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[redacted]

However, shrimp production is a risky venture, due to high investment requirements, market price fluctuations, varying salinity levels, and the high vulnerability of shrimps to bacterial and viral diseases as well as to agrochemical residues. In the Mekong Delta, paddy rice production has intensified during the last decades in order to maintain both national food security and Vietnam's leading position as one of the largest rice exporter worldwide (Ricepedia, n.d.; Toang, 2017). This intensification involved the use of high yield varieties as well as increased applications of fertilizer and pesticide (Phong et al., 2010; Anyusheva et al., 2012), resulting in a widespread environmental pollution particularly in surface water bodies, drinking water resources, as well as in soil and sediments (Toan et al., 2013; Chau et al., 2015a). Residues of currently used pesticides were determined in concentrations of up to 521 $\mu g\,kg^{-1}$ (buprofezin) in soil and sediment of paddy rice fields in the Mekong Delta (Toan et al., 2013). Surface water samples contained pesticides in the range of 0.15 (fenobucarb) to 1.10 $\mu g\,L^{-1}$ (difenconazole) (Chau et al., 2015a). Shrimps are also very sensitive to bacterial diseases (Sebesvari et al., 2012; *Own interview data*, 2015) and in the event of an outbreak, farmers typically lose the whole production cycle (*Own interview data*, 2015). To control this risk, farmers often apply prophylactic probiotics and veterinary antibiotics to their shrimp system (Holmström et al., 2003; Sapkota et al., 2008). The use of antibiotics is well documented in permanent shrimp production (e.g., Anh et al., 2010) but less so in the alternating rice-shrimp farming systems. Previous studies showed maximum antibiotic concentrations in sediment of shrimp ponds of 0.82 $\mu g\,kg^{-1}$ for sulfamethoxazole and 0.73 $\mu g\,kg^{-1}$ for trimethoprim in the Mekong Delta (Le and Munekage, 2004). Shimizu et al. (2013) detected sulfamethoxazole concentrations in the range of 0.002 to 0.914 $\mu g\,L^{-1}$ in shrimp pond water in Can Tho province of the Mekong Delta. The impact of antibiotic residues in the environment was demonstrated by Le et al. (2005) and Gao et al. (2012) who found antibiotic-resistant bacteria in water and sediment samples of shrimp and aquaculture farms. In any case, inasmuch as pesticides are to be avoided in shrimp habitats, the same holds true for antibiotics in the paddy rice fields. Antibiotic contamination in the soil of rice fields might lead to an accumulation of antibiotics in rice and rice-based food products and further to selection of resistant bacteria in the food chain, particularly when rice plants may take up the antibiotics from the soil (Dolliver et al., 2007). As the Mekong Delta is a system where the Mekong distributaries are connected by numerous man-made irrigation canals, it could easily allow pesticide and antibiotic cross-contamination between land use systems.

To estimate direct effects of land use systems on the environment, especially on drinking water quality, most studies analyse pesticides concentrations in water samples (e.g., Chau et al., 2015a; Lamers et al., 2011). However, most antibiotics and many of the commonly used pesticides tend to accumulate in the soil matrix because of their physico-chemical properties, i.e., low water solubility and rather polar properties (e.g., Müller et al., 2007; Thiele-Bruhn, 2003). Furthermore, concentrations of pesticides and antibiotics in water are highly influenced by environmental factors including heavy rain events or flow velocity in the channel/river, which often mean short-term exposure (Schäfer et al., 2011) as opposed to potential long-term exposure in soil. Consequently, to determine the prevalence of antibiotics and pesticides in different agricultural systems and to describe the pollution background of different land use systems, soil samples as long-term environmental compartments are more suitable. Even though, the high input of either pesticides or antibiotics in the prevailing agricultural land use systems (permanent rice, alternating rice-shrimp, and [redacted] major agricultural land use [redacted]

Therefore, the aim of this study was to understand actual pollution patterns and estimate the impact of current land use change pattern in terms of pollution. This is particularly relevant because with increased salinity intrusion, changes in agricultural land use systems towards more saline shrimp aquaculture could become a main adaptation strategy in the future. In this research, we first assessed the general management and particularly the use and application pattern of pesticides and antibiotics in the three prevailing land use systems of the coastal Mekong Delta: i) permanent rice system ii) alternating rice-shrimp systems, and iii) permanent shrimp systems. Second, we identified pesticide and antibiotic pollution patterns of these land use systems to provide the basis for a discussion about expected changes in the pollution patterns with changes of agricultural land use systems. To do so, we carried out household surveys with local farmers involved in the major land use systems and analysed frequently used pesticides and antibiotics in soil samples of permanent paddy rice fields, alternating rice-shrimp fields and permanent shrimp ponds in two coastal provinces of the Mekong Delta.

## 2. Material and methods

### 2.1. Study sites

The Vietnamese Mekong Delta is located at the southwestern tip of Vietnam. This low-lying area is intersected by a dense and complex network of rivers, channels and flood plains (Tuan et al., 2007). The climate is tropical monsoonal (mean annual precipitation: 1660 mm, average annual temperature: 27 °C), with a wet season from May to October (Delta Alliance, 2011). Up to 64% of the area in the delta is used for agricultural production (GSO, 2015). In Bến Tre the population density is 528 persons/km$^2$ (GSO, 2016) with 58.4 thousand ha paddy rice (GSO, 2017) and 45.2 thousand ha of aquaculture (GSO, 2017). In Sóc Trăng the population density is 369 persons/km$^2$ (GSO, 2016) with 348.2 thousand ha paddy rice and 74.1 thousand ha aquaculture (GSO, 2017).

For the purposes of this study, study sites were located in the districts My Xuyen and Vinh Chau in the province Sóc Trăng and one site was located in the district Thạnh Phu in the province Bến Tre. In both coastal provinces a salinity gradient was represented by three different land use systems (Renaud et al., 2015; *Own interview data*, 2015): i) permanent rice, ii) alternating rice-shrimp and iii) permanent shrimp (Fig. 1). In permanent rice areas, paddy rice is cultivated with two or three harvests per year. In the alternating rice-shrimp systems, shrimp and rice production are alternating between the dry and the wet season, respectively. In the permanent and intensive shrimp systems farmers cultivate two to four production cycles of shrimps per year (*Own interview data*, 2015).

### 2.2. Household survey

To understand farm management and in particular the pattern of use of agrochemicals in the three different land use systems, and to identify suitable agricultural fields for soil sampling, household surveys were carried out using semi-structured questionnaires from October 2015 to January 2016. In both provinces, 20 farmers were interviewed for each agricultural land use system, resulting in total of 120 household interviews using purposive sampling. The farmers had to fulfill certain criteria to be selected for the study: i) the interviewee is the actual manager of the particular field/pond at least since five years, ii) distance between the fields/ponds of the farmer should be as far as possible within the provinces to cover a large area, and iii) the farmer

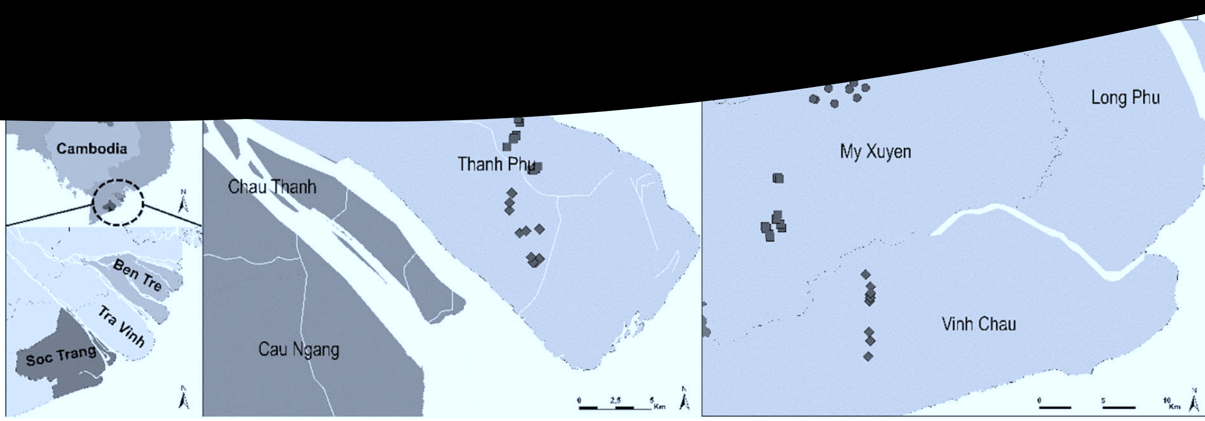


**Fig. 1.** Location of the study sites; symbols mark the sampled farms with different symbols representing the particular agricultural land use systems.

was willing to give permission to take soil samples from his/her fields. For each agricultural land use system, individual semi-structured questionnaires were developed. The first part of the questionnaire focused on general management of the farm and the second part on pesticide and/or antibiotic management.

### 2.3. Soil sampling

Based on interviews with local authorities and farmers for each land use system, ten representative field replicates were selected and sampled in January and February 2016 (see sampling Scheme SI 1). In these months, shrimp production is paused which enables the soil sampling. During the shrimp season the water level in the pond exceeds 2 m and the farmer's would not allow sampling at that time. In the alternating rice-shrimp systems, soil of the platform for rice-cropping and of the surrounding ditches for the shrimp cultivation, were sampled. In permanent rice systems (n = 20) and platforms of the alternating rice-shrimp system (n = 20), topsoil (0 to 15 cm) was sampled using a stainless steel shovel. In permanent shrimp systems (n = 20) and in ditches of the alternating rice-shrimp system (n = 20) top soil (0 to 10 cm) was taken using a sediment corer with 7 cm diameter (Hydro-Bios, Kiel, Germany) (Standard soil properties in Table S1). Sludge, defined as “uppermost fresh organic rich sedimented layer” was not sampled because it is typically removed by the farmers after the shrimp season at variable time intervals, for instance nearly every year in some shrimp ponds and in others whenever the farmer had time for this (*Own interview data*, 2015). Sludge and soil was differentiated on site based on differences in color and density. In total 80 composite samples were taken, comprising sampling from five randomly selected positions in each system or system compartment. Samples were directly packed in aluminum foil, cooled for short-distance transport (freezer −4 °C), frozen for the transport to Germany and stored at −20 °C until analysis.

### 2.4. Studied pesticides and antibiotics

The selection of the target pesticides and antibiotics were based on the following criteria: i) use by the farmers (frequency and amount applied) derived from former studies, the results of the household survey, and for pesticide, through observation of the presence of empty packages of commercial pesticide products in the study area, and ii) physico-chemical properties affecting environmental fate (i.e., water solubility, half-life values, Log $K_{ow}$, Tables 1 and 2). For pesticides, the risk potential for living organism based on the WHO toxicity classification (WHO, 2010), were also included for the selection. For antibiotics the application also in human medicine which could constitute a high risk of resistance formation with impact on the human health care by potential residues in the environment was one selection criterion.

### 2.5. Pesticide analysis

Pesticides were analysed as described by Braun et al. (2018). Prior to extraction processes, soil samples were lyophilized and sieved to < 2 mm. Delta-hexachlorocyclohexane (delta-HCH) has been added prior pesticide extraction. The selection of delta-HCH seemed justified since first test had revealed that the samples were free of this compound; however this assumption was apparently wrong for the whole sample set. During analyses, delta-HCH recovery eventually reached > 120%, pointing to additional overlapping target ion masses. Hence, delta-HCH was not further considered. Samples (15 g dry weight per sample) were extracted using accelerated solvent extraction (ASE; Dionex 350, temp. 100 °C, 5 min static time, two extraction cycles) after a modified method of Villaverde et al. (2008) using the solvent mixture of n-hexane: acetone (1:1, v/v). Prior to clean-up step, 300 µL toluene were added to the extract volume and reduced to 1.5 mL via rotary-evaporation. Subsequently, samples were cleaned-up according to Laabs et al. (2007), by using solid phase extraction. Then the extracts were loaded in glass cartridges, containing 1 g of aluminum oxide (Merck, Darmstadt, Germany) and 1 g Florisil (Merck, Darmstadt, Germany). The extracts were consecutively eluted with 20 mL diethylether:n-hexane (1:1, v/v) and the sample volume was reduced via rotary-evaporator. The samples were spiked with fluorene-$d_{10}$ as an internal standard for quantification. Pesticides were analysed via GC–MS (Agilent Technologies 6890N, Böblingen, Germany), equipped with Optima 5 MS column (30 m length × 0.25 mm ID × 0.5 µm film thickness; Macherey & Nagel, Düren, Germany). Helium was employed as carrier gas with a constant flow rate of 1 mL min$^{-1}$; injection volume was 1 µL using splitless mode. The MSD operated in selected ion monitoring (SIM) mode. The injector block was set to 250 °C. The oven parameters were set to the following conditions: initial temperature 85 °C held for 2.5 min, temperature increased 15 °C min$^{-1}$ until 220 °C were reached, followed by an increase to 280 °C with 10 °C min$^{-1}$ and held for 5 min and finally increased to 300 °C with 10 °C min$^{-1}$ and held for 5 min. For quality assurance and control, laboratory blank samples were introduced for each analytical batch. Additional lab replicates per field replicate were performed on every fifth sample. Method verification was done via recovery experiments at two concentration levels (n = 2; 0.2 and 2.5 µg for each pesticide in 15 g dry soil) based on previous work in a similar area of the Mekong Delta (Toan et al., 2013), using reference soil (RefeSoil 03-G, Frauenhofer Institut, Schmalenberg, Germany). The recovery for all studied pesticides was in the range of 70 to 120% (for details see Table S2). Routine limit of quantification (RLOQ) was for the majority of the pesticides 0.66 µg kg$^{-1}$ (lowest

[redacted]

| | | | | | | classes[b] | Chemical formula[a] |
|---|---|---|---|---|---|---|---|
| *Insecticide* | | | | | | | |
| Chlorpyrifos | 4.70 | 1.1 | 8151 | 21 | 150 | II | $C_9H_{11}Cl_3NO_3PS$ |
| Quinalphos | 4.44 | 17.8 | 1465 | 21 | 375 | II | $C_{12}H_{15}N_2O_3PS$ |
| Fipronil | 3.75 | 3.8 | – | 225 | 40 | III | $C_{12}H_4Cl_2F_6N_4OS$ |
| Fenobucarb | 2.78 | 420 | – | 18 | 600 | II | $C_{12}H_{17}NO_2$ |
| *Fungicide* | | | | | | | |
| Difenconazole | 5.44 | 15.0 | – | 130 | 63 | II | $C_{19}H_{17}Cl_2N_3O_3$ |
| Fludioxonil | 4.12 | 1.8 | 145,600 | 20 | 4.6 mL kg$^{-1}$[d] | U | $C_{12}H_6F_2N_2O_2$ |
| Tebuconazole | 3.78 | 36 | – | 47 | 250 | II | $C_{16}H_{22}ClN_3O$ |
| Propiconazole | 3.72 | 150 | 1086 | 214 | 50 | II | $C_{15}H_{17}Cl_2N_3O_2$ |
| Isoprothiolane | 3.30 | 54 | 1352 | 320 | 480 | II | $C_{12}H_{18}O_4S_2$ |
| Azoxystrobin | 2.50 | 6.7 | 589 | 180.7 | 100 | III | $C_{22}H_{17}N_3O_5$ |
| *Herbicide* | | | | | | | |
| Fenoxaprop-p-ethyl | 4.60 | 0.7 | 11,354 | 0.4 | 28 | U | $C_{18}H_{16}ClNO_5$ |
| Pretilachlor | 4.08 | 500 | – | 30 | 360 | U | $C_{17}H_{26}ClNO2$ |

[a] Source: PPDB (Pesticide properties database).
[b] WHO toxicity classes: Class III: slightly hazardous, Class II: moderately hazardous, U: unlikely to present acute hazard (WHO, World Health Organization, 2010).
[c] Estimated via the prescribed dosage of the commercial products.
[d] Dosage refers to seed treatment with ml/kg$^{-1}$ seed.

detected calibration standard: 0.01 µg kg$^{-1}$), for propiconazole, difenconazole and quinalphos RLOQ was 3.3 µg kg$^{-1}$ (lowest detected calibration standard 0.05 µg kg$^{-1}$) and for tebuconazole 6.6 µg kg$^{-1}$ (lowest detected calibration standard 0.05 µg kg$^{-1}$). Pesticides standards were supplied by LGC Standards (Wesel, Germany) with a purity > 97%. All used solvents were HPLC grade.

### 2.6. Antibiotic analysis

Samples (one sample 15 g dry weight) were lyophilized and sieved < 2 mm. The extraction was performed after Dalkmann et al. (2012) using accelerated solvent extraction (ASE; Dionex 350), with two extraction cycles including methanol:water solution (50:50; v/v) according to Gobel et al. (2005) and 50 mM aqueous phosphoric acid:acetonitrile solution (50:50; v/v) according to Golet et al. (2002). Clean-up was performed after Dalkmann et al. (2012): an aliquot of the sample extract (20 mL) was diluted with 300 mL of water and pH was adjusted to 2.4. Isotope-labelled standards were added to the samples before loading the sample on anion exchange cartridge (Chromabond SB, Macherey-Nagel, Düren, Germany) and adsorbent cartridge (OASIS HLB, Waters, Milford, United States). Samples were eluted from the column with respectively 5 mL of methanol, acetonitrile and acidified acetonitrile (0.1% 12 M HCL). After reducing the volume via rotary-evaporator close dryness, 1 mL of 50 mM phosphoric acid:acetonitrile solution (80:20, v/v) were added and filled into glass vials before measurement.

Antibiotics were analysed using liquid chromatograph tandem mass spectrometry (LC-MS/MS) equipped with a TSQ Quantum Ultra spectrometer (Thermo Finnigan, Dreieich, Germany) and a heated electrospray ionization ion source (HESI), operating in positive mode. The chromatograph was equipped with an XBridge C18 column (3.5 µm, 2.1 × 150 mm; Waters, Milford, MA, USA) combined with a guard column (Sentry 2.1 × 10 mm, Waters, Milford, MA, USA). The flow rate was 300 µl min$^{-1}$. Acidified methanol (A) and acidified Millipore water (B) (0.1% $CH_2O_2$) were used as solvents. Gradient started with 5% A, increasing linearly to 60% after 15 min. Further 16 min A was raised to

**Table 2**
Selected antibiotics and their properties.

| Compound | Water solubility 20 °C g L$^{-1}$ | Octanol-water partition coefficient log$K_{ow}$ | Soil- water partitioning coefficient $K_D$ L kg$^{-1}$ | pKa | Chemical formula[b] |
|---|---|---|---|---|---|
| *A) Sulfonamide* | | | | | |
| Sulfamethazine | 1.5[d] | 0.89[b] | 0.6 to 6.7[f] | 2.4;6.9[b] | $C_{12}H_{14}N_4O_2S$[b] |
| Sulfadiazine | 0.1[d] | −0.09[b] | 2.0[c] | 2.0;6.9[b] | $C_{10}H_{10}N_4O_2S$[b] |
| Sulfamethoxazole | 2.8[a] | 0.66[a] | 1.5[g] | 1.4;5.8[a] | $C_{10}H_{11}N_3O_3S$[b] |
| *B) Diaminopyrimidines* | | | | | |
| Trimethoprim | 1.0[a] | 0.59[a] | 9.7[g] | 3.2;6.7[e] | $C_{14}H_{18}N_4O_3$[b] |
| *C) Fluoroquinolones* | | | | | |
| Ciprofloxacin | 0.5[a] | 1.63[a] | 496[h] | 6.4;8.7[a] | $C_{17}H_{18}FN_3O_3$[b] |
| Enrofloxacin | 0.1[a] | 2.31[a] | 427[h] | 6.4;7.8[a] | $C_{19}H_{22}FN_3O_3$[b] |

[a] Dalkmann et al. (2012).
[b] https://www.drugbank.ca/drugs/DB00537, accessed April 4, 2018.
[c] Thiele-Bruhn et al. (2004).
[d] Pubchem Open Chemistry Database.
[e] Qiang and Adams (2004).
[f] Lin and Gan (2011).
[g] Liu et al. (2010).
[h] Nowara et al. (1997).

[redacted] gas and helium as collision gas (0.2 Pa). Method recoveries of the target antibiotic varied between 54 and 105% (Table S2). To determine the lower limit of quantitation for each substance the RLOQ was defined, which was 0.03 μg $kg^{-1}$ for all antibiotics (lowest detected calibration standard: 0.1 μg $L^{-1}$). Antibiotic standards and isotope-labelled, for internal standards, ciprofloxacin (carboxyl-$^{13}C_3$, quinolone-$^{15}N$, ≥98% pure), enrofloxacin hydrochloride (ethyl-$d_5$, ≥98% pure), trimethoprim (methyl-$^{13}C_3$, ≥98% pure), sulfamethoxazole (ring-$^{13}C_6$, ≥98% pure) and sulfamethazine (sulfamethazine-$D_4$, utilized also for sulfadiazine) were supplied by LGC standards (Wesel, Germany).

### 2.7. Statistical analysis

Statistical analyses were performed using IBM SPSS statistics 24. A Chi-quadrat test was used to test whether the use of pesticides and antibiotic, which the farmers named in the interviews, is dependent from the agricultural land use. For the analysis of differences of pesticide and antibiotic pollution between the different agricultural land use system and provinces, the data groups were first checked for normal distribution using a Shapiro-Wilk Test at $p = 0.05$. Differences in the antibiotic pollution (sulfonamide, fluoroquinolone and diaminopyrimidines concentration) between the two provinces were tested with Mann-Whitney-*U* test at $p = 0.05$ with the antibiotic pollution as the dependent variable and the provinces as the independent variable. For the statistical analysis the Kruskal-Wallis Test at $p = 0.05$ and a post-hoc Test (Dunn-Bonferroni-Test) were employed to test for differences i) in the level of pesticide concentration (insecticide, herbicide and fungicide concentrations) of the different agricultural land use systems of the respective province; ii) in the concentration of insecticides, fungicides and herbicides of the different agricultural land use systems; iii) in the antibiotic concentrations (sulfonamide, fluoroquinolone and diaminopyrimidines concentration) of the different agricultural land use system in the respective province; and finally to test for differences of the individual sulfonamide, fluoroquinolone and diaminopyrimidines concentrations of the agricultural land use system in the respective province. Individual pesticide and antibiotic concentration, fungicide, herbicide, insecticide, sulfonamide, fluoroquinolone, total pesticide pollution and total antibiotic pollution were defined as the dependent variable and different land use systems were the independent variable. Individual pesticide and antibiotic concentration, fungicide, herbicide, insecticide, sulfonamide, fluoroquinolone, total pesticide load and total antibiotic load were defined as the dependent variable and different land use systems were the independent variable.

## 3. Results

### 3.1. Household interviews

Household interviews revealed that in permanent rice systems, pesticide usage (90 to 100% of farmers) was significantly higher ($p > 0.05$) than in alternating rice-shrimp systems (10 to 90%) and permanent shrimp systems (0%, Table 3). In alternating rice-shrimp systems, pesticide use differed strongly between the two provinces. In Sóc Trăng, farmers used pesticides to nearly the same extent than permanent rice farmers, while in Bến Tre only 10% of alternating rice-shrimp farmers used pesticides as they protected their rice crops through seed pre-treatments (pickling) with fungicides (e.g., difenconazole) and/or insecticides (e.g., fipronil) (Table 3; *Own interview data,* 2015). The average application frequency of pesticides in the permanent rice system was more than twice as high as in the alternating rice-

[redacted] application of antibiotics was [redacted] than in the shrimp production of the alternating rice-shrimp systems ($p < 0.05$) (Table 3). Application frequency of antibiotics differed considerably between individual farmers. Mostly, farmers applied antibiotics during the juvenile phase, when shrimps are more vulnerable to diseases (*Own interview data,* 2015). During this period, farmer's applied antibiotics three times a day for a period of seven days in combination with feed (*Own interview data,* 2015). Here, one farmer applied antibiotics four times per day during the whole production cycle and stopped using antibiotics approximately five days before the harvest, which ended up to 336 antibiotic dispensations. In both provinces, the range of average total treatment applications of antibiotics per production cycle was between 1 and 336 applications (*Own interview data,* 2015) with no difference between Sóc Trăng and Bến Tre.

### 3.2. Pesticide and antibiotic screening

No significant differences in pesticide concentrations were observed between the two provinces for the respective agricultural land use systems ($p > 0.05$). Thus, in the following sections both provinces are discussed together.

In soils under permanent rice production, pesticide concentrations were significantly higher than in soil under alternating rice-shrimp and shrimp production ($p < 0.05$). Detection frequencies and average concentration of all individual fungicides follow the order permanent rice > alternating rice-shrimp > permanent shrimp (Fig. 2). In permanent rice system, fungicides were most frequently detected (26 to 100% detection frequency) and reached concentrations of up to 67.1 μg $kg^{-1}$ (for isoprothiolane; Fig. 2). Similarly, insecticides were detected in significantly higher concentrations in permanent rice systems than in the other agricultural land use systems ($p < 0.05$; Fig. 2). Detection of chlorpyrifos was limited to permanent rice systems, while fenobucarb was detected with concentrations of up to 2.3 μg $kg^{-1}$ in alternating rice-shrimp systems (Fig. 2). The maximum pesticide concentration in the alternating rice-shrimp system was 18.0 μg propiconazole/$kg^{-1}$ soil of the rice platform (Fig. 1, SI). The herbicide concentration in soil did not differ between the three agricultural land use systems (Fig. 2). Fenoxaprop-p-ethyl was the only pesticide, which was detected in ditches of the alternating rice-shrimp system and of the permanent shrimp system but not in permanent rice fields (Fig. 2). No insecticide was detected in permanent shrimp systems. The only detected fungicide in soil of shrimp ponds was isoprothiolane, with a concentration range of 0.3 to 3.7 μg $kg^{-1}$ (Fig. 1, SI).

In contrast to pesticide concentrations, significant differences were found in antibiotic concentrations of the two provinces. In Sóc Trăng, the total antibiotic load were significantly higher than in Bến Tre ($p < 0.05$). Concentrations of fluoroquinolone residue levels were significantly higher than sulfonamide and trimethoprim residue levels in the permanent rice and alternating rice-shrimp systems ($p < 0.05$), in Sóc Trăng. In Bến Tre, no significant differences between the concentrations of fluoroquinolone and sulfonamide/diaminopyrimidine were detected among all three land use systems ($p > 0.05$).

In both provinces soil under permanent shrimp production exhibited significantly higher antibiotic concentrations than soil under permanent rice and the platform of alternating rice-shrimp systems ($p < 0.05$; Table 4). Detection frequency and concentration of the target antibiotics did not differ between both provinces in the permanent shrimp system. Apart from sulfamethoxazole, the other target sulfonamides and fluoroquinolone were detected in 50% of the sampled shrimp ponds (Table 4).

In contrast, in the alternating rice-shrimp system, the ditch as well as the platform contained sulfonamide less frequently, but

[redacted]

| [redacted] | | | | | | Alternating rice-shrimp (n = 20) | Permanent shrimp (n = 20) |
|---|---|---|---|---|---|---|---|
| *I) Management aspects* | | | | | | | |
| Time of establishment of the current system (average) | (years) | > 20 | 20 | 15 | > 20 | 20 | 2 |
| Feed of shrimps | (%) | | | | | | |
| a) Natural food | | – | 0 | 0 | – | 75 | 0 |
| b) Natural food + supplement | | – | 30 | 0 | – | 5 | 0 |
| c) Industrial food | | – | 70 | 100 | – | 20 | 100 |
| Cropping system for rice cultivation (per year): | (%) | | | | | | |
| a) Single crop | | 0 | 100 | – | 0 | 100 | – |
| b) Double crop | | 100 | 0 | – | 25 | 0 | – |
| c) Triple crop | | 0 | 0 | – | 75 | 0 | – |
| *II) Pesticide management* | | | | | | | |
| Use of pesticides | (%) | 100 | 90 | 0 | 90 | 10 | 0 |
| Application frequency per season (average) | | 5 | 2 | 0 | 4 | 2 | 0 |
| Distribution of used pesticides | | | | | | | |
| Herbicide | (%) | 13 | 0 | – | 21 | 17 | – |
| Fungicide | | 36 | 75 | – | 32 | 63 | – |
| Insecticide | | 35 | 15 | – | 32 | 13 | – |
| Molluscicides[b] | | 16 | 0 | – | 15 | 7 | – |
| Pre-treatment of seeds | (%) | 90 | 35 | – | 100 | 90 | – |
| *III) Antibiotic management* | | | | | | | |
| Farmers who apply antibiotics | (%) | – | 0 | 90 | – | 5 | 90 |
| Application frequency per season[a] (median) | | / | 0 | 28 | / | 3 | 28 |

[a] Application amount is calculated as the total treatment dose of antibiotics in one production cycle, regardless the individual application pattern.
[b] Molluscicides were not pursued further for the pesticide screening as the analytical method is not suitable for the most commonly used molluscicides.

fluoroquinolone was detected in up to 90% of the samples (Table 4). In this system, all target antibiotics were more frequently detected in Sóc Trăng than in Bến Tre (Table 4). In Bến Tre ciprofloxacin was less frequent detected with more than seven-fold lower concentrations (Table 4).

In the permanent rice system, only enrofloxacin, trimethoprim, and sulfamethazine were detected in both provinces (Table 4). Among all land use systems, the highest antibiotic concentration was found for sulfamethazine (86 µg $kg^{-1}$) in the province Bến Tre.

## 4. Discussion

### *4.1. Management and use of pesticides and antibiotics in the three different land use systems of the coastal Mekong Delta*

The data of the household interviews confirmed the intensive usage of pesticides in the permanent rice systems. The average application frequency of pesticides in permanent rice systems was within the reported range for other provinces (An Giang and Can Tho) in the Mekong Delta (Chau et al., 2015a), ranging between 4 and 8 applications per cropping season. Fungicides and insecticides are frequently used to

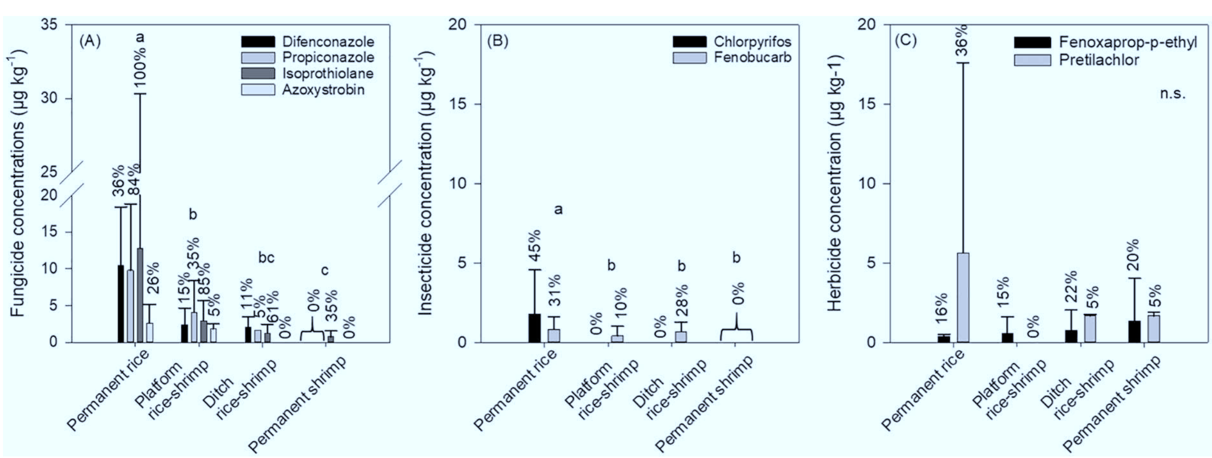


**Fig. 2.** Mean (A) fungicide, (B) insecticide, and (C) herbicide residue concentrations in soil (dry matter) of the different land use systems (permanent rice n = 19; alternating rice-shrimp platform n = 20; alternating rice-shrimp ditch n = 18; permanent shrimp n = 20). Detection frequencies are given in percentage above the respective bars. For mathematical calculations non-detected pesticides were calculated as routine limited of quantification ∗ 0.5 (RLOQ ∗ 0.5), non-quantified pesticides were calculated as RLOQ. Significances ($p < 0.05$) are marked with small letters above the bars – different letters indicate significant differences between systems, and n.s. implies no statistically significant differences.

**Table 4**
Range of antibiotic concentration in the three land use systems and per province in $\mu g\,kg^{-1}$ (dry matter). Detection frequencies (in %) are given in brackets for the particular antibiotic in the respec[redacted] mathematical calculations non-detected (n.d.) antibiotics were calculated as routine limit of quantification $*$ 0.5 (RLOQ $*$ 0.5), non-quantified antibiotics were calculated as RLOQ.

| Antibiotic | Permanent rice | | | Alternating rice-shrimp | | | | | | Permanent shrimp | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | Platform | | | Ditch | | | | |
| | Total (n = 20) | Sóc Trăng (n = 10) | Bến Tre (n = 10) | Total (n = 20) | Sóc Trăng (n = 10) | Bến Tre (n = 10) | Total (n = 20) | Sóc Trăng (n = 10) | Bến Tre (n = 10) | Total (n = 20) | Sóc Trăng (n = 10) |
| *Sulfonamides* | | | | | | | | | | | |
| Sulfadiazine | n.d. (0) | n.d. (0) | n.d. (0) | n.d. (0) | n.d. (0) | n.d. (0) | n.d.–0.1 (15) | n.d. (0) | n.d.–0.1 (30) | n.d.–0.3 (50) | n.d.– < 0.1 (40[redacted] |
| Sulfamethazine | n.d.–86.1 (10) | n.d. (0) | n.d.–86.1 (20) | n.d.–8.2 (10) | n.d.–8.2 (20) | n.d. (0) | n.d.–2.3 (10) | n.d.–2.3 (20) | n.d. (0) | n.d.–5.3 (60) | n.d.–3.3 (70) |
| Sulfamethoxazole | n.d. (0) | n.d. (0) | n.d. (0) | n.d.–2.9 (5) | n.d.–2.9 (10) | n.d. (0) | n.d. (0) | n.d. (0) | n.d. (0) | n.d.–0.8 (5) | n.d. (0) |
| *Diaminopyrimidines* | | | | | | | | | | | |
| Trimethoprim | n.d.–0.6 (15) | n.d. (0) | n.d.–0.6 (30) | n.d.–0.4 (50) | n.d.–0.4 (80) | n.d.–2.9 (20) | n.d.–0.2 (50) | n.d.–0.2 (70) | n.d.–0.18 (30) | n.d.–0.6 (50) | n.d.–0.3 (60) |
| *Fluoroquinolone* | | | | | | | | | | | |
| Enrofloxacin | n.d.–2.9 (50) | n.d.–2.1 (40) | n.d.–2.9 (70) | n.d.–4.3 (95) | 0.4–4.3 (100) | n.d.–0.5 (90) | n.d.–5.8 (85) | 0.3–5.8 (100) | n.d.–3.5 (70) | n.d.–1.0 (70) | n.d.–0.8 (80) |
| Cirprofloxacin | n.d. (0) | n.d. (0) | n.d. (0) | n.d.–12.0 (45) | n.d.–12.0 (90) | n.d. (0) | n.d.–7.4 (25) | n.d.–7.4 (40) | n.d.–1.0 (10) | n.d.–1.6 (40) | n.d.–1.0 (40) |

[redacted] pesticides carry risks for their saline aqua[redacted] production. Nevertheless, some pesticides were applied in alternating rice-shrimp farming systems, either because they were still needed for maintaining the rice growth or as chemical pre-treatment of the rice seeds (pickling).

In both provinces, application of antibiotics was a standard management practice in permanent shrimp cultivation. Antibiotics were often applied as a preventive measure to avoid diseases such as caused by *Vibrio* bacteria (*Own interview data*, 2015; Holmström et al., 2003). The significantly lower use of antibiotics in alternating rice-shrimp system is linked to an extensive shrimp production system (lower shrimp density) and lower risk of disease and pest outbreaks (Lu and Li, 2006). Nevertheless, because of hydrological interlinked channel systems, a transfer of these antibiotics into paddy rice fields cannot be excluded, as evidenced by our screening data (Table 4).

### *4.2. Pollution pattern in the three different land use systems in the coastal Mekong Delta*

In permanent rice systems, the frequent detection and high concentration of isoprothiolane were in agreement with our result from farmer's interview, which indicated that they sprayed mostly fungicides and insecticides. Earlier studies have already reported on the frequent detection of isoprothiolane in soil of arable land (Toan et al., 2013), but also in surface waters of the surrounding environment (Chau et al., 2015a) of the Mekong Delta. The omnipresence of isoprothiolane in all land use systems might be partly due to both its high recommended application dosage of $480\,g\,ha^{-1}$ as well as its frequent use. Isoprothiolane is used against rice-blast – one of the most important diseases in rice-cropping (Kihoro et al., 2013). Additionally, this active ingredient also persists in soil with the highest half-life ($DT_{50}$) value of 320 days of all target pesticides (Table 1). However, to the best of our knowledge, there is currently no further data available on the concentrations of recently used pesticides in soil under paddy rice production for the coastal Mekong Delta. The limited number of studies on pesticides residues in the Mekong Delta were rather focused on persistent pesticides like organochlorines (Carvalho et al., 2008; Minh et al., 2007) or on pesticides in surface water samples (Chau et al., 2015a) in the Mekong Delta.

In alternating rice-shrimp systems, the frequent detection of fungicides differs with the results from earlier studies but also with our results from the interviews, which suggested that farmers are aware of the negative effects of pesticide application on shrimp production in the subsequent season (Berg, 2001). Despite farmers' awareness, pesticides are still used during rice cultivation, although the application rate was 50% lower per season than in permanent rice (Table 3) and used pesticides were less toxic for the aquatic organisms (Berg and Tam, 2012; Stadlinger et al., 2018). Such a strategy is likely reflected in the observed pollution pattern of chlorpyrifos and fenobucarb. Both pesticides can be applied against the same pests, e.g., leaf rollers or steam bores (PPDB (Pesticide properties database)). However, the critical lethal concentration at which chlorpyrifos may kill 50% of the fish population ($LC_{50}$) is 1.3 ppb, which is much lower than that for fenobucarb at 1700 ppb for fish $LC_{50}$ (Table S3; Pucher et al., 2014). Our data clearly show elevated concentrations of chlorpyrifos in permanent rice, while no chlorpyrifos residues were found in the alternating rice-shrimp systems. In turn, fenobucarb was much more abundant in alternating rice-shrimp systems (Fig. 2). Earlier studies detected also fenobucarb in high frequency but with rather low concentration in arable soil of the Mekong Delta (Toan et al., 2013). Fenobucarb is a popular insecticide due to its low price, high efficiency, and product promotion (Berg and Tam, 2012), which might favour the use of this active ingredient by [redacted] but also other crops like vegetables (Toan et al., 2013; Chau et al., 2015a; *Own interview data*, 2015). However, some shrimp farmers use the pond banks for the cultivation of watermelons and confirmed the use of pesticides (*Own interview data*, 2015). Therefore, it is likely that this together with cross-contamination via irrigation water from interconnected channel system can favour the accumulation of pesticides also in shrimp ponds soils.

Fenoxaprop-P-ethyl was the only pesticide, which sometimes occurred at even higher frequency and concentration in soil of shrimp ponds than in that of permanent rice fields (Fig. 2). The fate of this herbicide in the environment is among other things influenced by the high log $K_{ow}$ value of 4.60, which promotes adsorption to the soil matrix (Table 2). Even though the half-life value of this pesticide is rather low (0.4 days, Table 1), due to the high potential to soil accumulation, the compound is protected from further degradation processes. Pesticides with this high soil sorption capacity can be transported with suspended sediments or dissolved organic matter from the field of application to surrounding areas (Whitford et al., 2010). A related study in the Red River Delta also suggested that log $K_{ow}$ values (log $K_{ow}$ > 4) control spatial distribution of pesticides in surface soil after dyke construction (Braun et al., 2018).

The major users of antibiotics among the three agricultural land use systems are permanent intensive shrimp farmers (Thuy et al., 2011), as also confirmed by our interviews. Nevertheless, detected antibiotic concentrations in the soil did not differ between the permanent shrimp farms and the ditches of the alternating rice-shrimp systems. Besides this, it should be noted that in the province of Sóc Trăng, the production of shrimp has started > 10 years earlier than in Bến Tre (Table 3). Antibiotic concentrations were elevated for the fluoroquinolones in Sóc Trăng, which are known to persist in the environment (e.g., Golet et al., 2002; Rosendahl et al., 2011). However, concentrations of other antibiotics such as sulfonamides were not elevated in Sóc Trăng (Table 4), suggesting that either there was no long-term accumulation of these antibiotics in the soil, and/or that recent misuse and overdosing was avoided more efficiently by the more experienced farmers in Sóc Trăng than in Bến Tre. The observed significant differences between antibiotic classes are likely due to the difference in their physico-chemical properties. For instance, fluoroquinolones are more persistent in soil than sulfonamides and trimethoprim, due to their higher log $K_{ow}$ values, higher soil-water partitioning coefficients and elevated pKa values (Table 2) (Thiele-Bruhn, 2003; Rosendahl et al., 2011; Rosendahl et al., 2012).

In permanent shrimp systems, the frequent detection of sulfonamides and trimethoprim was likely facilitated by their frequent and easy availability for farmers in local shops. Normally, veterinary antibiotics are mixed in commercial products. For instance, popular products were "Trimesul" and "Cotrim" consisting of either sulfamethazine or sulfamethoxazole mixed with trimethoprim (Thuy et al., 2011; Chau et al., 2015b). The low detection frequency of sulfamethoxazole might be due to the fast degradation of this antibiotic in soil (Dalkmann et al., 2012). Our observed concentration ranges for trimethoprim in the permanent shrimp soils are in line with previous findings of Le and Munekage (2004) with 0.01 to 0.73 μg kg$^{-1}$ in soil of shrimp pond bottom.

In our survey, farmers of alternating rice-shrimp systems refused the use of antibiotics in their low input system (Table 3), which is in line with previous studies reporting on extensive farming management (Lu and Li, 2006; Berg, 2001; Stadlinger et al., 2018). Likewise, the use of antibiotics in permanent rice systems is rather unlikely. The maximum value of sulfamethazine with 86.6 μg kg$^{-1}$ in permanent rice can be

[redacted] and enrofloxacin up to 12 ng L$^{-1}$ in surface water in the coastal Mekong Delta (Chau et al., 2015b). Shimizu et al. (2013) detected sulfamethazine in concentration ranging from 5 to 21 ng L$^{-1}$, sulfamethoxazole concentration ranging from 25 to 313 ng L$^{-1}$, and trimethoprim concentration ranging from 7 to 33 ng L$^{-1}$ in channel water in the Mekong Delta. Managaki et al. (2007) detected sulfamethoxazole, sulfamethazine and trimethoprim at concentrations between 7 and 328 ng L$^{-1}$ in the Mekong river water. All these data point to low (i.e., below known ecotoxicological thresholds) but ubiquitous occurrences of antibiotics in the delta.

Possible sources of antibiotics are urban wastewater (Dalkmann et al., 2012; Xu et al., 2009), livestock wastewater (Shimizu et al., 2013) or discharge of water from aquaculture production in general (Chau et al., 2015b) and shrimp production in particular at given site (Thuy et al., 2011). The facilitated exchange of water in the Mekong Delta, also enable the interchange of eroded sediment, for instance from shrimp ponds. Transport of antibiotics, especially for fluoroquinolone, could be via suspended solids, like eroded sediment or dissolved organic matter (Huang et al., 2011). Our observed pattern of detected antibiotics in the permanent rice and alternating rice-shrimp fields were similar to those in the permanent shrimp systems and followed, in general, the order permanent shrimp < integrated rice-shrimp < permanent rice (Table 4). This suggests that changing land-use patterns thus contribute to overall antibiotic dispersal. We thus have to add discharges from arable systems to already described main antibiotic sources in the Mekong River, apart from sewage, as outlined by Shimizu et al. (Shimizu et al., 2013).

The higher occurrence of fluoroquinolones in the alternating rice-shrimp system when compared to the permanent shrimp system could be linked to differences in production management. Alternating rice-shrimp farmers remove the sludge less frequently than permanent shrimp farmers. The sludge is either removed on the surrounding banks of the ponds, used as "manure" in the domestic vegetable production or directly pumped into the adjacent irrigation ditches (*Own interview data*, 2015). The sludge layer can act as sink for antibiotics, especially for fluoroquinolone, which can persist for months to years (Rosendahl et al., 2012; Du and Liu, 2012).

### *4.3. Risk assessment and changes in pollution patterns*

The majority of the frequently detected pesticides (e.g., isoprothiolane, propiconazole) in this study were classified as moderately hazardous (Class II) after the WHO toxicity classes (WHO, 2010). The soil samples of all land use systems also exhibited pesticide concentrations below respective critical concentration values for soil and sediment dwelling organisms (critical values listed in Table S3). This does not necessarily mean that there was no overuse of pesticides, as Chau et al. (2015a) reported that only a few farmers typically followed the recommended dose applications. However, it certainly reflects elevated dissipation rates as found for other tropical climates (Laabs et al., 2002; Chai et al., 2013) and that the monsoon rainfall remobilises and washes out the pesticides from soils and sediments (Toan et al., 2013).

Even though the data in this study do not provide evidence of direct use of pesticides in permanent shrimp systems, it is possible that the ongoing increasing change in land use towards shrimp aquaculture can also indirectly influence pesticide usage in the remaining paddy rice field's use. The reduced net area for paddy rice production likely increases the pressure on the remaining fields to produce high yields to ensure food security (Sebesvari et al., 2011; Laabs et al., 2002). In contrast, and compared to permanent rice and permanent shrimp systems, alternating rice-shrimp farming will remain as a low external quotient (HQ) between the measured environmental concentration (MEC) and the predicted no effect concentration (PNEC) values were calculated (Table S4). No antibiotic concentration poses a hazard for the soil organisms according to known threshold values (Table S4), therefore supporting earlier monitoring data of antibiotics in other regions of the Mekong Delta (Chau et al., 2015a).

Inasmuch as farmers might increasingly combine shrimp and rice or even switch to permanent shrimp production as an adaptation strategy to increased salinity intrusion, the use of antibiotics will increase significantly. At first glance, there may therefore be elevated risks for a long-term selection of antibiotic-resistant bacteria. Several studies already observed antibiotic-resistant bacteria in the environment of shrimp farms in Vietnam and the Philippines (Le et al., 2005; Tendencia and La Peña, 2001). Yet, permanent shrimp farming must not necessarily be performed in open ponds established on former paddy fields, but can also be done at fully engineered level in artificial pond systems with automatic control of shrimp growth and disease prevention, i.e., also automated aeration, fertilization, and antibiotic use, if needed. Such systems already exist in Vietnam. They require large investments, but provide also the opportunity to clean the water and control the quality of discharge water into the environment.

## 5. Conclusion

This study provides the first background data on the presence of currently used pesticides and antibiotics in soils of different agricultural land use systems in two coastal provinces of the Mekong Delta. The concentrations of pesticide residues were significantly higher in permanent rice systems than in alternating rice-shrimp and permanent shrimp systems. Antibiotic residues showed elevated concentrations in the permanent shrimp system compared to the alternating rice-shrimp and the permanent rice systems. As the alternating rice-shrimp systems are most sensitive to pollutant transfers among different land-use compartments, and since farmers are aware of this, any change in land use patterns which fosters alternating rice-shrimp farming will likely result in reduced pollutant loads of the delta, which is to be considered in future land use planning for the coastal area of the Mekong Delta that will be facing increased salinity intrusion in the future.


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## Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.envint.2019.03.038.

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