, in total 1.80 (1.06), 0.21 (0.17), 1.19 (0.39), and 0.14 (0.06) kg of fenobucarb, fenitrothion, imidacloprid, and dichlorvos were lost from the watershed at GS1 (GS2) by surface runoff, corresponding to 6.4 (9.2), 0.6 (1.3), 16.0 (13.4), and 0.4% (0.4%) of the applied pesticide mass, respectively. Figure 3 indicates that the physical–chemical properties water solubility and octanol–water partition coefficient of the parent pesticides determine the cumulative runoff losses. The runoff losses significantly increase with increasing water solubility and with decreasing octanol–water partition coefficient. With paddy rice, the situation of pesticide transport to adjacent environmental compartments is somewhat special, in that a considerable fraction of applied pesti-

cides directly enters the ponded surface water surface water with or shortly after application. Paddy fields are typically arranged in cascades and connected by channels or outlets favoring surface runoff to receiving streams. Hence, under the common management practices highly soluble pesticides are more prone to be transported directly to the stream via surface runoff shortly after application.

The results are in line with published data on pesticide loss with surface water in rice-cultivating areas in Asia. Sudo et al. [4], e.g., continuously measured the input of different herbicides from a paddy rice dominated watershed (3174 km^2) to Lake Biwa, Japan. The authors estimated the annual loss of herbicides to be in the range of 3% (pretilachlor) to 14.5% (bromobutide). Based on a comprehensive sampling pattern, [2] calculated the total runoff loss of nine kinds of herbicides shortly after application from paddy fields to the Kozakura River, Japan, to be in the range of 8–22%. The authors reported high correlations between runoff rates and physical–chemical pesticide properties. For a watershed (97 ha) in Japan mainly cultivated with paddy rice (86 ha), [21] reported cumulative losses of 8.8 and 7.7% of the applied mass of the pesticides dymrom and imazosulfuron, respectively. Numabe and Nagahora [22] measured pesticide runoff rates over a 3-year period in an watershed in Japan ranging from 0.3% for fenithion to 42% for benfuresate, indicating a high correlation between runoff rate of the pesticide and water solubility.
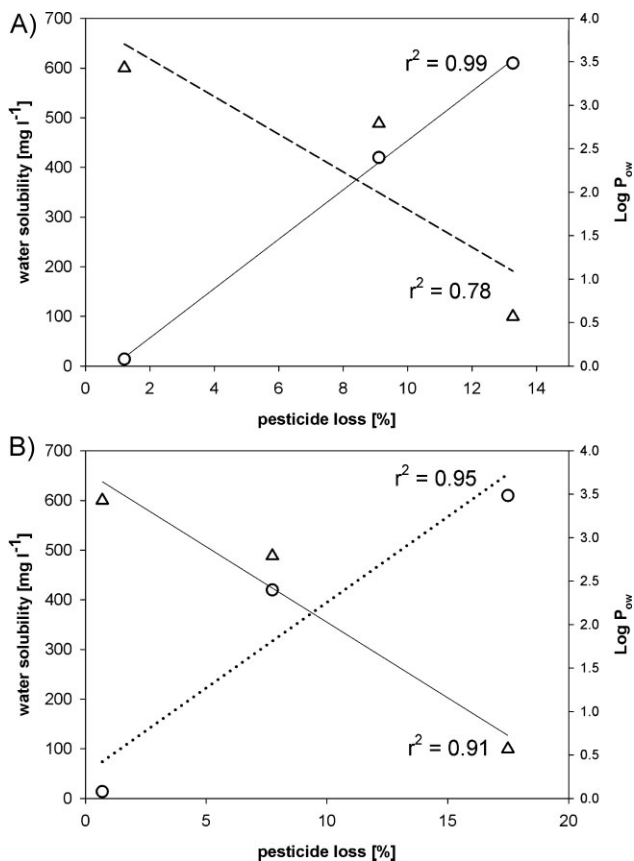
Figure 3. Correlation between total cumulative pesticide loss and water solubility or octanol–water partition coefficient for the insecticides imidacloprid, fenitrothion, and fenobucarb measured at sampling points GS1 (A) and GS2 (B).

3.3 Groundwater

Measured concentrations of target pesticides in groundwater sampled from eight wells (W1–W8) in the Chieng Khoi catchment are listed in Tab. 2. During the monitoring period, imidacloprid,

fenobucarb, and dichlorvos were detected in almost half (45%) of the groundwater samples. In contrast, concentrations of fenitrothion exceeding the detection limit were found in every water sample. The mean measured concentration for all wells and sampling dates could be clearly ranked in the following order: imidacloprid ($0.16 \, \mu g \, L^{-1}$), fenitrothion ($0.10 \, \mu g \, L^{-1}$), fenobucarb ($0.025 \, \mu g \, L^{-1}$), and dichlorvos ($0.007 \, \mu g \, L^{-1}$). Concentrations of fenitrothion, imidacloprid, fenobucarb, and dichlorvos in groundwater samples exceeded the European threshold for drinking water of $0.1 \, \mu g \, L^{-1}$ in 46, 13, 8, and 0% of all samples, respectively.

Imidacloprid was detected on every sampling date in water samples taken from W2 and W4. Highest imidacloprid concentrations of 1.15 and $1.53 \, \mu g \, L^{-1}$ were measured in samples from W4 and W5 on 28 August 2 to 3 wk after pesticide application for the summer crop season.

On sampling date 19 September, the fenitrothion concentration exceeded $0.1 \, \mu g \, L^{-1}$ in every sampled well. Highest concentrations of 0.46 and $0.48 \, \mu g \, L^{-1}$ were measured in samples taken from W1 and W2, respectively. For the spring crop season, however, the concentration exceeded the European threshold only in groundwater sampled from W5.

Fenobucarb was detected predominantly during the spring crop season (25 April), with concentrations ranging from 0.03 to $0.11 \, \mu g \, L^{-1}$. Dichlorvos, however, was detected only sporadically. Measured concentrations never exceeded $0.1 \, \mu g \, L^{-1}$. These results are in agreement with reported data from elsewhere. Bouwman et al. [23], e.g., measured pesticide concentrations in 54 domestic wells under rice-based production systems in Philippines from 1989 to 2000. The authors reported pesticide concentrations generally one to two orders of magnitude below the European drinking water quality standard. However, peak concentrations of $4.17 \, \mu g \, L^{-1}$ were detected temporarily.

In conclusion, our results strongly indicate that, under the current management practice, pesticide use in paddy fields poses a serious environmental problem in mountainous regions of northern Vietnam. Surface- and groundwater is multiply re-used for domestic purposes. This makes it indispensable to quantify and forecast pesticide losses to groundwater and surface water from paddy rice fields. Due to logistic problems in this remote area, however, the sampling intervals of the present study were too wide to accurately

Table 2. Concentrations of selected pesticides measured in groundwater samples taken from eight wells located in the Chieng Khoi sub-catchment. Values exceeding the European threshold for drinking water of $0.1 \, \mu g \, L^{-1}$ are printed in bold (n.d.: not detectable)

Sampling date	Well 1	Well 2	Well 3	Well 4	Well 5	Well 6	Well 7	Well 8
Imidacloprid ($\mu g \, L^{-1}$)								
April 25	n.d.	0.046	0.052	0.069	0.019	n.d.	n.d.	n.d.
August 28	n.d.	0.22	n.d.	1.15	1.53	n.d.	n.d.	n.d.
September 19	n.d.	0.06	0.03	0.06	n.d.	n.d.	0.01	n.d.
Fenitrothion ($\mu g \, L^{-1}$)								
April 25	0.004	0.002	0.021	0.005	**0.117**	0.003	0.002	0.003
August 28	0.004	0.005	0.007	0.005	**0.127**	0.007	**0.159**	0.002
September 19	**0.461**	**0.476**	**0.231**	**0.206**	**0.253**	**0.101**	**0.155**	**0.140**
Fenobucarb ($\mu g \, L^{-1}$)								
April 25	0.029	n.d.	0.056	**0.110**	0.073	0.032	0.050	0.028
August 28	n.d.	n.d.	n.d.	n.d.	0.030	n.d.	**0.168**	n.d.
September 19	n.d.	0.013	n.d.	n.d.	0.010	n.d.	n.d.	n.d.
Dichlorvos ($\mu g \, L^{-1}$)								
April 25	n.d.	n.d.	0.008	0.003	0.007	n.d.	n.d.	0.004
August 28	0.016	n.d.	n.d.	n.d.	0.015	n.d.	0.069	n.d.
September 19	0.015	0.005	n.d.	0.007	n.d.	n.d.	n.d.	0.023

capture peak concentrations, which would be important to precisely assess the total loss of pesticides from the area. More extensive measurements are needed to strengthen our conclusion and to improve our understanding of the factors that control pesticide losses from paddy rice systems to surface- and groundwater on the watershed scale under the environmental conditions of southeast Asia. Future studies, e.g., should focus on higher resolved temporal and spatial sampling strategies to favor a more detailed analysis of the temporal pattern of pesticide concentration in surface and groundwater. This would be an important step in evaluating management strategies to minimize the exposure of water pollution.

Acknowledgments

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