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Role of submerged vegetation in the retention processes of three plant protection products in flow-through stream mesocosms

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HIGHLIGHTS

- Aquatic macrophytes determine how dispersion and sorption mitigate PPPs in streams.
- Sparse vegetation fosters dispersion.
- Dense vegetation fosters mass retention.
- Compound related and time limited mass retention compensates diminished dispersion.

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ABSTRACT

Quantitative information on the processes leading to the retention of plant protection products (PPPs) in surface waters is not available, particularly for flow-through systems. The influence of aquatic vegetation on the hydraulic- and sorption-mediated mitigation processes of three PPPs (triflumuron, pencycuron, and penflufen; $\log K_{OW}$ 3.3–4.9) in 45-m slow-flowing stream mesocosms was investigated. Peak reductions were 35–38% in an unvegetated stream mesocosm, 60–62% in a sparsely vegetated stream mesocosm (13% coverage with *Elodea nuttallii*), and in a similar range of 57–69% in a densely vegetated stream mesocosm (100% coverage). Between 89% and 93% of the measured total peak reductions in the sparsely vegetated stream can be explained by an increase of vegetation-induced dispersion (estimated with the one-dimensional solute transport model OTIS), while 7–11% of the peak reduction can be attributed to sorption processes. However, dispersion contributed only 59–71% of the peak reductions in the densely vegetated stream mesocosm, where 29% to 41% of the total peak reductions can be attributed to sorption processes. In the densely vegetated stream, 8–27% of the applied PPPs, depending on the $\log K_{OW}$ values of the compounds, were temporarily retained by macrophytes. Increasing PPP recoveries in the aqueous phase were accompanied by a decrease of PPP concentrations in macrophytes indicating kinetic desorption over time. This is the first study to provide quantitative data on how the interaction of dispersion and sorption, driven by aquatic macrophytes, influences the mitigation of PPP concentrations in flowing vegetated stream systems.

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1. Introduction

The application of plant protection products (PPPs) is a common practice in conventional agriculture. As a result, PPPs can enter non-target aquatic ecosystems, either through spray-drift during application or edge-of-field runoff and drainage post application (Schulz, 2004). In addition to best management practices (BMPs), such as improved application techniques or vegetated buffer strips, vegetated treatment systems (VTSs) have been proposed and evaluated for the mitigation of PPP concentrations in receiving

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Abbreviations: PPP, plant protection product; BMP, best management practice; VTS, vegetated treatment system; HRT, hydraulic retention time; K_{OW}, octanol–water partitioning coefficient; FOF, fiber-optic fluorometer; SPE, solid phase extraction; ASE, accelerated solvent extraction; dSPE, dispersive solid phase extraction; UHPLC-MS, ultra high performance liquid chromatography-mass spectrometry; DIN, german institute for normalization; OTIS(-P), one-dimensional solute transport model for streams and rivers.

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waters (Reichenberger et al., 2007; Gregoire et al., 2008). A meta-analysis (Stehle et al., 2011) confirmed the effectiveness of VTSs for the mitigation of PPP concentrations and identified the high lipophility of PPPs in combination with the high plant coverage in the VTSs to be the major factors influencing the mitigation performance.

In fact, the ability of aquatic vegetation to interact with PPPs has been demonstrated in studies at the laboratory (Crum et al., 1999; Olette et al., 2008), microcosm (Bouldin et al., 2006), and mesocosm scale (Moore et al., 2009). The majority of mesocosm and field studies linked the sorption of lipophilic PPPs to aquatic macrophytes or, in limited cases, to sediments with the mitigation potential of the investigated VTSs (Bennett et al., 2005; Rogers and Stringfellow, 2009). Margoum et al. (2006) and Passeport et al. (2011) identified the ability of substrates, such as sediment, leaves, plants and soil taken from ditches, wetlands or forest buffers, respectively, to absorb and thus retain PPPs within these systems. Nevertheless, most of the studies published to date were performed in vegetated wetlands with negligible flow velocities (Schulz et al., 2003b; Moore et al., 2007; Gill et al., 2008).

The few studies that investigated the mitigation potential and the fate of the PPPs in flow-through vegetated treatment systems and streams were performed either as case studies under field conditions or in vegetated wetlands with irreproducible system properties. The reductions of azinphos-methyl concentrations (61–90%) were reported from a vegetated tributary of the Lourens River, South Africa, subsequent to a spray drift and runoff event (Dabrowski et al., 2006). Schulz et al. (2003a) revealed an overall reduction in the azinphos-methyl concentrations of 90% in a flow-through wetland in South Africa with an overall mass retention of 61%, of which 10.5% were initially adsorbed to macrophytes. Elsaesser et al. (2011) found merely 5% of the PPPs applied to the Lier wetland, Norway, adsorbed to macrophytes, and thus presumed that the increase of peak reductions in the vegetated wetland cells seemed to be a result of vegetation induced dispersion processes. However, those studies pursued relatively simplistic sampling strategies that relied on the comparison of few concentrations at the inlets and the outlets of the investigated systems and did not focus on a quantitative description of the processes leading to the mitigation of the PPP concentrations.

In addition to being a potential sink for PPPs through sorption processes, aquatic vegetation constitutes a key factor determining the hydraulic conditions in streams and wetlands (Sukhodolov and Sukhodolova, 2012). Depending on the plant density and geometry, increased velocity shear and turbulent mixing lead to enhanced longitudinal dispersion of fluid momentum (Nepf, 2012). The roughness or flow types (Nikora et al., 2007) differ significantly between emergent vegetation, which extends throughout the entire water column, and submerged vegetation, which is always superposed by a free floating water layer (Shucksmith et al., 2011). Within emergent macrophyte canopies, the relevant length scales for turbulent mixing are limited by the stem diameter and spacing. Potentially larger vortices at the canopy-scale are generated in a shear layer between the canopy and the overflow in the non-vegetated part of the water column (Nepf et al., 2007). Hydrodynamic aspects of flow-plant interactions have been studied in detail from the scales of individual stems and leaves (Albayrak et al., 2011) up to patches of vegetation (Sukhodolov and Sukhodolova, 2012; Sukhodolova and Sukhodolov, 2012). Recently, different aspects of the reactive transport of dissolved substances were investigated at the laboratory (Hansen et al., 2010) up to the field (Schuetz et al., 2012) scale. In spite of its potential importance in many engineered as well as natural aquatic ecosystems, studies that conjointly investigated the influence of aquatic vegetation on both the hydraulic and sorption processes, and thus, on the mitigation of PPPs in slow-flowing streams, are not existent so far.

The vegetated flow-through stream mesocosms in which the present study was performed facilitated the modifications of individual system inherent properties while ensuring reproducible experimental conditions. For the present study, the coverage and the spatial distribution of macrophytes were the only system properties that were modified among the stream mesocosms. Based on this experimental setup, this study aimed to quantify the role of longitudinal dispersion and sorption processes as a result of submerged vegetation and its density-related spatial distribution on the peak concentration and mass retention of three moderately lipophilic PPPs ($\log K_{OW}$ 3.3–4.9) at the mesocosm scale.

2. Materials and methods

2.1. General study outline

The study phase lasted from 31st of July 2011 until 12th of August 2011 and consisted of two experimental phases. The tracer experiment was conducted in total darkness during the night from 31st of July 2011 to 1st of August 2011. The PPP experiment occurred with two separate dosing events of the stream mesocosms on the 2nd and 10th of August 2011. Generally, the tracer experiment, as well as both PPP applications were carried out during windless and rainless weather conditions, respectively. However, during the second PPP application in the unvegetated stream mesocosm the occurrence of wind gusts with wind speed over 2 on the Beaufort scale affected the application. In order to avoid any impact of precipitation on the PPP experiment, the stream mesocosms were transiently covered with tarpaulins when rain showers occurred during the entire experimental phase.

2.2. Vegetated stream mesocosms

The study was performed in three out of a total of sixteen independent stream mesocosms of the Landau stream mesocosm facility (Elsaesser et al., 2013) (Fig. 1). Each of the stream mesocosms consisted of U-shaped concrete tubs (length = 45 m; width at the bottom = 0.37 m and width at the top = 0.38 m; depth = 0.5 m) and contained a 0.26-m water layer on top of a sediment layer (0.12 m) of medium loamy sand (total organic carbon = 1.0 ± 0.4%, $n = 15$). Two of the stream mesocosms were planted with different densities of the submerged western waterweed (*Elodea nuttallii*). Immediately after the termination of the PPP experiment, macrophytes were removed from three sites (each 0.2 m^2) in each vegetated stream mesocom, lyophilized and weighed to assess the macrophytes biomass (dry weight) and relative density, respectively (Table S1, Supplemental material). Stream 3 was densely covered by macrophytes, while the biomass in stream 2 was reduced to 13% relative to the densely vegetated stream 3 prior to the start of the experimental phase. Macrophytes in stream 3, however, homogeneously covered the entire cross-sectional area from the bottom boundary to the water surface. Macrophytes in stream 2 protruded only to about half of the water depth and were covered by a free flow zone in the upper part of the cross-sectional area (Fig. 2a–c).

Outside of both experimental phases, the stream mesocosms were run in a circulation mode, where the outflowing water was pumped back to the stream inlets by centrifugal pumps and the evaporation loss was compensated with tap water. However, in contrast to other studies, the stream mesocosms were run in the flow-through mode during the tracer experiment, as well as during the entire experimental phase of the PPP experiment (13 d), with water being fed to the stream mesocosms from a water reservoir at the streams head end and subsequently discharged at the stream outlets. For both the tracer and the PPP experiment, the

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