



Tracking litter-derived dissolved organic matter along a soil chronosequence using ^{14}C imaging: Biodegradation, physico-chemical retention or preferential flow?



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ABSTRACT

The cycling of dissolved organic matter (DOM) in soils is controversial. While DOM is believed to be a C source for soil microorganisms, DOM sorption to the mineral phase is regarded as a key stabilization mechanism of soil organic matter (SOM). In this study, we added ^{14}C -labelled DOM derived from *Leucanthemopsis alpina* to undisturbed soil columns of a chronosequence ranging from initial unweathered soils of a glacier forefield to alpine soils with thick organic layers. We traced the ^{14}C label in mineralized and leached DOM and quantified the spatial distribution of DO^{14}C retained in soils using a new autoradiographic technique. Leaching of DO^{14}C through the 10 cm-long soil columns amounted up to 28% of the added DO^{14}C in the initial soils, but to less than 5% in the developed soils. Biodegradation hardly contributed to the removal of litter- DO^{14}C as only 2–9% were mineralized, with the highest rates in mature soils. In line with the mass balance of ^{14}C fluxes, measured ^{14}C activities in soils indicated that the major part of litter DO^{14}C was retained in soils (>80% on average). Autoradiographic images showed an effective retention of almost all DO^{14}C in the upper 3 cm of the soil columns. In the deeper soil, the ^{14}C label was concentrated along soil pores and textural discontinuities with similarly high ^{14}C activities than in the uppermost soil. These findings indicate DOM transport via preferential flow, although this was quantitatively less important than DOM retention in soils. The leaching of DO^{14}C correlated negatively with oxalate-extractable Al, Fe, and Mn. In conjunction with the rapidity of DO^{14}C immobilization, this strongly suggests that sorptive retention DOM was the dominating pathway of litter-derived DOM in topsoils, thereby contributing to SOM stabilization.

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1. Introduction

Dissolved organic matter (DOM) represents the mobile phase of soil organic matter and plays an important role in many biogeochemical processes. Although the fluxes of DOM are small compared to litter inputs and respiration on a local and global scale (Battin et al., 2009; Kindler et al., 2011), the leaching of DOM from topsoils and its sorption in the mineral soil can contribute significantly to the long-term accumulation and preservation of SOM (Kaiser and Guggenberger, 2000; Kaiser and Kalbitz, 2012). In addition, DOM is the major export pathway for essential nutrients

such as nitrogen, phosphorous and sulfur from forest ecosystems (e.g. Hagedorn et al., 2000; Michalzik et al., 2001; Kaiser et al., 2003) and it plays a key role in the transport of metals and organic contaminants (Tipping, 2002).

While the fluxes and functional role of DOM have been addressed in a number of studies, the mechanisms behind DOM cycling in soils – mobilization and immobilization – are still not fully understood (Kaiser and Kalbitz, 2012). Principally, DOM derives from leaching of plant litter and microbial detritus, exudation by roots, and solubilisation of soil organic matter, but the relative contribution of each of these sources is still uncertain and differs among ecosystems, soil types and soil depths (e.g. Hagedorn et al., 2004; Sanderman and Amundson, 2008; Malik and Gleixner, 2013). In mineral soils, DOM concentrations typically decrease with soil depth, indicating a net DOM retention. However, different concepts

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for the underlying retention mechanisms exist among scientific disciplines. A biology-centered view assumes DOM to be highly bio-available, because soil microorganisms take up C in soluble forms (e.g. Kuzyakov, 2010; Kiikkilä et al., 2012) and hence, biodegradation has been assumed to be of primary importance at least for fresh litter leachates (Cleveland et al., 2004; De Troyer et al., 2011). In contrast, organic geochemists consider that DOM is mainly removed in soils by sorption to reactive surfaces or precipitation, and DOM transport is primarily controlled by physico-chemical properties of DOM (Jardine et al., 1989; Kaiser et al., 1996; Kaiser and Guggenberger, 2000). In a survey of DOM fluxes in 12 European sites differing in land-use, Kindler et al. (2011) observed that the fraction of DOM retained in subsoils correlated positively with the ratio between poorly crystalline Fe and Al-oxides (as a measure of mineral surface reactivity) and soil organic matter (representing a DOM source or the occupation of sorption sites). However, during storm events, the sorptive DOM retention might get by-passed by preferential flow, leading to a more rapid and longer transport than expected based on the surface reactivity of the soil matrix (Hagedorn et al., 2000; Kaiser and Guggenberger, 2005).

Tracer experiments are powerful to disentangle the retention but also the mobilization mechanisms of DOM in soils. Experimental additions of labelled litter show that only a minor fraction of litter-derived DOM reaches deeper soil horizons, while total DOC concentrations decrease only slightly with soil depth at least in topsoils (Fröberg et al., 2007a; Müller et al., 2009; De Troyer et al., 2011; Kammer and Hagedorn, 2011; Tipping et al., 2012). So far, DOM tracer studies using ^{13}C -labelled organic matter or ^{14}C -‘bomb’ carbon have been case studies in specific ecosystems and soils (e.g. forests: Fröberg et al., 2007b, Sanderman and Amundson, 2008; grasslands: Hagedorn and Joos, 2014). While these experiments all indicate a strong retention of litter-derived ‘new’ DOM, they have failed to identify the mechanism and spatial dimension of DOM retention as the tracer signal disappears in the large SOM ‘reservoir’ in soils. The aim of our study was to gain insight into the fate of ‘new’ litter-derived DOM in soils and to identify the mechanism that control DOM retention. We have set up a soil column experiment, in which we added highly ^{14}C -labeled litter-derived DOM and traced the ^{14}C signal in soil-respired CO_2 and leached DOM, but also in the soil solid phase. In addition, we applied ^{14}C imaging by autoradiography (Stiehl-Braun et al., 2011) to visualize the small-scale spatial distribution of ^{14}C and hence DO^{14}C retention in soils. For our experiment, we used soils of a chronosequence from a glacier forefield, providing a gradient in soil properties reaching from ‘initial’ unweathered soils with low SOM contents to ‘old’ fully developed soils with thick organic layers and high contents of pedogenic oxides (Bernasconi et al., 2011; Dümig et al., 2011; Smittenberg et al., 2012). We hypothesized that (1) the retention of litter DOM will increase with soil development, (2) biodegradation will increase with soil age as a result of an increasing biomass and diversity of soil microbial communities, (3) the sorptive retention will be highest in intermediately developed soils with the highest contents of potential sorbents, pedogenic oxides relative to soil organic matter, (4) preferential flow leads to a bypassing of the soil matrix, thereby reducing the biotic and abiotic DOM retention.

2. Materials and methods

2.1. Study sites and soil sampling

We selected soils covering a gradient in soil development from ‘initial’ unweathered soils to strongly developed soils with organic layers, hence soils that represent a great variability in soil

properties potentially affecting DOM cycling. We chose soils from acidic bedrock in the Swiss Alps: five soils of the chronosequence of the Damma glacier (46°38' N, 8°27' E; 1950–2050 m a.s.l.) developed from coarse-grained granite (Bernasconi et al., 2011) and one treeline soil (Cambic Podzol), with a thick mor-type organic layer from Stillberg near Davos (46°46' N, 9°52' E; 2100 m a.s.l.), developed on siliceous Orthogneiss of the crystalline Silvretta nappe (Hagedorn et al., 2008; Müller et al., 2009). The Damma glacier has not receded continuously since the end of the last ice age in 1850 due to glacier advances from 1920 to 1928 and from 1970 to 1992, resulting in two small terminal moraines (Bernasconi et al., 2011). Hence, the soil chronology consists of four distinct groups of soil ages: <15 y, 60 to 80 y, 110 to 140 y (Hyperskeletal Leptosols, WRB, 2006) and a reference site outside the forefield (Haplic Cambisol) with a soil age of at least 700 y. In our study, we have sampled soils from each of these age classes avoiding fresh alluvial material. At the youngest site, we took samples from sandy frequently flooded depressions and gravelly mounds (named 10 y sand, 10 y gravel), which were assumed to have distinctly different flow regimes due to their texture. Moreover, *in situ* measurements have indicated that DOC and CO_2 fluxes differed strongly among these microsites (Guelland et al., 2013a,b). While the vegetation is patchy at the initial sites, grasses dominate at the intermediate sites (named 70 y) (*Agrostis gigantea*, *Deschampsia cespitosa* (L.) Roem. and Schult., *Athyrium alpestre* (Hoppe) Milde.). The vegetation cover is full at the older sites (named 120 y) with dwarf shrubs such as *Rhododendron ferrugineum* L. and *Salix* spp., and grasses like *A. gigantea*. The reference site of the Damma glacier, the so-called ‘grassland’ is covered by the grass *A. gigantea* and ferns while the ‘treeline’ site is dominated by the dwarf shrubs *Vaccinium myrtillus* L., *V. gaultherioides* Bigelow and *Empetrum nigrum* ssp. *hermaphroditum* (Hagerup) Böcher. Details and additional information on climate, vegetation and soil properties can be found in Hagedorn et al. (2008), Bernasconi et al. (2011) and Smittenberg et al. (2012).

In August 2012, we sampled four soil columns at each of the six sites (10 y sand; 10 y gravel; 70 y; 120 y, grassland, treeline). The columns, which were located at least 10 m apart, were sampled by hammering PVC tubes of 11 cm height and 10 cm in diameter into the ground and by carefully excavating them with a spade. For transport, they were capped with PVC lids at the bottom and the top and stored at 4 °C in a cooling chamber during several weeks until the beginning of the experiments. At all locations, additional soil samples were taken with a steel cylinder of 11 cm height and 10 cm diameter for the analysis of soil properties. The excavated columns were horizontally divided into five 2 cm-soil slices.

For analysis, soils were first dried at 60 °C for five days, weighed and then sieved to <2 mm. Stones were weighed and their volume >2 mm was determined by water displacement in a graduated cylinder. Porosity of the soil columns was calculated from bulk density ρ_b [g cm^{-3}] and particle density ρ_p [g cm^{-3}] assuming particle densities of 2.65 g cm^{-3} for stones and 1.4 g cm^{-3} for organic matter. Soil pH was determined from the <2 mm soil fraction in 0.01 M CaCl_2 with a soil/solution ratio of 1:2. Pedogenic oxides were characterized by extracting them with dithionite-citrate-bicarbonate (Al_d , Fe_d , Mn_d) using the method of Mehra and Jackson (1960) and by NH_4 -oxalate (Al_{ox} , Fe_{ox} , Mn_{ox}) according to Schwertmann (1964). In the extracts, metal concentrations were determined by ICP-OES (Optima 3000, Perkin-Elmer, Waltham, MA, USA). For the analysis of soil organic C (SOC) and nitrogen (N) contents, aliquots of the <2 mm fraction were finely powdered with a swing mill (Schwingmühle MM 20, Retsch, Haan, Germany). Samples were then measured with a C–N elemental analyser (CE Instruments NC 2500, ThermoQuest Italia SpA, Radano, Italy).

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