



# Contribution of the vertical movement of dissolved organic carbon to carbon allocation in two distinct soil types under *Castanopsis fargesii* Franch. and *C. carlesii* (Hemsl.) Hayata forests

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## Abstract

• **Key message** The vertical transport of dissolved organic carbon (DOC) is an important determinant of carbon distribution across a soil profile. The transport of DOC down a soil profile can be largely influenced by incoming DOC and soil organic carbon (SOC) levels, which insulate DOC from adsorption processes regulated by soil texture and Fe/Al mineralogy.

• **Context** Uncertainties about how soil properties affect DOC transport through the soil profile require study because soils can differ strongly with respect to texture or Fe/Al mineralogy and yet retain similar quantities of DOC.

• **Aims** This study aimed to assess the role of incoming DOC and native SOC in regulating DOC migration in soils and investigate the contribution of DOC movement to SOC allocation.

• **Methods** We leached a standard DOC solution extracted from *Castanopsis carlesii* litter through two distinct soil types, using two leaching strategies: single leaching and sequential leaching. The two soil types under a natural *Castanopsis carlesii* (Hemsl.) Hayata forest and a natural *Castanopsis fargesii* Franch. forest, respectively, differ strongly with respect to soil texture, Fe/Al oxide abundances, and SOC nature.

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## Contribution of the co-authors

Dr. Youtao Si participated in every step of the study and especially put effort in experimental design, DOM characterization, data analysis, and finally writing the paper.

Li Xiong performed part of the experiment.

Yuehmin Chen supervised the work and wrote part of the paper.

Jinmao Zhu ran part of the data analysis.

Jinsheng Xie provided advices on how to write the paper.

Ren Gao coordinated the research project.

Yusheng Yang supervised the work.

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• **Results** With single leaching, where each of six soil layers making up an entire 0–100-cm soil depth profile received single doses of standard DOC solution, deeper soil layers retained more DOC than upper soil layers, with native SOC largely masking the effects of soil texture and Fe/Al mineralogy on DOC migration. Following sequential leaching, where a sixfold larger amount of standard DOC solution sequentially percolated through the six soil layers, the upper soil layers generally retained more DOC than deeper layers. Nevertheless, in sequential leaching, desorption-induced transfer of carbon from upper soil layers to deeper soil layers resulted in greater total carbon retention than in single leaching.

• **Conclusion** Forest subsoils (40–100 cm) are well below C saturation, but DOC vertical movement from top soils only transfers limited organic carbon to them. However, DOC vertical movement may greatly alter SOC allocation along the top soil profile (0–40 cm), with part of outer sphere native SOC displaced by incoming DOC and migrating downwards, which is a natural way to preserve SOC.

**Keywords** Dissolved organic carbon · Adsorption · Desorption · Soil organic carbon allocation

## 1 Introduction

Soil organic carbon (SOC) is a sensitive indicator of soil quality and a large component of the global carbon budget. Dissolved organic carbon (DOC) is the most reactive component of SOC because it is highly mobile and contains diverse functional groups. It is a major vehicle for the movement of carbon from surface to deep soils and contributes significantly to SOC dynamics and sequestration (Kalbitz et al. 2000; Tipping et al. 2012; Toosi et al. 2012). It was reported that 25–98% of SOC is associated with the fine soil fraction, and the dominant mechanism is chemically sorptive protection of DOC (Basile-Doelsch et al. 2007; Jagadamma et al. 2014).

Early work on DOC chemistry assumed that only surface-reactive (highly sorptive) substances such as phenolic acids were preferentially removed from incoming DOC by adsorption, whereas no desorption of native SOC occurred (Guggenberger and Kaiser 2003; Jason and Gregory 2001). A cycling downwards model was then proposed, in which, besides adsorption, desorption of microbial processed carbon by fresh plant-derived carbon also drives DOC movement (Kaiser and Kalbitz 2012). Scott and Rothstein (2014) used DOC fractionation to show that DOC collected from deeper soils included desorbed SOC from upper soils and corroborated that desorption was involved in net sorption dynamics across a soil profile. The idea of potential solubility of sorbed organic carbon has now been widely incorporated into hydrological modeling (Dusek et al. 2017; Yurova et al. 2008).

DOC adsorption and desorption occur throughout the soil profile, and the balance of these processes depends on interactions between soil surfaces and incoming DOC. Forest soil properties like soil texture and abundance of Fe/Al oxides may influence these interactions (Kaiser and Guggenberger 2000; Kothawala et al. 2009; Schmidt et al. 2011). Nevertheless, the extent to which soil properties impact DOC migration is uncertain. Scott and Rothstein (2014) found that six contrasting types of forest soil retained similar quantities of DOC when supplied with a common DOC solution, despite wide variation

in textural properties (clay content ranged from 0.1 to 5.2%) and Fe/Al mineralogy (iron hydrous oxide content ranged from 2.9 to 11.6%). Top soils (typically 0–40 cm deep) with higher SOC contents often have limited binding sites, but in the fields, they often show higher DOC retention than subsoils (typically 40–100 cm deep) which contain more clays and Fe/Al oxides (Kalbitz and Kaiser 2008; Kothawala et al. 2009; Lilienfein et al. 2004).

Solving this paradox will require more fully understanding the nature of SOC and the covering of mineral soil surfaces, which contrasts juvenile minerals. It has been demonstrated that SOC can impede exogenous DOC from reacting with soil minerals (Guggenberger and Kaiser 2003), thereby reducing the sorptive capacity of soil particles and perhaps causing soils that differ with respect to texture or mineralogy to exhibit similar retention rates. Inner sphere SOC is directly associated with minerals, and reduces the availability of binding sites on the soil surface, and so is it typically associated with long residence times. In contrast, outer sphere SOC is loosely bound and interacts more readily with exogenous DOC (Guggenberger and Kaiser 2003; Kleber et al. 2015). The outer sphere SOC either adsorbs exogenous DOC or is displaced by the DOC, depending on the nature of the exogenous DOC and the outer sphere SOC itself. Hot water-extractable organic carbon (HEOC) often represents the most reactive part of the outer sphere SOC (Bu et al. 2010), with total SOC amount and chemistry determining outer sphere SOC amount and chemistry, which in turn can determine interactions with DOC inputs. Critically, few studies have explored the role of SOC in regulating the vertical movement of DOC through soils with contrasting chemical and physical properties.

A second entry point into solving the above paradox is understanding how the amount and quality of incoming DOC affect adsorption and desorption dynamics across a soil profile. The more highly sorptive DOC is preferentially retained in upper soil layers, which diminishes the quantity and sorptive quality of the DOC that enters deeper soil layers

and so reduces retention rates in deeper soils (Guggenberger and Kaiser 2003; Kothawala et al. 2009). There is, however, very little information on how DOC chemistry and amount affect retention dynamics across a soil profile. We suggest that it is possible to examine the factors that affect DOC retention rates by examining retention efficiency and sorption/desorption dynamics across a soil profile by subjecting contrasting soil profiles and different soil layers within each profile to fixed amounts of a common DOC.

Organic matter fractionation into hydrophobic and hydrophilic components is a standard method for analyzing organic carbon quality in aqueous solutions. It is widely believed that hydrophobic compounds are more sorptive than hydrophilic compounds (Gu et al. 1994; Kaiser and Guggenberger 2000). Hydrophilic compounds are more likely to be displaced or remain in soil solution and move into deeper soils. Therefore, their concentration would increase with increasing soil depth (Yano et al. 2005). Ultraviolet-visible (UV) and fluorescence spectroscopy have recently been applied in ecological studies as simple but efficient ways to characterize DOC (Bu et al. 2010; Bu et al. 2011; Choe et al. 2010; Colombo et al. 2014; Dai et al. 2006; Foresti et al. 2003; Kim and Hyun 2014; Kleber et al. 2011). The aromaticity index (AI), obtained by dividing the UV-specific absorbance at 254 nm by DOC concentration, can reflect the content of aromatic compounds, whose conjugated  $\pi$  electron system is highly sorptive (Corvasce et al. 2006; Kalbitz et al. 2003; McKnight et al. 1992; Weishaar et al. 2003). The humification index (HIX) calculated from fluorescence spectra is indicative of the microbially processed molecules, such as products derived from microbial decomposition or the condensation of simple structural components (Michel et al. 2006; Ohno 2002; Zsolnay et al. 1999). The combination of fractionation and spectroscopic characterization can provide a more comprehensive view of DOC chemistry.

In this study, we investigated DOC leaching dynamics and chemical changes through two types of soils, each spanning the 0–100-cm depth range and physically divided into six fixed but separated depth layers. Two leaching strategies were used: (i) single leaching, in which each soil layer received a fixed amount of a standard DOC solution and (ii) sequential leaching, where a sixfold larger amount of a standard DOC solution percolated successively through each of the six layers. The standard DOC solution was extracted from the *Castanopsis carlesii* litter. The two soil types are from a natural *C. carlesii* forest and a natural *C. fargesii* forest in mid-subtropical China, respectively. The two soil types were chosen to demonstrate the effect of native soil conditions on DOC leaching dynamics. Besides significant differences in soil texture and Fe/Al oxide abundances, differing vegetation composition between the two forest stands results in distinct SOC nature, which are expected to significantly influence DOC dynamics. In addition, because many natural *C. fargesii* stands

have been converted to *C. carlesii* plantations, using DOC extracted from the *C. carlesii* litter as the standard source of DOC could also provide insight into how forest conversion would affect SOC allocation along depth by altered above-ground DOC input. Combining organic matter fractionation and spectroscopic characterization, this study aimed to (1) assess the roles of incoming DOC and native SOC chemistry in regulating soil DOC migration and (2) investigate the contribution of DOC adsorption/desorption to SOC allocation.

## 2 Materials and methods

### 2.1 Study area

Two dominant types of forest stands in Fujian Province of subtropical China (25° 29'~27° 07' N, 116° 22'~118° 39' E) were selected for soil sampling. Both stands are located in a maritime monsoon climate zone where the mean annual temperature is about 19 °C. One stand is a natural *C. carlesii* forest, located in the *Castanopsis kawakamii* Hayata Nature Reserve of Sanming county (26° 10' N, 117° 27' E). This region has a mean annual precipitation of 1749 mm. Soils in this area are classified as sandy clay loam Ferralic Cambisols (hereafter, FC) according to FAO/UNESCO. The other stand is a natural *C. fargesii* forest located in Ninghua county (26° 19' N, 116° 49' E). This region has a mean annual precipitation of 1688 mm and soils are classified as Humic Alfisols (hereafter, HA).

### 2.2 Soil sampling and soil core preparation

To obtain a wide range of soil conditions, soils were sampled down to 100-cm depth. In July 2015, we selected three sampling points in each of the two stand types with a distance of  $\geq 10$  m between points. At each point, we collected six soil segments at 0–10, 10–20, 20–40, 40–60, 60–80, and 80–100 cm deep. Visible litter and rocks were removed and the soils were sieved (mesh size 2 mm). Portions of the soil samples were analyzed for soil pH, SOC content, particle size distribution, and Fe/Al contents. The remainder of each sample was air-dried and split into two portions—one to extract HEOC (see section 2.5) and the other packed into 4 cm  $\times$  10 cm (diameter  $\times$  length) PVC columns equipped with glass fibers at the bottom to prevent soil loss during leaching. Each PVC column was filled with 151-g air-dried soils to ensure uniform soil core density (1.2 g cm<sup>-3</sup>). Disturbed soil cores rather than intact soil cores were used in this study to reduce soil heterogeneity. All soil cores were kept in a refrigerator for 2 weeks to equilibrate before leaching to eliminate post-disturbance effects.

### 2.3 The standard DOC solution

To isolate the effects of soil properties on leachate chemistry, we created a standard DOC solution for initial DOC input. In July 2015, newly fallen, undecomposed litter from *C. carlesii* was collected, air-dried, and mixed. The standard DOC solution was prepared by extracting the chemically homogeneous litters with ultrapure water (litter mass: ultrapure water volume = 1:100) for 24 h at room temperature and filtering the extract through 0.45- $\mu\text{m}$  polyethersulfone membranes. The extraction time and dilution ratio were chosen in an attempt to extract all the soluble ingredients. The standard DOC solution was freshly made before each use. The concentration of DOC in the standard solution was  $209.1 \pm 8.6 \text{ mg C L}^{-1}$  ( $n = 12$ ), which is comparable to DOC concentrations in Oi and Oe horizon extracts (Yano et al. 2005). Its pH was  $5.6 \pm 0.3$ , with the hydrophobic compound fraction constituting  $80.5 \pm 2.2\%$ .

### 2.4 Soil core leaching

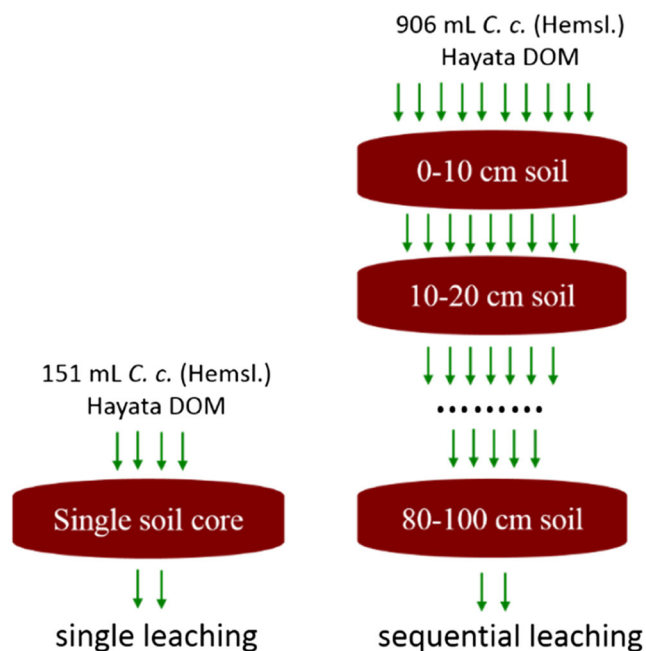
Two leaching methods were used in this study. With single leaching, every soil core was equally and individually supplied with 151-mL standard solution (Fig. 1, left). With sequential leaching, 906 mL ( $6 \times 151 \text{ mL}$ ) of the standard DOC solution was first percolated through the 0–10-cm soils, followed by the leachate through the 10–20-cm layer, and so on until application to the last 80–100-cm soil depth (Fig. 1, right). The standard DOC solution was applied at an input rate of 2 mL/min. Portions of all leachates were collected, filtered through a 0.45- $\mu\text{m}$  polyethersulfone membrane, and promptly analyzed.

### 2.5 Hot water-extractable organic carbon

The air-dried soils (equal to soils for PVC package, see section 2.2) were extracted for HEOC, which was considered as the most reactive part of native SOC. Forty-milliliter ultrapure water and 8-g soils were added to a 50-mL centrifuge tube, then shaken at 80 °C for 1 h. The suspension was centrifuged at 3500 rpm for 15 min and the supernatant was filtered through a 0.45- $\mu\text{m}$  polyethersulfone membrane for immediate analysis.

### 2.6 Analytical methods

SOC content was determined with a soil elemental analyzer (Vario MAX-1, Elementar, Germany). Soil pH was determined in 1:2.5 soil:water suspension (McLean 1982). Soil particle size distribution was determined by the hydrometer method (Zhang et al. 2000). The citrate-dithionite-bicarbonate method was used to determine the content of free Fe and Al oxides ( $\text{Fe}_d$ ,  $\text{Al}_d$ ) (Mehra and Jackson 1960). The contents of amorphous Fe and Al oxides ( $\text{Fe}_o$ ,  $\text{Al}_o$ ) were estimated by extraction with ammonium oxalate at pH 3.0 (McKeague 1978).



**Fig. 1** Schematic representation of single leaching (left) and sequential leaching (right)

All DOC samples were analyzed within 8 h of preparation. The organic carbon concentrations of all DOC solutions (including the standard solutions, HEOC, and leachates) were determined on a TOC-VC/PH/CPN (Shimadzu, Japan). The differences in total DOC contents between incoming solutions and leachates are regarded as total retention. The standard DOC solutions and leachates were fractionated into hydrophobic and hydrophilic organics using XAD-8 exchange resin at pH = 2. Organic carbon absorbed onto XAD-8 was defined as the hydrophobic fraction, not absorbed as hydrophilic fraction (Leenheer 1981). The differences in hydrophobic DOC contents between incoming solutions and leachates are regarded as hydrophobic retention.

Spectroscopic characterization was conducted for all DOC solutions. Fluorescence emission spectra were recorded with a Hitachi F-7000 (Japan,  $\lambda_{\text{ex}}$  254 nm, slit 10 nm;  $\lambda_{\text{em}}$  300–480 nm, slit 10 nm; scanning speed 1200 nm  $\text{min}^{-1}$ ). The ratio of the peak area in the  $\Sigma(435\text{--}480)\text{-nm}$  range to the peak area in the  $\Sigma(300\text{--}345)\text{-nm}$  range was defined as the humification index (HIX) (Zsolnay et al. 1999). UV spectra were obtained on a UV-2450 (Shimadzu, Japan) in cuvettes with 1-cm edge length. Absorption at 254 nm was used to calculate the aromaticity index (AI) according to the equation  $\text{AI} = (\text{UV}_{254} / \text{DOC concentration}) \times 100$  (Zsolnay 2003).

### 2.7 Statistical analyses

For each soil type, basic soil properties (such as sand, silt, clay) or DOC traits (total retention, hydrophobic retention,



**Table 1** Selected characteristics of Ferralic Cambisols (FC) and Humic Alfisols (HA) soils (mean  $\pm$  SD,  $n = 3$ )

Soil depth (cm)	pH	Soil organic carbon (g kg <sup>-1</sup> )	Sand (%) (0.25–0.05 mm)	Silt (%) (0.05–0.002 mm)	Clay (%) (< 0.002 mm)	Fe <sub>d</sub> -Fe <sub>o</sub> (g kg <sup>-1</sup> )	Al <sub>d</sub> (g kg <sup>-1</sup> )
FC soils							
0–10	4.33 $\pm$ 0.18aA	28.24 $\pm$ 2.49aB	60.5 $\pm$ 0.17aA	13.82 $\pm$ 0.66bB	25.69 $\pm$ 0.56aA	21.56 $\pm$ 0.28aA	8.04 $\pm$ 0.43aA
10–20	4.37 $\pm$ 0.13aA	15.27 $\pm$ 1.57bA	60.2 $\pm$ 1.62aA	12.20 $\pm$ 1.96aB	27.60 $\pm$ 1.55aA	24.19 $\pm$ 1.55bA	10.11 $\pm$ 1.64bcA
20–40	4.53 $\pm$ 0.11aA	6.58 $\pm$ 0.98cA	52.4 $\pm$ 0.59bA	15.58 $\pm$ 1.27cB	32.04 $\pm$ 1.08bA	27.82 $\pm$ 1.01cA	8.95 $\pm$ 1.00abA
40–60	4.46 $\pm$ 0.18aA	3.14 $\pm$ 0.84dA	51.2 $\pm$ 1.82bA	15.62 $\pm$ 1.11cB	33.22 $\pm$ 2.02bcA	28.63 $\pm$ 1.44cdA	10.37 $\pm$ 0.87bcA
60–80	4.46 $\pm$ 0.04aA	2.44 $\pm$ 0.25 dB	50.4 $\pm$ 0.41bcA	14.78 $\pm$ 0.53bcB	34.80 $\pm$ 0.84cA	30.34 $\pm$ 1.39dA	9.79 $\pm$ 1.25bcA
80–100	4.41 $\pm$ 0.28aA	2.29 $\pm$ 0.40dA	48.9 $\pm$ 1.94cA	16.25 $\pm$ 1.23cB	34.82 $\pm$ 2.66cA	32.72 $\pm$ 2.06eA	11.02 $\pm$ 0.63cA
HA soils							
0–10	4.64 $\pm$ 0.14aA	61.81 $\pm$ 3.18aA	37.29 $\pm$ 10.15aB	43.55 $\pm$ 8.65aA	19.17 $\pm$ 1.80aB	8.59 $\pm$ 0.65aB	1.44 $\pm$ 0.14aB
10–20	4.73 $\pm$ 0.11abB	16.59 $\pm$ 4.18bA	34.97 $\pm$ 10.19bB	43.97 $\pm$ 7.83abA	21.06 $\pm$ 2.58abB	10.09 $\pm$ 0.67abB	1.48 $\pm$ 0.14abB
20–40	4.71 $\pm$ 0.14abA	6.50 $\pm$ 1.31cA	33.55 $\pm$ 9.83bcB	43.55 $\pm$ 6.01bA	22.90 $\pm$ 3.83cbB	11.32 $\pm$ 0.57bB	1.60 $\pm$ 0.05abB
40–60	4.88 $\pm$ 0.17abB	4.21 $\pm$ 0.68cA	33.34 $\pm$ 9.72cB	42.81 $\pm$ 5.82cA	23.85 $\pm$ 3.94cbB	13.96 $\pm$ 0.83cB	1.72 $\pm$ 0.08bcB
60–80	4.89 $\pm$ 0.17abB	3.83 $\pm$ 0.43cA	32.62 $\pm$ 9.74cB	41.31 $\pm$ 3.75dA	26.07 $\pm$ 6.19cdA	16.90 $\pm$ 1.79 dB	1.90 $\pm$ 0.24cB
80–100	4.93 $\pm$ 0.13bB	3.13 $\pm$ 0.51cA	30.27 $\pm$ 11.34 dB	42.31 $\pm$ 6.02dA	27.42 $\pm$ 5.39dA	18.09 $\pm$ 2.98 dB	2.34 $\pm$ 0.15 dB

Lowercase letters following the data in the same column indicate significant difference across depths within one soil type; capital letters show significant difference between two soil types at the same depth. The significance level was set at 0.05

leachate HIX, and leachate AI) across all depths were compared with analysis of variance (ANOVA) in a randomized block design. Soil depth was treated as fixed effect and sampling point (i.e., block, three blocks in each soil type) was included as the random factor. The least significant difference (LSD) method was applied for means comparisons in all ANOVA. Soil properties for two different soil types at the same depth were compared with a *t* test for independent samples. The Pearson correlation coefficients (*r* value) were calculated for total retention, hydrophobic retention, leachate AI, and leachate HIX. Multiple linear regressions were performed to assess the relationship between individual traits of single leaching (dependent variables: total retention, hydrophobic retention, leachate AI, and leachate HIX) and multiple soil properties (independent variables: soil pH, SOC, sand, silt, clay, Fe<sub>d</sub>, Al<sub>d</sub>, Fe<sub>o</sub>, Al<sub>o</sub>, Fe<sub>d</sub>-Fe<sub>o</sub>, HEOC quantity, HEOC AI, HEOC HIX). The stepwise method was used to select significant independent variables.

All statistical analyses were performed using SPSS (SPSS 20 for Windows, SPSS Inc. Chicago, IL, USA). Relative proportion of DOC input, DOC retention, and SOC of each soil horizon during sequential leaching were calculated as individual value of each horizon divided by the sum of six (for 0–100-cm FC soil) or five (for 10–100-cm HA soil) horizons.

**Data availability** The datasets generated and/or analyzed during the current study are available in the figshare repository (Si 2018; <https://figshare.com/s/3b4864e304ef0bed3fec>; <https://doi.org/10.6084/m9.figshare.6632942>). The dataset was not peer-reviewed.

## 3 Results

### 3.1 Soil properties

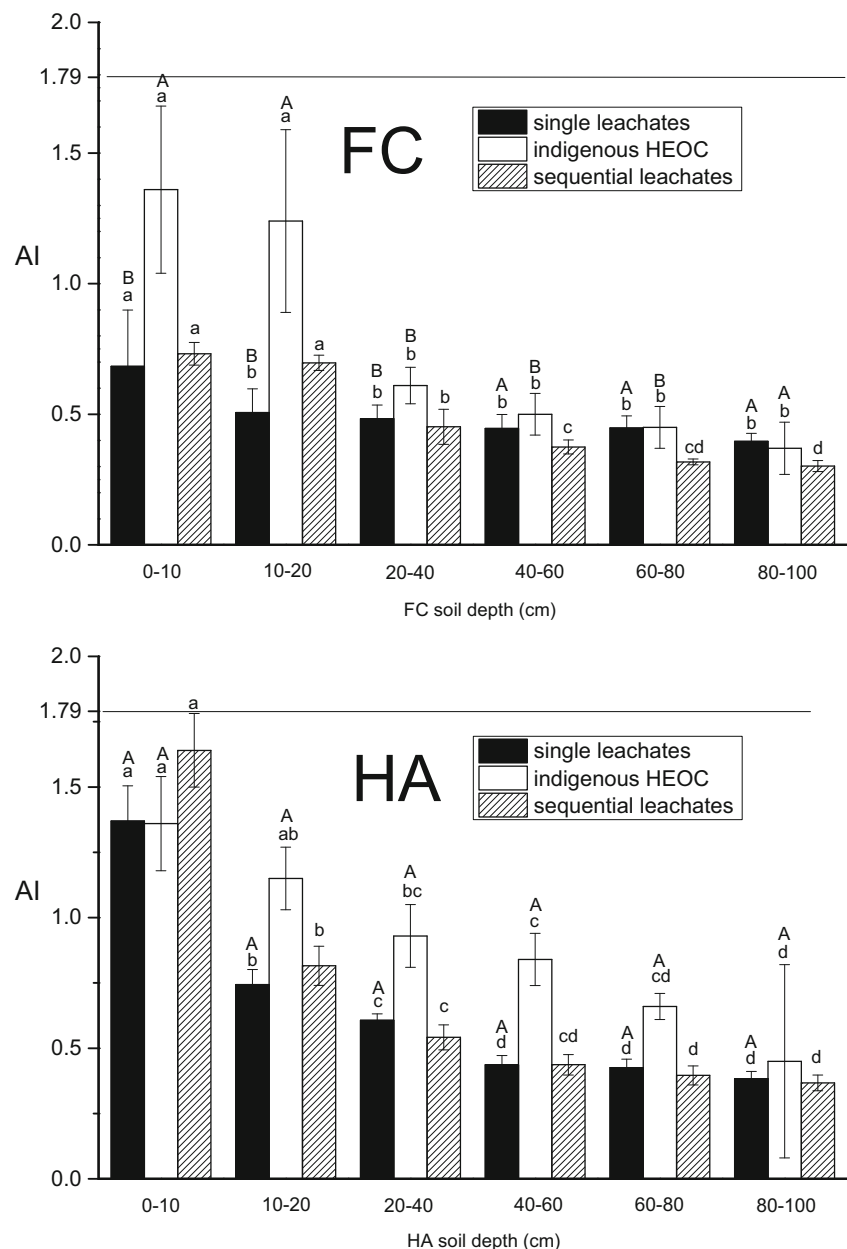
Between the two soil types, four out of six layers differed significantly in pH (Table 1). FC soils were more acidic and had significantly higher Fe<sub>d</sub>-Fe<sub>o</sub>, Al<sub>d</sub>, sand, and clay contents but significantly lower silt contents than HA. SOC content decreased with depth for both soil types, and HA soils had higher SOC content than FC soils (Table 1). HEOC of all FC soil layers, except for the 10–20-cm segments, had lower AI values than those of HA. HEOC of FC soils had higher HIX than those of HA soils (Figs. 1 and 2, blank columns).

### 3.2 DOC retention

#### 3.2.1 Single leaching

With single leaching, total retention and hydrophobic retention increased with depth within each soil type (Fig. 4). Different from all the other soil segments, the 0–10-cm HA soils showed negative total retention and negative hydrophobic retention (Fig. 4, HA). FC soils retained more DOC than HA soils (total retention 160.8 vs. 78.3 mg C kg<sup>-1</sup> soil, hydrophobic retention 148.7 vs. 76.6 mg C kg<sup>-1</sup> soil). For both soil types, total retention was significantly positively related to hydrophobic retention, but significantly negatively related to leachate HIX (Table 2).

**Fig. 2** AI (special UV absorption) of leachates from soil cores (black columns for single leaching and shadow columns for sequential leaching) and AI of HEOC (blank columns) for Ferralic Cambisols (FC) and Humic Alfisols (HA) soils. Error bars represent standard deviation ( $n = 3$ ). Lowercase letters over the same type of columns denote significant difference between horizons within each single Figure. Capital letters show significant difference between FC soils and HA soils, and only soil horizons of the same depth range are compared. The significance level was set at 0.05. Horizontal lines at AI = 1.79 indicate the AI of the standard DOC input



### 3.2.2 Sequential leaching

With sequential leaching, the quantity of DOC input to various horizons decreased with depth for each soil type (Fig. 5). Total retention and hydrophobic retention in FC soil decreased with depth (Fig. 4, FC). A sixfold higher amount of DOC input with sequential leaching made the 0–10-cm HA horizon release 17 times more DOC compared to single leaching, while retention over 10–100-cm HA soil layers was positive throughout but decreased along the soil profile (Fig. 4, HA). On average, FC soils retained more DOC than HA soils (total retention 168.2 vs. 153.2 mg C kg<sup>-1</sup> soil, hydrophobic retention 142.8 vs. 133.7 mg C kg<sup>-1</sup> soil).

### 3.3 UV absorption

Movement through soils reduced AI of DOC solutions (Fig. 2). In single leaching, leachates from deeper soil layers had lower AI than those from upper soil layers, and AI of leachates from 0 to 40-cm HA soils were significantly higher than those from 0 to 40-cm FC soils (Fig. 2, FC). In sequential leaching, by the time the percolating solutions reached the 60-cm depth, their AI had dropped significantly compared with that of the standard DOC, whereas the AI of the solution percolating through the 60–100-cm layers only slightly decreased. Notably, leachates from the 0–10-cm HA soil layers had the highest AI (1.6) but declined sharply (0.8) after they passed through 10–20-cm soils (Fig. 2, HA).

**Table 2** The Pearson correlation coefficients between total retention, hydrophobic retention, leachate AI, and leachate HIX. Only data from single leaching were used. For Ferralic Cambisols (FC) soils, data from all six soil layers were included and  $n = 18$ ; for Humic Alfisols (HA) soils, only data from five layers within 10–100 cm were included and  $n = 15$ ; the top layer was excluded due to the net release of DOC

	Total retention	Hydrophobic retention	Leachate AI	Leachate HIX
<b>FC soils</b>				
Total retention	1			
Hydrophobic retention	0.961**	1		
Leachate AI	-0.401	-0.385	1	
Leachate HIX	-0.774**	-0.724**	0.693**	1
<b>HA soils</b>				
Total retention	1			
Hydrophobic retention	0.984**	1		
Leachate AI	-0.967**	-0.969**	1	
Leachate HIX	-0.901**	-0.896**	0.898**	1

\*\* $p < 0.01$

### 3.4 Fluorescence spectroscopic analysis

As shown in Fig. 3, leachates always had lower HIX than incoming solutions. For each soil type in single leaching (Fig. 3 FC), the HIX of leachates of the four layers at the 0–60-cm depth differed significantly, whereas that of the leachates out of the three layers at the 40–100-cm depth showed no significant difference. Leachates from the 40–100-cm HA soil layers had significantly higher HIX than those from the 40–100-cm FC soil layers.

For each soil type in sequential leaching (Fig. 3, HA), HIX of leachates from six layers significantly differed from each other. Leachates from HA soils had slightly lower HIX than those from FC soils.

### 3.5 Multiple linear regression

To isolate the effect of soil conditions on leaching dynamics, only data from single leaching was used to carry out the regression analysis. Data about 0–10-cm HA soils was excluded due to net release. DOC movement through FC soils was predominantly influenced by SOC content, which was the only significant variable predicting total and hydrophobic retention. DOC movement through 10–100-cm HA soils was mainly affected by HEOC AI or HEOC HIX (see relative contribution, Table 3).

## 4 Discussion

### 4.1 Single leaching

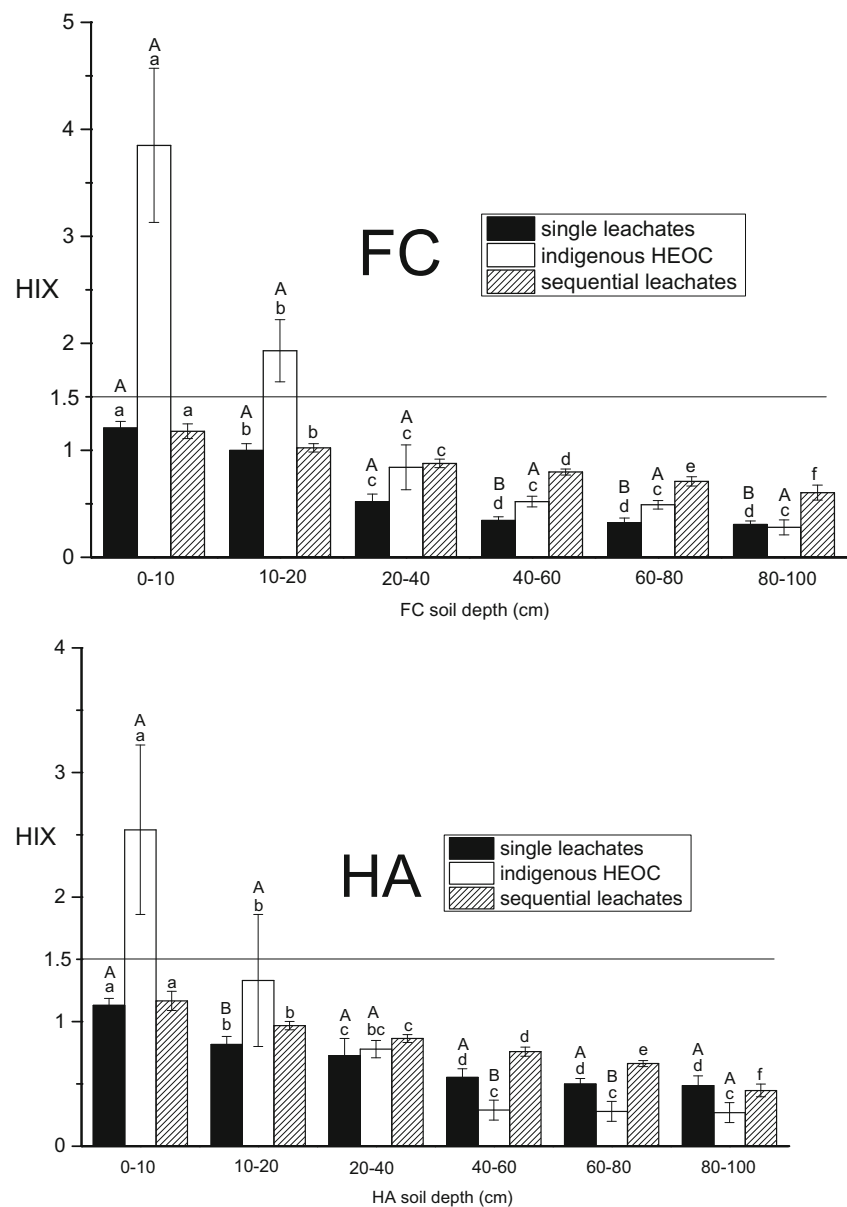
By single leaching, we demonstrated that within a soil type, deeper soils had higher retentive capacity than upper soils.

Lower SOC concentrations of deep versus surface soils may mean deep soils are further from carbon saturation than surface soils. Hydrophobic fraction accounted for the majority of total retention (Fig. 4) and they were significantly positively correlated (Table 2), consistent with the findings of Kaiser and Zech (1997). Aromatic substances, especially polycyclic aromatic compounds, are highly conjugated and have active  $\pi$  electrons that readily react with Fe/Al oxides and hydroxides (Weishaar et al. 2003). These substances are highly reactive and are preferentially retained at the soil surface. The lower AI of leachates out of deeper soils thus means more retention. HIX can indicate the percentage of microbe-processed organic carbon in solutions (Zsolnay et al. 1999). Lower HIX of leachates out of 40–100-cm soils meant these soils released less native SOC and retained more incoming DOC than 0–40-cm soils (significantly negative correlation between leachate HIX and total retention, Table 2).

SOC of FC soils greatly masked the effect of soil texture and Fe/Al mineralogy, because SOC is the only variable incorporated in the model to predict total retention or hydrophobic retention by FC soils, as seen in Table 3. FC soils were collected from a natural *C. carlesii* forest, whose SOC is structurally similar to our standard DOC and has been accumulated for hundreds of years, which makes sense as this litter source has been contributing to SOC formation for hundreds of years, perhaps explaining SOC effects on retention patterns (Table 3). Lack of SOC might result in similar retention, as the 60–80-cm and 80–100-cm FC soil layers showed similar and overall strong sorptive capacities with minimal SOC coverage, despite significant difference in their  $Fe_d$ - $Fe_o$  contents, consistent with Scott and Rothstein (2014). Dynamic exchange between incoming DOC and outer sphere SOC could be seen from the FC leachate AI, which was influenced by HEOC AI (Table 3). This implies that outer sphere SOC was displaced into leachates. Moreover, leachate AI was generally lower than HEOC AI (Fig. 2, FC), suggesting that higher aromatic substances were more competitive for soil surfaces. The amount of displaced SOC could be estimated by leachate HIX, which was positively controlled by SOC content (Table 3), suggesting that larger SOC content was associated with larger releases of SOC.

HA soils were collected from a natural *C. fargesii* stand, whose SOC nature should be different from that of the *C. carlesii* litter used to produce the standard DOC. As expected, we saw greater interaction between the standard DOC and outer sphere HA SOC, with stronger desorption and consequent less retention compared to FC soils. The strongest desorption due to the highest SOC content led to net DOC release from the top 10-cm HA soils. This provided unambiguous evidence for dynamic exchange and competition between incoming DOC and native SOC for soil surfaces. Displacement of native

**Fig. 3** Humification index (HIX) of leachates from soil cores (black columns for single leaching and shadow columns for sequential leaching) and HIX of hot water-extractable soil organic carbon (HEOC) (blank columns) for Ferralic Cambisols (FC) and Humic Alfisols (HA) soils. Error bars represent standard deviation ( $n = 3$ ). Lowercase letters over the same type of columns denote significant difference between horizons within each single figure. Capital letters show significant difference between FC soils and HA soils, and only soil horizons of the same depth range are compared. The significance level was set at 0.05. Horizontal lines at HIX = 1.50 indicate the HIX of the standard DOC input



SOC from other HA soil layers could be deduced from (i) leachate AI from the top 20-cm HA soils which was higher than leachate AI of the top 20-cm FC soils, despite the two kinds of soils having similar HEOC AI, and (ii) although HEOC HIX of 40–100-cm HA soils was less than or comparable to that of 40–100-cm FC soils, leachates out of the former soils showed significantly higher HIX than leachates out of the latter soils, indicating the former released much more microbe-processed SOC. For HA, outer sphere SOC therefore regulated DOC movement to a greater extent than in FC soils. This is in line with the fact that HEOC HIX and/or HEOC AI were included in the regression models predicting total retention, hydrophobic retention, and leachate HIX by 10–100-cm HA soils (Table 3).

## 4.2 Sequential leaching

In contrast to single leaching, deeper soils retained less DOC than upper soils with sequential leaching (except for the 0–10-cm HA soil horizon). As we have already demonstrated that deeper soils had stronger retentive capacities than upper soils, the retentive pattern for sequential leaching was clearly attributed to the changes in quantity and sorptive quality of incoming DOC.

Although decreasing with depth, DOC input to 0–80-cm FC and 0–100-cm HA soils for sequential leaching was still more than DOC input to these soils for single leaching (Fig. 5). Nevertheless, the 10–80-cm FC and the 60–100-cm HA soil layers retained less DOC with sequential leaching versus single leaching (Fig. 4). This suggests that reduced



**Table 3** Multiple linear regression model to assess the relationship between individual leaching traits (dependent variables, including total retention, hydrophobic retention, leachate AI, and leachate HIX) and soil properties (independent variables, including soil pH, SOC, sand, silt, clay,

$Fe_d$ ,  $Al_d$ ,  $Fe_o$ ,  $Al_o$ ,  $Fe_d-Fe_o$ , HEOC quantity, HEOC AI, HEOC HIX). The stepwise method was used to select significant independent variables. VIF (variation inflation factor) of selected variables were all below 7.2

	Adjusted $R^2$ (%)	Relative contribution of independent variables
FC soils (of 0–100-cm depth range)		
Total retention = $-1.573 \times SOC + 176.031$	62.1	SOC:100%
Hydrophobic retention = $-1.104 \times SOC + 159.390$	49.3	SOC:100%
Leachate AI = $0.492 \times HEOC\ AI - 0.003 \times sand - 0.032\ Al_d + 1.977$	85.8	HEOC AI: 56.9%, sand: 26.6%, $Al_d$ : 16.5%
Leachate HIX = $0.018 \times SOC + 0.004 \times sand - 1.783$	96.8	SOC: 49.1%, sand: 50.9%
HA soils (of 10–100-cm depth range)		
Total retention = $-29.447 \times HEOC\ AI + 2.579 \times Fe_d-Fe_o + 90.371$	81.3	HEOC AI: 49.2%, $Fe_d-Fe_o$ : 50.8%
Hydrophobic retention = $-14.492 \times HEOC\ HIX + 1.488 \times Fe_d-Fe_o + 0.123 \times silt - 22.812 \times HEOC\ AI + 50.507$	90.2	HEOC HIX: 28.7%, $Fe_d-Fe_o$ : 33.6%, silt: 9.0%, HEOC AI: 28.7%
Leachate AI = $0.015 \times SOC - 0.021 \times Fe_d + 0.735$	85.7	SOC: 54.0%, $Fe_d$ : 46.0%
Leachate HIX = $0.187 \times HEOC\ HIX + 0.218 \times HEOC\ AI + 0.331$	81.6	HEOC HIX: 54.8%, HEOC AI: 45.2%

quantities of DOC input do not necessarily result in less retention. Declining affinity of DOC input along soil depth was thus the key factor explaining the retentive pattern of sequential leaching.

Reduction in the affinity of incoming DOC to deeper soils for soil surfaces was attributed to (i) the quantity and percentage of hydrophobic compounds of input solutions decreased with depth (Guggenberger and Kaiser 2003; Kaiser and Kalbitz 2012). (ii) DOC input to deeper soils had lower AI (Fig. 2, shadow columns). By the time the percolating solution reached the 60-cm depth, AI had declined drastically compared to the standard DOC (Fig. 2). Subsequently, the 60–100-cm soils only slightly decreased the AI of the percolating solution, consistent with their small retention. (iii) Relative more weaker-binding compounds were input to deeper soils. Leachates from sequential leaching generally had higher HIX than those from single leaching. This suggested that leachates out of upper soils included relative more native fraction, which showed weaker affinity for soil surfaces and accumulated in the leachate solutions.

Despite six times greater DOC input than that of single leaching, the total retention by top 10-cm FC soils increased only by a factor of 4.2. This might be due to the already high SOC concentration and the chemical similarity between the standard DOC and native FC SOC. This implies that retention amount should not be considered linear with DOC input as assumed by some models (Tipping et al. 2012). However, sequential leaching widened the gap in SOC between the two uppermost FC horizons. The relative DOC retention by 0–10-cm FC soil layer was slightly higher than its relative SOC content (higher blank columns vs. shorter gray columns for this soil horizon in Fig. 5, FC). In contrast, the 10–20-cm horizon retained much less DOC compared to its relative SOC content. Under field conditions, rapid

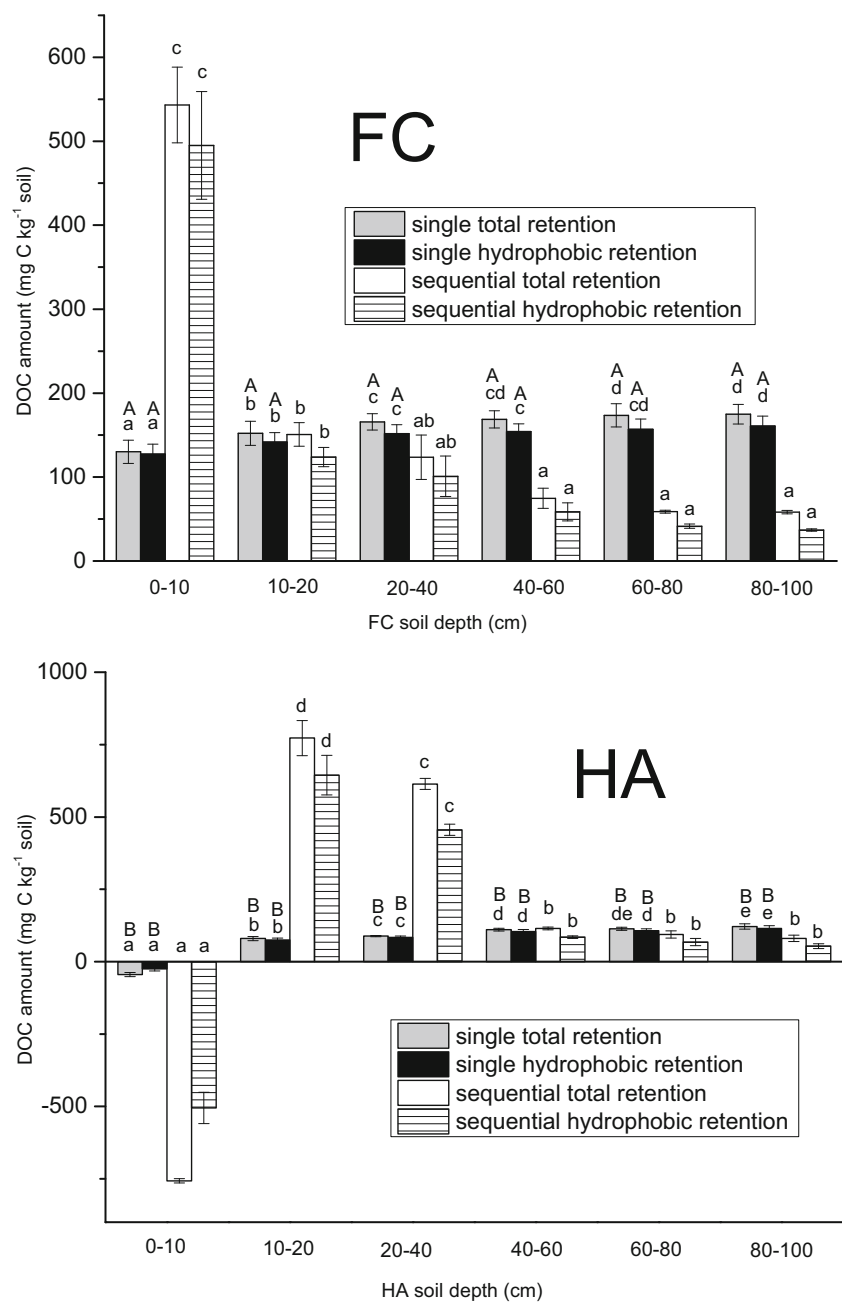
degradation of SOC in the top 10-cm soils and migration of soil solution with rainfall, as well as carbon input from fine roots, would further transport organic carbon to 10–20-cm soils. The 20–100-cm FC soils also retained higher percentages of DOC than their native SOC content (Fig. 5, FC), probably because under field conditions these soils had little chance to receive so much DOC input as in our experiment and therefore sequestered lower C.

The sixfold greater DOC input to the 0–10-cm HA soil layers increased the DOC release by a factor of 17 (Fig. 4). Obviously, this DOC release was caused not only due to scouring by the incoming DOC, but more importantly by desorption of native SOC. If scouring alone, six times of input would have produced at most a sixfold increase in the DOC release. Moreover, the AI of the leachates (1.6) from this soil horizon was highest of all leachates, exceeding HEOC AI and leachate AI of single leaching (Fig. 2). This implies that desorption-induced release of native SOC in sequential leaching was even more intensive than hot water extraction, and more aromatics were released. This caused much DOC input to the 10–40-cm HA horizons, and consequently, these soils retained the most DOC, with the leachate AI dropping sharply from 1.6 to 0.5 (Fig. 2, HA). Especially, the relative DOC retention by the 20–40-cm HA horizon was much greater than the relative SOC content in this layer (Fig. 5, HA). Due to the strong retention, the 40–100-cm soil layers retained lower percentages of DOC relative to SOC content.

### 4.3 Single leaching vs. sequential leaching

Despite the sorptive capacity of deeper soils was limited, the organic carbon sequestration was more in sequential leaching than in single leaching for each soil type, because (i) sequential leaching retained more hydrophilic fraction; (ii) leachates

**Fig. 4** Amount of HEOC and DOC retained by Ferralic Cambisols (FC) and Humic Alfisols (HA) soils. Error bars represent standard deviation ( $n = 3$ ). Lowercase letters over the same type of columns denote significant difference between horizons within each single figure. Capital letters show significant difference between FC and HA, and only soil horizons of the same depth range are compared. The significance level was set at 0.05. Single leaching means soil core was equally and individually supplied with 151 mL (averagely 31.6 mg C) standard solution. Sequential leaching refers to that 906 mL (averagely 189.5 mg C) standard DOC solution percolated through the 0–10-cm soils, followed by the leachate passing through the 10–20-cm layer, and so on until it penetrated the soil at 80–100-cm depth

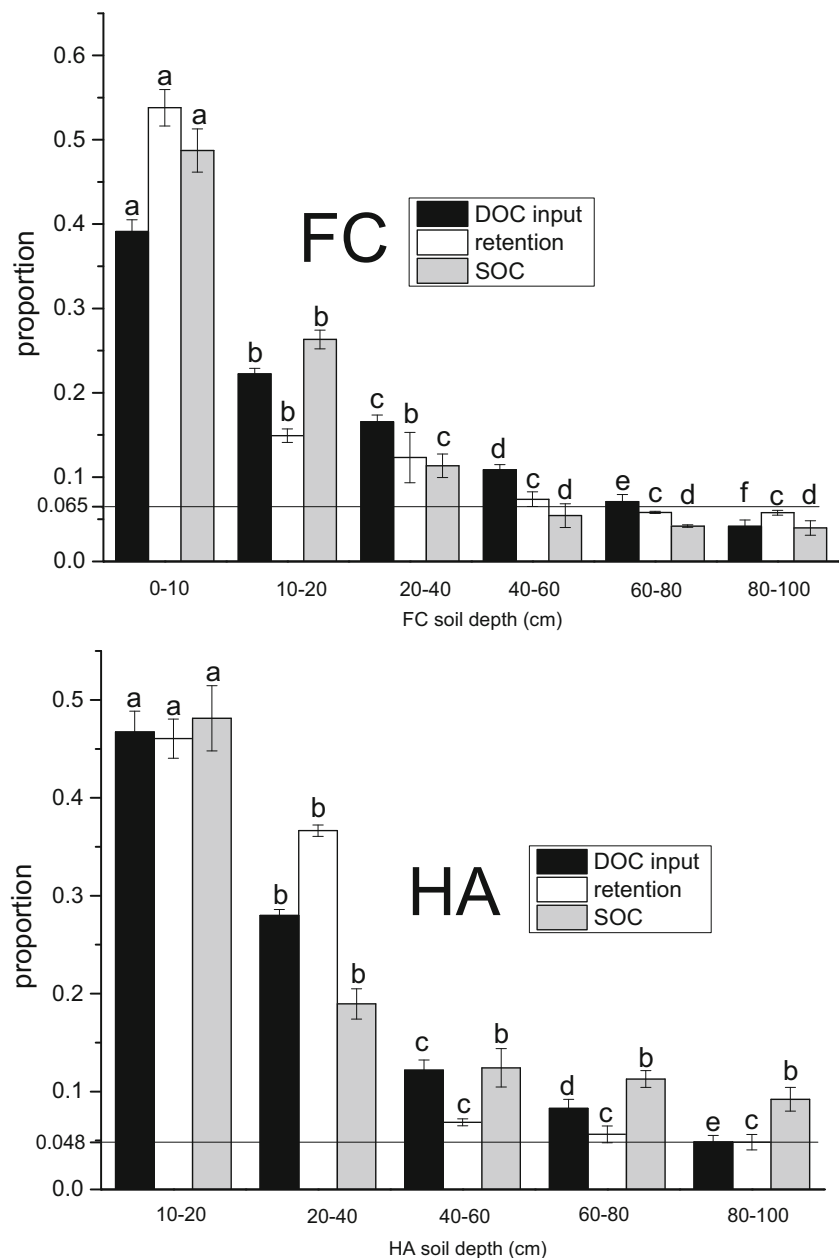


from the four 20–100-cm horizons in sequential leaching had lower AI than those from single leaching (Fig. 2, black columns vs. shadow columns) and therefore retained more weaker-binding fraction than single leaching; and (iii) the HIX of the percolating solution steadily decreased during sequential leaching, so some of desorbed native SOC from the upper soil layers was retained by deeper soil layers. This desorption-induced transfer of organic carbon from upper soils to deeper soils shows that with time weaker-binding organic carbon will be associated with deeper soils, which is a natural way to preserve SOC, in agreement with the cycling downwards model (Kaiser and Kalbitz 2012).

#### 4.4 Ecological significance

Our results do not reflect actual DOC dynamics of in situ soils for obvious reasons—we relied on modified soils and we applied DOC in a way that does not reflect actual hydrological processes. The sorptive capacity of soil surfaces for DOC shown in laboratory studies may not control the transport of DOC in soils if macropore fluxes dominate under field conditions. Soil macropores can facilitate DOC solution flow, either downward or lateral, and often serve as hotspots with high microbial activity, rapid organic matter mineralization and more soluble microbial products (Kuzyakov and

**Fig. 5** Proportion of DOC input, retention, and SOC of each soil horizon during sequential leaching for Ferralic Cambisols (FC) and Humic Alfisols (HA) soils. Error bars represent standard deviation ( $n = 3$ ). Horizontal lines indicate DOC input during single leaching. Proportions for FC soils were calculated as individual data of each horizon divided by sum of all six horizons; proportions for HA soils were calculated as individual data of each horizon divided by sum of five horizons between 10 and 100 cm. Lowercase letters over the same type of columns denote significant difference between horizons within one soil type



Blagodatskaya 2015). However, this study focused on the sorptive capacity of soil surfaces and reduced the influence of macropore by packing disturbed soils to PVC columns.

Traditional aboveground litter decomposition studies considered only the remaining recalcitrant particulate litter residue as contributing stable SOC and underestimated DOC fluxes to the mineral soil (Berg and Mcclaugherty 2008). Cotrufo et al. (2015) identified the importance of DOC leaching from litter followed by high microbial incorporation, i.e., the DOC-microbial pathway, for the formation of mineral-associated SOC at the early stage of litter decomposition. While they found no new SOC formation from recently decomposed litter in soils below 20-cm soil depth, our results highlighted that aboveground DOC input can actually alter SOC processes and

composition to the 100-cm depth. This is particularly important for forest conversion. DOC from *C. carlesii* litter would displace much native *C. fargesii* SOC of 0–10-cm depth range and move them downwards, which greatly alter the SOC nature of 10–40-cm soils and subsequent microbial community structure and activity. This desorption-induced transfer could explain why the oldest organic matter being located in the youngest part of the soil profile (the deep mineral subsoil) and why subsoil DOC is of microbial origin. Another possible fate of the displaced SOC from upper soil layers is the potential to be leached into aquatic environments, which is of course not good to SOC accumulation. Such desorption-induced SOC loss might explain rapid SOC loss during early stages of post-harvest stand growth (Chen et al. 2004; Chen et

al. 2010). While we focused on the role of DOC and SOC nature in controlling DOC adsorption/desorption in this study, other environmental variables may be equally important. A warmer atmosphere may lead to higher microbial activity and more microbial products and might promote SOC desorption and DOC migration to deeper soils (Allison et al. 2010). Nitrogen deposition could also affect DOC movement by altering litter decomposition rates and DOC input rates to soils (Knorr et al. 2005).

Although soil DOC dynamics has been incorporated into some mechanistic SOC models (Jason and Gregory 2001; Sanderman et al. 2008; Tipping et al. 2012), our results can still provide useful information to optimize such models. First, through single leaching, this study revealed that deeper soil horizons have stronger sorptive capacities than upper horizons. However, models, such as DyDOC (Tipping et al. 2012), do not include a sorptive capacity factor for different soil horizons. Secondly, a sixfold higher amount of DOC input to the 0–10-cm soils (both FC and HA) with sequential leaching did not result in six times the retention or release compared with single leaching, but Tipping et al. (2012) assumed there was a linear relationship between retention and DOC input. Thirdly, Sanderman et al. (2008) proposed a “potentially exchangeable or reactive soil carbon pool” responsible for the composition of soil DOC. Beyond this concept, our results further demonstrate that the size of this pool is highly dependent on the nature of the incoming DOC.

## 5 Conclusion

This study highlighted the importance of the native SOC and incoming DOC in regulating DOC leaching dynamics. By single leaching, we demonstrated deeper soil layers had stronger sorptive capacities than upper soil layers, and SOC or the HEOC was the most important factors regulating DOC movement through soils. With sequential leaching, however, changing DOC quantity and chemistry with declining affinity for soil surfaces increasingly restricted carbon sequestration in deeper soils. Nevertheless, desorption-induced transfer of organic carbon from upper to deeper soils ensured that sequential leaching always retained more organic carbon than single leaching, and DOC vertical movement greatly altered SOC formation and loss along the top soil profile. Clearly, our results demonstrate a need for soil biogeochemistry and hydrology models to more accurately address DOC and SOC dynamics and interactions.

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## Compliance with ethical standards

**Conflict of interest** The authors declare that they have no conflict of interest.

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