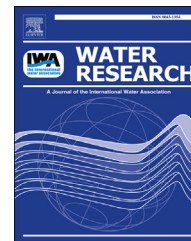


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Phosphorus losses from agricultural land to natural waters are reduced by immobilization in iron-rich sediments of drainage ditches



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ABSTRACT

Redox reactions involving iron (Fe) strongly affect the mobility of phosphorus (P) and its migration from agricultural land to freshwater. We studied the transfer of P from groundwater to open drainage ditches in an area where, due to Fe(II) rich groundwater, the sediments of these ditches contain accumulated Fe oxyhydroxides. The average P concentrations in the groundwater feeding two out of three studied drainage ditches exceeded environmental limits for freshwaters by factors 11 and 16, but after passing through the Fe-rich sediments, the P concentrations in the ditch water were below these limits. In order to identify the processes which govern Fe and P mobility in these systems, we used diffusive equilibration in thin films (DET) to measure the vertical concentration profiles of P and Fe in the sediment pore water and in the ditchwater. The Fe concentrations in the sediment pore water ranged between 10 and 200 mg L⁻¹ and exceeded those in the inflowing groundwater by approximately one order of magnitude, due to reductive dissolution of Fe oxyhydroxides in the sediment. The dissolved P concentrations only marginally increased between groundwater and sediment pore water. In the poorly mixed ditchwater, the dissolved Fe concentrations decreased towards the water surface due to oxidative precipitation of fresh Fe oxyhydroxides, and the P concentrations decreased more sharply than those of Fe. These observations support the view that the dynamics of Fe and P are governed by reduction reactions in the sediment and by oxidation reactions in the ditchwater. In the sediment, reductive dissolution of P-containing Fe oxyhydroxides causes more efficient solubilization of Fe than of P, likely because P is buffered by adsorption on residual Fe oxyhydroxides. Conversely, in the ditchwater, oxidative precipitation causes more efficient immobilization of P than of Fe, due to ferric phosphate formation. The combination of these processes yields a natural and highly efficient sink for P. It is concluded that, in Fe-rich systems, the fate of P at the sediment–water interface is determined by reduction and oxidation of Fe.

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1. Introduction

Excessive fertilization of agricultural land has resulted in large losses of phosphorus (P) from soils to the aquatic environment. This has caused cultural eutrophication of lakes, streams, and estuaries in many developed countries worldwide (Correll, 1999; Schindler, 2012; Smith, 2003). Losses of P from agricultural land may occur either by soil erosion and overland flow or by leaching, i.e. vertical transport through the soil profile. Globally, overland flow is the principal pathway in most catchments (Hansen et al., 2002). Such losses are related to erosion of particulate P during storm events, mostly in fine-textured soils (Sharpley et al., 2001). The transport and environmental effects of eroded P have been studied in great detail (Delgado and Scalenghe, 2008; Ekholm, 1994). Conversely, in well-drained soils with low P sorption capacity, P losses predominantly occur through leaching (Van der Zee, 1988). Such percolates may cause P-enrichment of shallow groundwater and may ultimately reach the aquatic environment via the groundwater flow. The lowland areas of Belgium and the Netherlands have acid sandy soils with a long history of excessive fertilizer application. These soils are P-saturated and exhibit large leaching losses to shallow groundwater (Lookman et al., 1995; Van der Zee et al., 1990). In such soils, it is difficult to balance agronomic needs (optimal crop production) and environmental concerns (limited P leaching) (del Campillo et al., 1999).

The transport of P from percolate or groundwater through drains to the aquatic environment is often assumed to be conservative, i.e. it is not much affected by chemical processes (Schoumans et al., 2013; van der Salm et al., 2011). However, as P-enriched groundwater surfaces, it seeps through micro-oxic and highly redox-active environments, such as the linings of drainage tubes or the sediments of streams and ditches. In such environments, P may undergo complex chemical interactions with several redox-active elements, most notably with Fe. As hydrous ferric oxides are submerged and become anoxic, they may undergo reductive dissolution, which is often microbially mediated (Lovley, 1997). Conversely, in oxic or micro-oxic environments, the reduced and highly mobile Fe(II) may be re-oxidized either chemically (Davison and Seed, 1983) or biotically (Druschel et al., 2008). The oxidized Fe(III) readily precipitates as authigenic hydrous ferric oxides, such as ferrihydrite or lepidocrocite (Baken et al., 2013; Duckworth et al., 2009; Fortin et al., 1993).

Several studies have already addressed the interactions between Fe and P at the interface between anoxic and oxic environments. Reductive dissolution of hydrous ferric oxides generally releases associated P (Liu, 2010), and the release of P from reconstructed wetlands is of environmental concern (Zak and Gelbrecht, 2007; Zak et al., 2010). However, in some cases, the release of P from anoxic sediments is limited, possibly due to formation of vivianite (Heiberg et al., 2012, 2008). The subsequent oxidation of Fe(II) produces hydrous ferric oxides which combine a high specific surface area with a high affinity for oxyanions (Dzombak and Morel, 1990). They may bind P by adsorption or coprecipitation and are very efficient sinks for P. The formation of various ferric phosphate minerals with low molar Fe:P ratios, in some cases even down to unity, have been reported in synthetic solutions (Kaegi

et al., 2010; Mayer and Jarrell, 2000; Voegelin et al., 2010), stream sediments (Hyacinthe and Van Cappellen, 2004), and lakes (Gunnars et al., 2002; Lienemann et al., 1999).

The coupling of reductive dissolution and oxidation of Fe may lead to intense Fe cycling, which has been observed in a variety of environments including sediments, groundwater seeps, and lakes (Blöthe and Roden, 2009; Davison, 1993; Gault et al., 2011; Kleeberg et al., 2013). Diffusive techniques, such as Diffusive Gradients in Thin films (DGT) or Diffusive Equilibration in Thin films (DET), are highly suited for probing the chemical composition of such environments. Some recent methodological studies have already simultaneously determined spatially resolved Fe and P concentrations using diffusive techniques (Cesbron et al., 2014), and a study by Xu et al. (2013) showed a strong relationship between Fe and P concentrations in the pore waters of an incubated sediment. However, few studies have examined Fe–P interactions in systems with combined reduction and oxidation reactions. Based on column experiments, Kjaergaard et al. (2012) speculate that if P and Fe(II) are concomitantly released from submerged soils, the export of P may be attenuated by the downstream re-oxidation of Fe(II) and subsequent binding of P. Zak and Gelbrecht (2004) and Zak et al. (2010) showed that even if high P concentrations are measured in the pore waters of reconstructed wetlands, the risk of P export is limited if molar Fe:P ratios in soils and pore waters do not fall below critical values. Immobilization of P by such processes may be a key process limiting P export from selected environments, but the limited evidence warrants further studies.

This study was set up in order to identify the processes which govern the dynamics of Fe and P at the anoxic-oxic boundary. More specifically, we set out to determine how chemical Fe–P interactions in the sediment of drainage ditches can reduce the dissolved P concentrations from far above to well below environmental limits. The transport of dissolved P from Fe(II)-bearing groundwater to open drainage ditches was monitored in an agricultural area where P losses predominantly occur through leaching (Lookman et al., 1995; Van der Zee, 1988 and references therein). Vertical concentration profiles of dissolved P and Fe at the sediment–water interface of open drainage ditches were measured at 1 cm resolution using the DET (diffusive equilibrium in thin films) technique. Our approach relied on field measurements, since laboratory studies of redox-sensitive systems are inherently prone to artefacts (Brand-Klibanski et al., 2007). We hypothesize that P export from drainage ditches is reduced by binding on freshly produced hydrous ferric oxides, and that P is scavenged in the oxic zone as long as the Fe:P ratio in the source (groundwater or sediment) exceeds a threshold value.

2. Materials and methods

2.1. Study sites

The study sites are located in the Kleine Nete catchment (Northern Belgium) which is characterized by acid sandy soils with iron-rich groundwater. The geography, geology, and hydrology of this catchment have been described in detail elsewhere (Vanlierde, 2013; Vanlierde et al., 2007). The land use in

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