Science of the Total Environment 550 (2016) 60–68

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Influence of colloids on the attenuation and transport of phosphorus in alluvial gravel aquifer and vadose zone media

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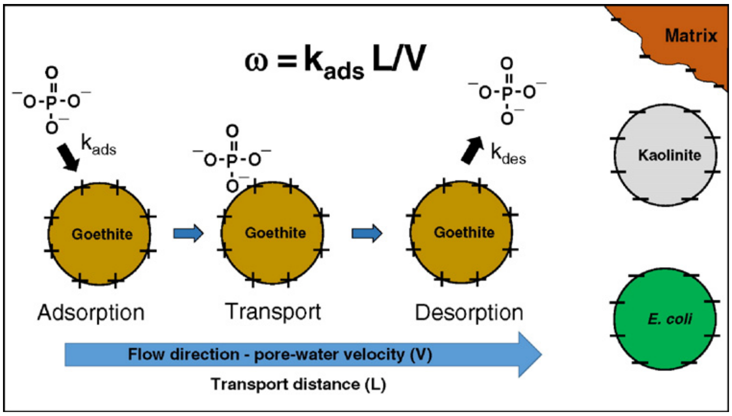
HIGHLIGHTS

- Without model colloids, P readily traveled through permeable gravel aquifer media.
- 4.2 log reduction in P concentration would need 53 m distance in a gravel aquifer.
- In the presence of kaolinite, goethite and *E. coli*, P attenuation was enhanced.
- Fine materials, goethite and low flow rate promoted colloid-assisted P attenuation.
- Colloid-assisted P attenuation could be utilized for prevention of P leaching.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 13 October 2015
Received in revised form 12 January 2016
Accepted 13 January 2016
Available online 22 January 2016

Editor: A. Sarmah

Keywords:
Phosphorus
Colloids
Groundwater
Alluvial gravel
Adsorption
Transport

ABSTRACT

Phosphorous (P) leaching (e.g., from effluents, fertilizers) and transport in highly permeable subsurface media can be an important pathway that contributes to eutrophication of receiving surface waters as groundwater recharges the base-flow of surface waters. Here we investigated attenuation and transport of orthophosphate-P in gravel aquifer and vadose zone media in the presence and absence of model colloids (*Escherichia coli*, kaolinite, goethite). Experiments were conducted using repacked aquifer media in a large column (2 m long, 0.19 m in diameter) and intact cores (0.4 m long, 0.24 m in diameter) of vadose zone media under typical field flow rates. In the absence of the model colloids, P was readily traveled through the aquifer media with little attenuation (up to 100% recovery) and retardation, and P adsorption was highly reversible. Conversely, addition of the model colloids generally resulted in reduced P concentration and mass recovery (down to 28% recovery), and increased retardation and adsorption irreversibility in both aquifer and vadose zone media. The degree of colloid-assisted P attenuation was most significant in the presence of fine material and Fe-containing colloids at low flow rate but was least significant in the presence of coarse gravels and *E. coli* at high flow rate. Based on the experimental results, setback distances of 49–53 m were estimated to allow a reduction of P concentrations in groundwater to acceptable levels in the receiving water. These estimates were consistent with field observations in the same aquifer media. Colloid-assisted P attenuation can be utilized to develop mitigation strategies to better manage

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http://dx.doi.org/10.1016/j.scitotenv.2016.01.075
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effluent applications in gravelly soils. To efficiently retain P within soil matrix and reduce P leaching to groundwater, it is recommended to select soils that are rich in iron oxides, to periodically disturb soil preferential flow paths by tillage, and to apply a low irrigation rate.

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1. Introduction

Phosphorus (P) is an essential nutrient for plant growth. Additions of P to P-limited surface waters will promote growth of aquatic vegetation and algal blooms. Eutrophication can deplete the level of oxygen available to aquatic life, stimulate the growth of toxic cyanobacteria, and decrease the recreational value of rivers and lakes. Concentrations of dissolved P as low as 10 μg/L can cause eutrophic and hyper-eutrophic conditions (Sharpley et al., 2000). The Australian and New Zealand Environment and Conservation Council (2000) guidelines suggest that the trigger values for New Zealand river systems are dissolved reactive phosphorus (DRP) concentrations of 9 μg/L or total phosphorus (TP) 26 μg/L for upland rivers and DRP 10 μg/L or TP 33 μg/L for lowland rivers.

Many point- and nonpoint-sources of P can contribute to eutrophication of surface water. These include erosion of soils and banks, fertilizers, grazing animals, land disposal of animal, human and industrial effluent and sludge, on-farm livestock burials, and decomposing plants. Overland flow and drains are the main pathways of transporting P directly to surface waters. However P can also leach through permeable subsurface media and/or transport through preferential flow paths into groundwater (Fuchs et al., 2009), which recharges the base-flow of surface waters.

In the past, the pathway of P transport through groundwater discharge to surface water has been overlooked (Holman et al., 2008) under a common assumption that P is strongly adsorbed by soils and assimilated by plants, and thus would not leach to groundwater. However recent studies (Tesoriero et al., 2009; Holman et al., 2010; Domagalski and Johnson, 2011; McDowell et al., 2015) have suggested that P does migrate to groundwater, and that subsurface movement of P can be an important transport mechanism that contributes to eutrophication of receiving surface waters. For example, a survey carried out in the UK and Ireland shows that significant numbers of groundwater bodies have median P concentrations above ecologically significant thresholds for freshwaters (Holman et al., 2010). Thus when P-rich groundwater recharges to surface streams, it will contribute to eutrophication of surface water.

Alluvial gravel subsurface media are particularly vulnerable to leaching of contaminants because they are highly permeable and contain significant preferential flow paths. This allows contaminants to be rapidly transported. In New Zealand, many land-use activities (e.g., intensification of dairy farming over the last three decades) have been developed on alluvial plains where stony soils overlie unconfined alluvial gravel aquifers, e.g. Canterbury, Southland and Hawkes Bay. From analysis of national databases, McDowell et al. (2015) have found that aquifers with gravel and sand lithology showed increasing P concentrations in locations used for dairying, with as little as 10-years data.

Colloids can play a significant role in influencing the attenuation and transport of strongly adsorbing contaminants in subsurface media (Kretzschmar et al., 1999; McCarthy and McKay, 2004). For example, contaminants adsorbed to immobile colloids that deposit in porous media can be retained in small pores, reducing their subsurface transport. On the other hand, contaminants adsorbed to mobile colloids can be transported through large pores, enhancing their transport velocity (Walshe et al., 2010). These processes are expected for P because it has a high affinity for binding to colloids, especially to iron oxides (Zhang, 2008). For example, it has been found that P retention in soils increased with increasing iron oxides, aluminum oxides, and aluminosilicate minerals (Brennan et al., 1994). Leaching of P from soils has also been often associated with mobile colloids through preferential flow paths (Schelde et al., 2006; de Jonge et al., 2004). Many types of colloids exist in subsurface media, such as metal oxides, clay minerals, natural organic matter and biocolloids (viruses, bacteria, and protozoa). Significant variability exists among colloids for their influence on P attenuation and transport due to their different physicochemical properties.

The purpose of this study is to examine the influence of a few common colloids on P attenuation and transport in permeable alluvial gravel aquifer and vadose zone media. These include kaolinite, goethite and *E. coli* as model colloids for clay minerals, metal oxides, and biocolloids, respectively.

2. Materials and methods

2.1. Phosphorus and model colloids

Inorganic soluble phosphorus, in the form of orthophosphate (PO_4^{3-}), was used in this study. Orthophosphate is also referred to as “bioavailable or reactive phosphorus” and is the form required by plants. The orthophosphate used in this study was potassium dihydrogen phosphate (KH_2PO_4) purchased from Sigma-Aldrich. It is readily soluble in water with a water solubility of 222 g/L at 20 °C.

Kaolinite, one of the most common clay minerals, was used as the model clay colloid. It is produced by weathering of feldspar and muscovite (Banfield and Eggleton, 1990) and is present in abundance in most weathered soils and environmental waters. Kaolinite ($Al_2Si_2O_5(OH)_4$) is a 1:1 alumina silicate compromising a tetrahedral silica sheet bonded to an octahedral alumina sheet by shared oxygen atoms (Mitchell and Soga, 2005). It has a cation exchange capacity (CEC) of 17 meq/100 g and density of 2.3 g/cm^3 (Demirbaş et al., 2013). The kaolinite used in this study was sourced from Merck KGaA. It had a BET specific surface area (SSA) of 14.54 (±0.08) m^2/g as determined in this study using N_2 physisorption method with Micromeritics Gemini VI 2385-C at 77 K. This is consistent to the previously reported BET SSA previously reported values of 17.2 m2/g (Demirbaş et al., 2013) or 16.6 m2/g (Wei et al., 2014).

Goethite, one of the most abundant ferric (hydr)oxides in natural environments (Cornell and Schwertmann, 2006), was used as the model colloid for metal oxides. Goethite (α-FeOOH) is an iron oxyhydroxide containing ferric iron and has a density of 4.26 g/cm^3 (Cornell and Schwertmann, 2006). Goethite often forms through the weathering of other iron-rich minerals and is ubiquitous in soils. The goethite used in this study was purchased from Sigma-Aldrich. Its BET specific surface area (SSA value) determined in this study was 71.93 (±1.97) m^2/g, which is somewhat higher than similar to the value of 51.2 m^2/g reported by Wei et al. (2014).

Escherichia coli was used as the model bio-colloid as it is the most commonly used bacteria indicator. *E. coli* has a buoyant density of 1.08–1.10 g/mL (Kubitschek et al., 1983). The strain used in this study was *E. coli* J6-2 (Sinton, 1980), which is a non-pathogenic, lactose negative, nalidixic acid-resistant derivative of *E. coli* K-12. It is rod-shaped, 0.76 μm in diameter and 1.42 μm long (Pang et al., 2005). Cells were cultured in Brain Heart Infusion Broth (BBL, Sparks, MD, USA) at 37 °C, washed, re-suspended in saline solution and stored at 4 °C prior to use.

To examine the affinity of the selected model colloids to anionic P, zeta potential (ζ) of the colloidal solutions were measured using a Zetasizer Nano ZS (Malvern Instruments) in near neutral pH in Milli-Q water. Concentration used in the measurements was 2.5 mg/L for

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