



Leaching of natural colloids from forest topsoils and their relevance for phosphorus mobility

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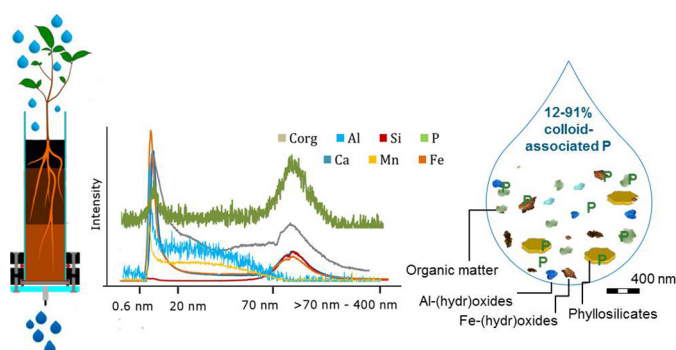
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HIGHLIGHTS

- Natural colloids up to a size of 400 nm leached from forest topsoil columns.
- Leached colloids were dominated by soil organic matter.
- Large proportions of P leached from loamy soils were bound to colloids.
- The fraction of P bound to colloids decreased with rising P leachate concentrations.

GRAPHICAL ABSTRACT



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ABSTRACT

The leaching of P from the upper 20 cm of forest topsoils influences nutrient (re-)cycling and the redistribution of available phosphate and organic P forms. However, the effective leaching of colloids and associated P forms from forest topsoils was so far sparsely investigated. We demonstrated through irrigation experiments with undisturbed mesocosm soil columns, that significant proportions of P leached from acidic forest topsoils were associated with natural colloids. These colloids had a maximum size of 400 nm. By means of field-flow fractionation the leached soil colloids could be separated into three size fractions. The size and composition was comparable to colloids present in acidic forest streams known from literature. The composition of leached colloids of the three size classes was dominated by organic carbon. Furthermore, these colloids contained large concentrations of P which amounted between 12 and 91% of the totally leached P depending on the type of the forest soil. The fraction of other elements leached with colloids ranged between 1% and 25% (Fe: 1–25%; C_{org}: 3–17%; Al: <4%; Si, Ca, Mn: all <2%). The proportion of colloid-associated P from the forest surface soil did not increase with increasing bulk soil P concentrations and were also not related to tree species. The present study highlighted that colloid-facilitated P leaching can

Abbreviations: LC0.6–20, leached colloids 0.6 nm–20 nm; LC20–70, leached colloids 20 nm–70 nm; LC70–400, leached colloids 70 nm–400 nm; WDC, water-dispersible colloids; OM, organic matter; BBR, Bad Brückenau; VES, Vessertal; LUE, Lüss; FFF, field-flow fractionation; TEM, transmission electron microscopy; EDX, energy-dispersive X-ray spectroscopy; OCD, organic carbon detector; DLS, dynamic light scattering; UV, ultraviolet; ICP-MS, inductively coupled plasma mass spectrometry; P_{org}, organic phosphorus; Al_{ox}, oxalate extracted aluminium; Fe_{ox}, oxalate extracted iron; Fe_{DCB}, dithionite extracted iron; MRP, molybdenum reactive phosphorus; MUP, molybdenum un-reactive phosphorus; P_{tot}, total P concentration of the leachate.

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1. Introduction

Phosphorus (P) is one of the limiting nutrients for all organisms (Stevenson and Cole, 1999). The soil is the major source for P for the flora and fauna in terrestrial ecosystems like forests. The primary source of P in soils is rock weathering (Stevenson and Cole, 1999). However, during pedogenesis the recycling of organic P (P_{org}) gets increasingly important (George et al., 2017; Lang et al., 2016; Walker and Syers, 1976). Microorganisms have a key function in P status and cycling of soils (Bergkemper et al., 2016). In forest soils, P_{org} and ortho-phosphate, being released during the decomposition of organic matter, become transferred from the organic surface layer into and through the mineral soil with the percolating soil water. It was shown that P is translocated e.g. due to podzolisation and transferred from top- to sub-soil (Backnas et al., 2012; Turner et al., 2012). However, the P retention was shown to be high in the upper mineral soil (Jobbágy and Jackson, 2001) and related to the soil clay and sesquioxide content (Vaananen et al., 2008). Furthermore, the soil composition and pH affects P binding to the soil matrix. The main reasons for large P retention in soils are that ortho-phosphate and P containing compounds are highly surface-affine. Under acidic soil conditions which prevail in most forest soils, P becomes partly fixed to soil mineral compounds in particular to iron- and aluminium-(hydr)oxides (Missong et al., 2017; Regelink et al., 2015; Vaananen et al., 2008; Vincent et al., 2012). Additionally, ortho-phosphate can form associations with dissolved and particulate organic matter through polyvalent cation bridges (Borie and Zunino, 1983; Tipping et al., 1995; Whittinghill and Hobbie, 2012). In a recent agricultural soil study, 0.3–1.8% of P was leached from bacterial biomass (Glaesner et al., 2016).

Metal-(hydr)oxides, phyllosilicates and large organic matter molecules were often reported to be typical components of natural nanoparticles (<100 nm) and colloids (<1 μm) (Tsao et al., 2011). The binding of P to such colloids is mostly strong and due to their small size and large surface areas, colloids can be enriched in P per mass unit in comparison to the bulk soil (Missong et al., 2017). In literature, it was shown that colloids containing P can also be mobilized and transported in soils, meaning that P transport can proceed not only in dissolved form, but also in association with colloids (de Jonge et al., 2004b; Granger et al., 2007; Rick and Arai, 2011; VandeVoort et al., 2013; Vendelboe et al., 2011). Column studies reported the colloid-associated P release from soils (de Jonge et al., 2004a; Holzmann et al., 2016; Ilg et al., 2005; Siemens et al., 2004; Siemens et al., 2008). A recent study showed that colloid-associated P could even dominate total P leaching in comparison to dissolved reactive P (Sharma et al., 2017). Moreover, it was shown, that the P transport in well aggregated soils occurs to large extend along preferential-flow paths (Julich et al., 2017b), which were also described as flow-path for colloids. Little information is available regarding the proportion of colloid-facilitated P transport in relation to total P transport as well as the nature of colloids that carry the P, especially in forest soils (Bol et al., 2016). One reason for this lack of information is the standard (filter) pore size of 450 nm that is often used for separating dissolved and particulate substances, overlapping with the size range of natural colloids (Gottselig et al., 2017a, 2017b; Haygarth et al., 1997; Hens and Merckx, 2001). Therefore, in studies applying standard analytical protocols involving a filtration with a pore size of 450 nm (Haygarth et al., 1997), the P associated with nanoparticles and fine colloids (100 < d < 400 nm) was included in the so-called “dissolved” fraction (Gottselig et al., 2017a, 2017b).

The soil properties influencing the P and colloid transfer in soils are diverse. The mobilization of colloids and associated P is affected by soil

texture, structure and porosity. Additionally, the pore water chemistry, the dynamics of soil water contents, and water flow velocity influence the colloid mobilization and mobility (de Jonge et al., 2004a; Heathwaite and Dils, 2000; Henderson et al., 2012; Vendelboe et al., 2011; Vendelboe et al., 2012). Moreover, the particle size and type affect the stability of suspensions of nanoparticles and colloids (Kaplan et al., 1993), and with it the colloid-facilitated leaching of P. The common source for mobile colloids leached from surface soils is the (top)soil itself. In general the water movement imposes shear stress on the soil matrix thereby causing colloid release. Therefore, the composition of colloids in soil leachate is probably mostly similar to the composition of the fine size fractions in soils (Kretzschmar et al., 1999). Moreover, the organic carbon (C_{org}) concentration is an important parameter, since organic matter attached to the particle surface increases the dispersibility of colloids in soil solution by shifting their surface charge to more negative values and with it impede flocculation or attachment to the soil matrix also by steric hindrance (Kretzschmar et al., 1995). This colloid stabilization mechanisms were also described for ortho-phosphate (Celi and Barberis, 2005) and P_o (Ognalaga et al., 1994) as highly colloid-stabilizing compounds, e.g. for Fe-oxides (Ilg et al., 2008; Siemens et al., 2004; Swartz and Gschwend, 1998). However, it is to note that in most colloid studies, the size ranges chosen for colloidal size classes (e.g. 220 nm–450 nm) were operationally related to filter pore sizes. Further studies are needed characterising the composition and size of soil colloid associated with P to better understand leaching and re-distribution of P in forest soils (Bol et al., 2016).

Field-Flow Fractionation (FFF) is increasingly used to separate natural nanoparticles and fine colloids according to their size (Baken et al., 2016; Gottselig et al., 2014; Gottselig et al., 2017a, 2017b; Jiang et al., 2015a; Jiang et al., 2015b; Missong et al., 2017; Neubauer et al., 2013; Regelink et al., 2013; Regelink et al., 2014; von der Kammer et al., 2011). These studies showed that colloids in stream water and water dispersible colloids (WDC) can be separated into different nanoparticulate and fine colloidal size fractions. We expected that the composition of leached colloids could be intermediate between WDC extracted from the soils (Missong et al., 2017) and colloids present in stream waters (Gottselig et al., 2017a, 2017b). Furthermore, we also anticipated that due to the thick organic layer covering the mineral soil, a large fraction of organic matter colloids and of organo-mineral aggregates become leached.

Lang et al. (2016) proposed that P cycling and P-losses from forest soils vary systematically with soil P stocks. Therefore, we selected three forest soils with differing P bulk soil concentrations for answering the following questions:

- 1) Do the specific size fractions of natural colloids leached from forest topsoils have characteristic element distributions (“fingerprints”) and how does this coincide to soil WDC and stream colloids?
- 2) Do the proportions of dissolved and colloidal P leaching concentrations differ between the three forest sites with different P stocks?
- 3) To what extent are the abiotic or biotic factors (e.g. soil texture, structure or vegetation) important for the colloidal and total P leaching?
- 4) How do type and size fraction of colloids quantitatively influence total P leaching?

We addressed these questions in a mesocosm experiment with undisturbed soil columns of three forest topsoils that were irrigated with artificial rain. The soil substrates were sandy to loamy, with different degree of pedogenetic transformation of primary P-minerals and varying

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