



Multiple exchange processes on mineral surfaces control the transport of dissolved organic matter through soil profiles



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ARTICLE INFO

Keywords:

Cascade model

DOM

Reactive minerals

¹³C

NanoSIMS

Microbial community composition

ABSTRACT

Organic topsoil layers are important sources of dissolved organic matter (DOM) transported to deeper soil layers. During passage through the mineral soil, both organic matter (OM) quality and quantity change markedly. Whether these alterations are due to sorption processes alone or to additional stepwise exchange processes of OM on mineral surfaces (“cascade model”) is not fully understood. To test the “cascade model”, we conducted a laboratory flow cascade experiment with undisturbed soil columns from three depths of two different soil profiles (Dystric and Eutric Cambisol) using carbon (C) isotope labelling. Each of the connected topsoil and subsoil columns contained a goethite (α -FeOOH) layer either with or without sorbed ¹³C-labelled OM to assess the importance of OM immobilization/mobilization reactions with reactive soil minerals. By using a multiple method approach including ¹³C analysis in the solid and solution phases, nanometer scale secondary ion mass spectrometry (NanoSIMS), and quantitative polymerase chain reaction (qPCR), we quantified organic carbon (OC) adsorption and desorption and net OC exchange at goethite surfaces as well as the associated microbial community patterns at every depth step of the column experiment. The gross OC exchange between OM-coated goethite and the soil solution was in the range of 15–32%. This indicates that a considerable proportion of the mineral associated OM was mobilized and replaced by percolating DOM. We showed that specific groups of bacteria play an important role in processing organic carbon compounds in the mineral micro-environment. Whereas bulk soils were dominated by oligotrophic bacteria such as *Acidobacteria*, the goethite layers were specifically enriched with copiotrophic bacteria such as *Betaproteobacteria*. This group of microorganisms made use of labile carbon derived either from direct DOM transport or from OM exchange processes at goethite surfaces. Specific microorganisms appear to contribute to the cascade process of OM transport within soils. Our study confirms the validity of the postulated “cascade model”, featuring the stepwise transport of OM within the soil profile.

1. Introduction

Dissolved organic matter (DOM) mobilized in topsoil and transported to subsoil horizons is an important source of carbon (C) throughout the soil profile. In most soils the concentration of dissolved organic carbon (DOC) declines strongly with soil depth, with up to 90% net loss in the first meter of the soil profile (Michalzik et al., 2001). This is likely due to its adsorption to reactive minerals such as iron (Fe)

oxides (Kaiser and Guggenberger, 2000) and clay minerals (Saidy et al., 2013), or to co-precipitation with aluminum (Al) and Fe (Scheel et al., 2008; Mikutta et al., 2014). According to Kalbitz and Kaiser (2008), as much as 19–50% of the organic matter (OM) in the subsoil of a sandy Podzol can be derived from DOM.

Positive correlations between the content of pedogenic Al and Fe with the content of soil organic carbon (OC) (Kaiser and Guggenberger, 2000) and of OC resistant to chemical degradation (Mikutta et al.,

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2006) suggest that the association of OM with Fe and Al oxides is an important stabilization mechanism for OM. Especially in acidic soil environments, such as in Dystric Cambisol subsoils, Fe oxides with high specific surface area (SSA) provide reactive surfaces that are important for the binding of OM (Eusterhues et al., 2005). Gu et al. (1994) and Kaiser et al. (1997) concluded that the adsorption of DOM to Fe oxides is dominated by ligand exchange involving carboxyl and hydroxyl functional groups, promoting oxidized aromatic moieties (i.e., lignin-degradation compounds) over carbohydrates in the sorption process (Chorover and Amistadi, 2001; Kaiser, 2003). Thus not only does the concentration of DOC decrease with soil depth but the composition of DOM also changes. A relative enrichment in carbohydrates and sugars along with the selective removal of carboxylated aromatic moieties from soil solution has been observed in batch experiments (Kaiser et al., 1997), saturated soil column experiments (Guo and Chorover, 2003), and in field experiments (Kaiser et al., 2004; McCarthy et al., 1996). Binding of DOM to Fe oxides such as goethite is not only selective for specific compounds, but can also lead to a strong reduction in SSA due to clogging of micropores by selective sorption at the mouths of pores (Kaiser and Guggenberger, 2007). At higher OM loadings, multiple layers of OM are possibly formed on the mineral surfaces, suggesting stronger binding and stabilization of OM that is in direct contact with the mineral surface (Kaiser and Guggenberger, 2000) and enhanced exposure of the outermost OM layer to desorption and exchange processes.

A major source of DOM in temperate forest soils is leaching from the forest floor, leading to highest DOC concentrations directly below the soil organic layer (Michalzik et al., 2001). Most of this DOM is either retained or consumed in the upper centimeters of the mineral soil (Fröberg et al., 2007), whereas DOM at greater soil depths is not directly derived from the organic layer (Hagedorn et al., 2004). Based on such observations, Kaiser and Kalbitz (2012) proposed a stepwise cascade of adsorption to reactive minerals, microbial transformation, and re-release into the soil solution during the transport of DOM down the soil profile. This cascade assumes that previously bound, more degraded OM moieties get remobilized by the input of fresh highly surface-reactive plant-derived DOM and are transported further down the soil profile where they replace and consequently remobilize even older OM. This conceptual view, therefore, could link the chemical fractionation of DOM during passage down the soil column with the increasing ^{14}C age of DOM and OM as soil depth increases (Sanderman et al., 2008), but experimental data is lacking.

The microbial community adapts to the quantity of bioavailable OM; thus microbial community composition indicates co-evolution with concurrently changing OM properties as soil depth increases (Heckman et al., 2013). In deeper soil, the magnitude of OM processing differs considerably between different soil compartments. Microbial densities are mostly low and heterogeneously distributed depending on OM availability and chemical composition (Preusser et al., 2017; Angst et al., 2016). In contrast to the generally slow process rates in subsoil, reactive minerals such as goethite are known as hotspots of biogeochemical interactions (Eusterhues et al., 2005) and may therefore be of major importance for OM processing in deeper soil layers. Nevertheless, several physicochemical characteristics of water-extractable OM (apparent molar mass, pH, and electrical conductivity) may modify

responses of the bacterial and fungal communities in the presence of Fe and Al oxide phases (Heckman et al., 2013).

The objective of this study, therefore, was to test the “cascade model” of Kaiser and Kalbitz (2012) by elucidating whether changes in amounts and composition of DOM within soil profiles are due to sorption processes alone or to additional stepwise exchange of OM on mineral surfaces, processes which may be driven in part by microbial activity. We hypothesize that (i) the input of plant-derived DOM to mineral topsoils leads to selective adsorption of plant-derived compounds to reactive surfaces; (ii) fresh DOM input partially replaces older mineral-associated OM, which subsequently gets remobilized and further transported to deeper soil; and (iii) these mineral-organic associations act as biogeochemical hotspots of high resource availability leading to changes in microbial abundance and community composition. These hypotheses were tested by a column experiment using three connected undisturbed soil horizons from two soils (Dystric and Eutric Cambisol), each horizon containing a thin interspersed layer of goethite coated with ^{13}C -labelled OM. Incorporation of the labelled OC made it possible to quantify net OC retention and DOM-induced exchange of goethite-associated OM. For selected samples, nanoscale secondary ion mass spectrometry (NanoSIMS) was used to study the microscale (im) mobilization of OM at the goethite surfaces. Concomitant shifts in microbial community composition in the bulk soil and the goethite layers were analysed by domain- and taxa-specific quantitative polymerase chain reaction (qPCR) assays.

2. Materials and methods

2.1. Soil sampling

Undisturbed soil cores of 100 cm^3 (ϕ 5.7 cm, h 4.0 cm) were taken from two sites in Lower Saxony, Germany; a sandy Dystric Cambisol (IUSS Working Group WRB, 2014) developed from Pleistocene fluvial and aeolian sandy deposits at the Grindewald ($52^\circ 34'22.12''\text{ N}$, $9^\circ 18' 49.76''\text{ E}$), and a silty Eutric Cambisol (IUSS Working Group WRB, 2014) developed from basalt near Dransfeld ($51^\circ 28' 35.60''\text{ N}$, $09^\circ 45' 32.46''\text{ E}$). Both sites were covered with evenly aged (~ 100 years) European beech (*Fagus sylvatica* L.) forest. Soil cores were taken from three depths: 4 cm, 30 cm and 100 cm at the sandy site, and 4 cm, 12 cm and 26 cm at the silty site, because core sampling in deeper soil layers at the silty site was impossible due to high stone content near the solid bedrock (Table 1).

2.2. Preparation of goethite coated with ^{13}C -labelled organic matter

Goethite was produced by increasing the pH of a 0.5 M FeCl_3 solution to 12 with 5 M NaOH and subsequent aging of the precipitate to goethite at 55°C for 48 h (Schwertmann and Cornell, 2000). The suspension was dialyzed against double deionized water until the conductivity was $< 10\ \mu\text{S cm}^{-1}$ and subsequently frozen in liquid nitrogen, freeze-dried, and sieved $< 200\ \mu\text{m}$. For the organic matter coating, ^{13}C -labelled DOM was prepared by extraction of a mixture of 10 g labelled beech leaves (12.3 atom% ^{13}C) (IsoLife BV, Wageningen, Netherlands) and 240 g of naturally grown beech leaves in 2500 mL double deionized water (1:10, w/v), to limit the need for expensive ^{13}C -labelled plant

Table 1

Basic physical and chemical soil properties of the three sampling depths in each of the two soils before the experiment.

Texture	Horizon	Depth (cm)	pH (CaCl_2)	C (mg g^{-1})	N (mg g^{-1})	C/N	$\delta^{13}\text{C}$ (‰)	Fe_d (mg g^{-1})	Sand (%)	Silt (%)	Clay (%)
Sand	AE	4	3.23	56.31	0.80	27.46	-28.32	2.97	54	41	5
Sand	Bw	30	4.27	5.99	0.32	18.80	-26.62	2.46	56	39	5
Sand	Cw	100	4.28	0.20	0.06	3.63	-26.41	1.38	76	23	1
Silt	A	4	3.85	48.79	3.75	13.02	-26.54	20.85	0	89	11
Silt	Bw	12	3.88	44.80	3.57	12.53	-26.28	21.49	0	87	13
Silt	Bw	26	4.27	30.25	2.77	10.94	-25.96	22.4	1	89	11

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