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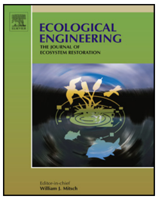

Water residence time and pesticide removal in pilot-scale wetlands

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ABSTRACT

The use of agricultural pesticides may lead to environmental contamination. Constructed wetlands (CWs) are often recommended for decreasing non-point source pollution by pesticides. The aim of this study was to evaluate the influences of water residence time (WRT) on the effectiveness of two CWs for decreasing pollution in drainage water at the pilot scale. Thus, two successive charge/discharge steps were conducted successively with high and low flows before conducting a step with no flow. For both pilot experiments, the measured effectiveness was between 22 and 100% of the applied quantities of the five pesticides. These results were correlated with the physicochemical properties of the pesticides, particularly their adsorption and desorption capacities. In addition, the pilot scale wetlands were less effective during short WRT (from 15.2 to 100%) than during long WRT (from 17.8 to 100%), despite the greater remobilisation observed during the discharge steps with low flows. After stopping flow, the DT_{50} values of the pesticides in the aqueous phase were less than 4 days in the two pilot wetlands, likely due to their rapid transfer to the solid phase (soil or straw). In addition, the DT_{50} values measured in the solid phases ranged from 3.8 days to more than one year. Thus, the WRT must be optimised in the CW by maintaining vegetation or installing weirs, to favour retention processes (sorption, sedimentation) during the drainage periods (October to May). Next, the pesticides stored in the solid phase could be subjected to degradation processes, particularly when moisture, residence times and temperature conditions are favourable (summer).

1. Introduction

Pesticides applied in agriculture may alter water quality through several transfer pathways such as runoff/erosion, spray drift, leaching or drain flow (Reichenberger et al., 2007). Several strategies are proposed to reduce pesticide pollution, including best management practices such as using grass cover bands and maintaining untreated areas. However, the drainage water can bypass these mitigation strategies. Constructed wetlands (CWs) are proposed to reduce the pesticide pollution of runoff or drainage water, and their effectiveness has been demonstrated in several studies (Elsaesser et al., 2011; Moore et al., 2008; Rose et al., 2006; Schulz and Peall, 2001). In Lorraine, approximately 20% of agricultural lands are drained (Recensement Agricole, 2010) which contributes to environmental contamination. Two studies have reported peak pesticide concentrations in drainage water of 5.2–395.3 $\mu g\,L^{-1}$ (Dousset et al., 2004; Novak et al., 2001). These concentrations exceed the limits (0.1 $\mu g\,L^{-1}$) given by the European water framework directive (JOCE, 2000).

Rural areas require low-cost autonomous treatment systems to reduce pesticide concentrations in drainage water before reaching ditches and rivers. Thus, nine experimental CWs were installed in grass buffer strips in Lorraine, France, and nearly 70 frequently applied pesticides were monitored at the inlets and outlets of the CWs. At the field scale, pesticide concentrations were reduced between the inlets and outlets of the CWs. However, a wide range of behaviours were observed regarding the different pesticide molecules. Several studies reported that the removal rates of pesticides or their degradation products ranged from a few percent to 100% (Budd et al., 2009; Maillard et al., 2011; Rose et al., 2006; Tournebize et al., 2013). However, Passeport et al. (2013) presented negative removal rates (−32%) which likely resulted from the remobilisation processes. Many parameters have been proposed to explain the wide range of efficiency observed in CWs: water residence time, vegetation, retention processes, degradation processes and the physico-chemical properties of pesticides such as solubility, K_{ow} or DT_{50} (Vymazal and Březinová, 2015).

The water velocity, type of pesticide and water residence times (WRT) are often important parameters influencing the effectiveness of CW (Blankenberg et al., 2006; Passeport et al., 2010; Rodgers and Dunn, 1992). Other parameters, such as plant density, wetland dimensions and flow rate, influence the WRT. In Lorraine, nine

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Table 1
Main properties of the studied pesticides (University of Hertfordshire, 2013).

	2,4-MCPA	Isoproturon	Napropamide	Boscalid	Prochloraz	Tebuconazole
Water solubility at 20 °C ($mg\,L^{-1}$)	29,400	70.2	74.0	4.6	26.5	36.0
Vapour pressure at 25 °C (mPa)	4.00×10^{-1}	5.50×10^{-3}	2.20×10^{-2}	7.20×10^{-4}	1.50×10^{-4}	1.30×10^{-3}
Sorption coefficient K_{oc} ($L\,kg^{-1}$)	n.d.	122 (36–241)	839 (435–1609)	809 (750–1200)	500 (n.d.)	769 (102–1249)
Half-live at 20 °C (days)	24 (7–41)	12 (7.2–18.2)	308 (120–400)	246 (27–372)	223.6 (22.1–936.1)	365 (n.d.)

n.d.: not available data.

CWs were installed within a grass cover band to reduce the pesticide concentration from drainage water. During the drainage period (i.e., from October to May), a succession of low (<0.5 $L\,s^{-1}$) and high (>5 $L\,s^{-1}$) flow rates into these CWs were measured. Additionally, the pesticide concentrations measured in the inlet and outlet water samples ranged from a few $ng\,L^{-1}$ to more than 25 $\mu g\,L^{-1}$ depending on the pesticide molecules (ANSES, pers.com). At the end of each drainage period, water can become stagnant in CWs that favour longer residence times. Thus, the objectives of this study were (i) to assess the impacts of WRT on the effectiveness of pilot scale wetlands for reducing the concentrations of dissolved pesticides during the drainage period and (ii) to determine the fate of pesticides accumulated in wetlands during the water stagnation period between drainage periods.

2. Materials and methods

2.1. Experimental sites

Among the nine CWs installed in Lorraine, two were monitored more frequently at the field scale. In addition, pesticide sorption and dissipation were realised under laboratory conditions (Vallée et al., 2014, 2015). The CW at Jallaucourt (Moselle, France) consisted of a 13 long, 6 m wide ditch with a water level of 1 m and a water storage capacity of 4 m^3. To reduce the rate of water flow and lengthen the residence time, a bundle of straw was placed in the middle of the ditch. Spontaneous vegetation (dominated by *Glyceria fluitans*, *Ranunculus Repens*, *Juncus conglomeratus* and *Juncus effusus*) was developed for total recovery after 3 years. The CW at Ollainville (Vosges, Lorraine, France) consisted of a triangular pond (20.5 m × 15.5 m × 11 m) with a water level of 0.6–0.8 m and a water storage capacity of 100 m^3. Spontaneous vegetation (dominated by *Typha latifoli*, *Ranunculus Repens*, *Callitriche platycarpa* and *Juncus conglomerates*) was developed, and the entire CW was recovered after one year.

2.2. Selected pesticides

Among the 70 pesticides followed in the CWs, six were chosen for this study. The applied amounts, the concentrations in the drainage water, and the diversities of their physico-chemical properties were the main criteria used to choose the pesticides. Three fungicides: boscalid (2-chloro-N-(4′-chlorobiphenyl-2-yl)nicotinamide) (BCL), prochloraz (N-propyl-N-[2-(2,4,6-trichlorophenoxy)ethyl]imidazole-1-carboxamide) (PCZ) and tebuconazole (1-p-chlorophenyl-4,4-dimethyl-3-(1H-1,2,4-triazol-1-ylmethyl)pentan-3-ol) (TBZ) and three herbicides: 2,4-MCPA (2-methyl-4chlorophenoxyacetic acid) (MCPA), isoproturon (3-(4-isopropylphenyl)-1,1-dimethylurea) (IPU) and napropamide (N,N-diethyl-2-(1-naphthyloxy)propionamide) (NPP) were selected. Analytical standards (>98%) were purchased from Sigma–Aldrich (Seelze, Germany), and the main characteristics of these pesticides are given in Table 1.

2.3. Pilot set-up

The Jallaucourt and Ollainville pilots were built of polyvinyl chloride at scales of 1:10 and 1:20 (Fig. 1), respectively. The soils, straw and representative vegetation were sampled on 08 April 2013. The soils and straw were air-dried, and the soils were passed through a 2 mm sieve and homogenised. The main characteristics of the soils and straw are provided in Table 2. The plants were transplanted in pots and grown in a phytotron over 8 months. For the Jallaucourt pilot, 22 kg of soil was deposited at a depth of 5–10 cm

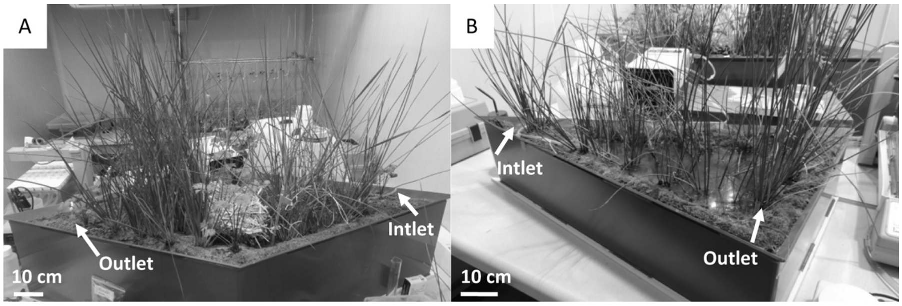

Fig. 1. Experimental pilots of Jallaucourt (A) and Ollainville (B) in the phytotronic room after 75 days of experiment.

Table 2
Main characteristics of the pilot substrates.

Substrate	Clay ($g\,kg^{-1}$)	Silt ($g\,kg^{-1}$)	Sand ($g\,kg^{-1}$)	pH (H_2O)	Organic carbon ($g\,kg^{-1}$)	Total nitrogen ($g\,kg^{-1}$)	Total calcareous ($g\,kg^{-1}$)	CEC[a] ($cmol+\,kg^{-1}$)
Jallaucourt soil	504	376	120	7.84	16.5	1.45	2.5	33.4
Jallaucourt straw	–	–	–	–	359.0	12.2	–	–
Ollainville soil	604	343	53	6.93	18.6	1.86	1.4	31.5

[a] CEC cationic exchange capacity.