

## RESEARCH ARTICLE

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## Key Points:

- Particulate organic carbon half-lives were low compared to the dissolved fraction with only  $17 \pm 3$  days
- High particulate organic carbon loss rate despite sustained concentrations in inland waters requires replenishment of the particles
- Transformation of terrestrial dissolved into particulate organic carbon might sustain rapid loss via particle formation

## Supporting Information:

- Supporting Information S1
- Data Set S1

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## Organic Carbon Processing During Transport Through Boreal Inland Waters: Particles as Important Sites

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**Abstract** The degradation and transformation of organic carbon (C) in inland waters result in significant CO<sub>2</sub> emissions from inland waters. Even though most of the C in inland waters occurs as dissolved organic carbon (DOC), studies on particulate organic carbon (POC) and how it influences the overall reactivity of organic C in transport are still scarce. We sampled 30 aquatic ecosystems following an aquatic continuum including peat surface waters, streams, rivers, and lakes. We report DOC and POC degradation rates, relate degradation patterns to environmental data across these systems, and present qualitative changes in dissolved organic matter and particulate organic matter during degradation. Microbial degradation rates of POC were approximately 15 times higher compared to degradation of DOC, with POC half-lives of only  $17 \pm 3$  (mean  $\pm$  SE) days across all sampled aquatic ecosystems. Rapid POC decay was accompanied by a shift in particulate C:N ratios, whereas dissolved organic matter composition did not change at the time scale of incubations. The faster degradation of the POC implies a constant replenishment to sustain natural POC concentrations. We suggest that degradation of organic matter transported through the inland water continuum might occur to a large extent via transition of DOC into more rapidly cycling POC in nature, for example, triggered by light. In this way, particles would be a dominant pool of organic C processing across the boreal aquatic continuum, partially sustained by replenishment via flocculation of DOC.

## 1. Introduction

Inland waters receive large amounts of terrestrial carbon (C) and approximately half of this C is lost along the aquatic continuum before being discharged into oceans via rivers (Aufdenkampe et al., 2011; Tranvik et al., 2009; Ward et al., 2017). In aquatic systems, the C content of organic matter is often categorized as dissolved organic carbon (DOC) or particulate organic carbon (POC) based on filtering with filter pore sizes ranging from  $\sim 0.1$  to  $\sim 0.7 \mu\text{m}$  (Thurman, 1985). To better understand the biogeochemical processes in inland waters, the sources, forms, and fates of these groups of organic C must be considered. This need is particularly true for POC in the water column, which, despite making up about half of the total flux of organic C in world rivers (Meybeck, 1982; Schlesinger & Melack, 1981) and being the terrigenous component most directly incorporated into coastal marine sediments (Keil et al., 1998; Prahl et al., 1994), has been poorly studied relative to DOC.

So far, studies on POC imported to inland waters have focused on processes regulating the transport of POC from land to the aquatic system and their environmental controls on POC concentrations. The simultaneous input of DOC and POC during storm events has been studied intensively (Dhillon & Inamdar, 2013; Jeong et al., 2012; Wiegner et al., 2009). These studies emphasize that large amounts of POC are flushed into inland waters during high-intensity storm events and that those inputs can even exceed the input of DOC by 6 to 8 times during storms (Dhillon & Inamdar, 2013). A recent analysis of streams and rivers in the coterminous United States reveals that suspended sediment and chlorophyll-*a* were the most important factors explaining the variability in POC concentrations (Yang et al., 2016). However, there was still a high variation ( $\sim 60\%$ ) that could not be explained by the investigated environmental factors highlighting the need to better constrain the variability and transformation processes of POC in aquatic systems.

While in transit, the composition of organic matter can be modified by several processes, including photo-oxidation, respiration, leaching, flocculation, sorption, and settling (Eckard et al., 2007; Einarsdottir et al., 2017; Galy et al., 2011; Spencer et al., 2010; von Wachenfeldt & Tranvik, 2008). Most studies on

organic C processing by microbes focused on the dissolved fraction as it is the largest contributor to the organic C pool in inland waters (Thurman, 1985). In the past years, a major effort has been set in disentangling the factors controlling molecular composition and reactivity of dissolved organic matter (DOM) along inland waters. Kellerman et al. (2014) identified mean annual temperature and precipitation as well as water retention time (WRT) as major controlling factors of DOM molecular composition in lakes. In addition, the microbial degradability of DOM appears to decrease with longer WRT, suggesting an “aging process” of DOM that points toward a crucial role of intrinsic molecular characteristics of the DOM pool for transformation processes in inland waters (Marín-Spiotta et al., 2014). Despite the potential relevance of POC for global C fluxes, substantially contributing to the total organic C (TOC) export from inland waters to the ocean (Lal, 2003; Ludwig & Probst, 1996), there are few studies exploring the transformation of suspended POC in aquatic systems along the inland water continuum. So far, earlier studies mainly focused on specific types of particles such as algae (e.g., Grossart & Ploug, 2001) or pollen (Wurzbacher et al., 2014) and suspended POC quantities, qualities, and related turnover were less studied. This is especially interesting in regions with high terrestrial influence such as boreal landscapes, where DOC, POC, and sediments are dominated by terrestrial organic C (Kothawala et al., 2014; von Wachenfeldt & Tranvik, 2008).

Therefore, we aimed to explore if and how POC and DOC reactivity changes along the boreal aquatic continuum and if these changes are related to the WRT. We tested whether POC reactivity decreases with increasing WRT similar to the patterns observed for DOC (Catalán et al., 2016). To explore this question, we incubated water from freshwaters representing different WRTs, characterized the natural DOM and particulate organic matter (POM) composition, and investigated changes after microbial decomposition in the DOM and POM pools over the course of 1 week.

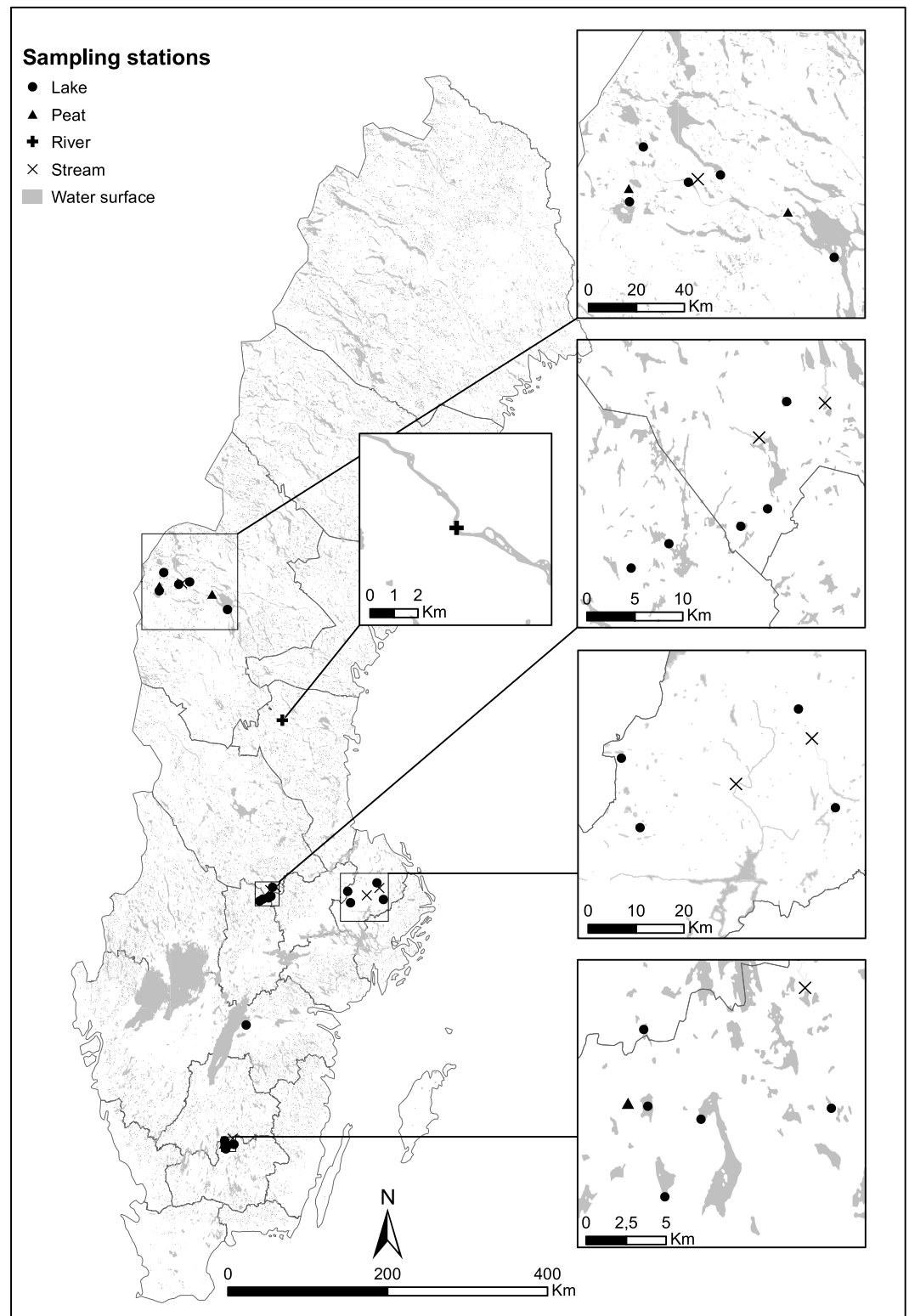
## 2. Materials and Methods

### 2.1. Sampling

A total of 30 freshwater systems were sampled in four regions in Sweden in 2016: Småland (28–29 August), Jämtland (31 August to 2 September), Bergslagen (4 September), and Uppland (6 September; Figure 1). We sampled water from three wetland peats, seven streams and rivers (hereafter referred to as *running waters*), and 20 lakes, covering a wide range of boreal aquatic systems (Table 1). We selected a gradient of different WRTs: first, surface water from peats, for which WRT was considered as zero, representing headwater peatlands and thus water prior to entering surface waters and being transported through the inland water continuum; running waters as a second group with very low WRT (calculated as explained below); and finally, lakes with different WRTs retrieved from previous studies (Table 1). Specifically, WRT in lakes was retrieved from the literature and is the ratio between the volume of a lake ( $V$ ;  $\text{m}^3$ ) and the volumetric flow rate ( $Q$ ;  $\text{m}^3/\text{s}$ ). This gradient represents the aquatic continuum from terrestrial sites to lakes with longer WRTs where it is expected that the organic matter had been subjected to a longer period of biogeochemical transformation. At each site, we took a water sample ( $\sim 25$  L) and measured temperature, oxygen and conductivity (HQ40d, Hach, Loveland, Colorado, USA), and pH (Hanna HI991300; Woonsocket, Rhode Island, USA) directly in the systems. Peat water samples were taken from standing surface waters in the wetland peat, running water samples from the surface, and the lakes were sampled at 0.5 m depth as close as possible to their outlet. Subsequently, the samples were transported back to the laboratory and further processed within 1 to 2 days. The 1- to 2-day storage can lead to some change in the particulate pool, which might have led to a slight underestimation of the true degradation rates determined by the incubations (see section 2.3). The following parameters were measured in the original samples: DOC, total phosphorus (TP), total nitrogen (TN), chlorophyll-*a*, absorbance and fluorescence of DOM, anions ( $\text{F}^-$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$ ), cations ( $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$ ) and organic acids (acetate and glycolate), high-resolution mass spectrometry of DOM, POM dry weight, POC, particulate nitrogen (PN) and the carbon to nitrogen ratio (C:N) of particulates, bacterial abundance and production. An overview of all data is provided in the supporting information.

### 2.2. Determination of Stream WRT

Running waters' WRTs were calculated based on discharge and catchment stream length. The discharge of the running waters was measured during the sampling campaign if possible. For the small headwater stream (8; Hisshult), discharge ( $Q$ ;  $\text{L/s}$ ) and water velocity ( $v$ ;  $\text{m/s}$ ) were determined through the slug addition of NaCl (Gordon et al., 2004). In the case of sampling sites 16, 22, and 24,  $Q$  and mean velocity were obtained using a



**Figure 1.** Map of Sweden with sampling sites. © Lantmäteriet Gävle: Permission i2014/00601.

portable in-stream current meter ( $\mu$ P-TAD, Höntzsch Instruments, Waiblingen, Germany). For sites 17, 26, and 28, discharge was not available due to the high-flow conditions or lost data. In these cases, data downloaded from the interface HYPE from the Swedish Meteorological and Hydrological Institute

**Table 1**
*Environmental Parameters of All Sites Included in This Study*

ID	Sampling site	Aquatic system	GPS coordinates	WRT (years)	Water temp (°C)	pH	Conductivity (μS/cm)	POC (mg/L)	DOC (mg/L)	Color <sub>420</sub> (m <sup>-1</sup> )	Chla (μg/L)	TP (μg/L)	TN (mg/L)
1	Vättern	Lake	58°28'0 N 14°55'2 E	70	16.8	8.3	147	0.41	3.0	0.2	0.36	4.4	0.56
2	Skärshultsjön	Lake	57°10'1 N 14°31'2 E	0.121	18.7	6.5	50	0.90	15.5	5.3	1.54	15.7	0.58
3	Förhultasjön	Lake	57°07'2 N 14°30'4 E	0.719	18.7	7.3	57	0.65	11.5	2.0	2.10	8.4	0.45
4	Peat Småland	Peat	57°07'2 N 14°29'3 E	0	12.3	4.8	44	4.88	30.4	11.8	3.83	35.2	0.97
5	Fiolen	Lake	57°04'7 N 14°32'5 E	6.5	18.3	7.1	50	0.58	7.4	1.0	2.71	11.4	0.43
6	Stråken	Lake	57°07'0 N 14°34'5 E	10.7	18.2	6.8	61	0.64	10.5	2.0	2.92	11.4	0.44
7	Klintsjön	Lake	57°07'5 N 14°42'4 E	4.7	18.4	5.9	38	0.33	4.2	0.6	0.98	5.5	0.25
8	Hissshult	Stream	57°11'2 N 14°40'2 E	0.000031	16.8	5.9	100	5.62	24.2	10.0	29.15	46.3	1.15
9	Storsjön	Lake	63°07'4 N 14°19'0 E	2	14.5	7.5	44	0.27	3.4	0.5	0.68	3.1	0.21
10	Peat near Storsjön	Peat	63°17'3 N 13°55'3 E	0	12.0	6.2	64	21.38	30.5	9.9	3.70	179.7	2.84
11	Ännsjön	Lake	63°18'3 N 12°36'2 E	1.1	12.9	7.8	56	0.59	3.6	0.7	0.54	6.8	0.19
12	Peat near Ännsjön	Peat	63°21'2 N 12°35'4 E	0	11.5	5.8	44	1.46	37.3	14.8	1.76	26.1	0.75
13	Häggsjön	Lake	63°30'5 N 12°41'5 E	0.032	13.4	7.3	37	0.17	4.5	0.9	0.61	3.1	0.17
14	Kallsjön	Lake	63°25'2 N 13°21'1 E	2.3	12.4	7.4	29	0.26	3.1	0.8	0.09	6.3	0.23
15	Åresjön	Lake	63°23'3 N 13°05'1 E	0.028	13.7	7.8	40	0.35	3.1	0.6	0.52	3.2	0.16
16	Mountainous stream	Stream	63°24'1 N 13°10'0 E	0.00087	12.5	7.7	32	0.22	2.7	0.6	0.19	1.1	0.11
17	Ljusnån	River	61°53'3 N 15°40'4 E	0.0030	15.3	7.8	31	0.29	6.8	2.3	1.20	5.0	0.21
18	Gäddtjärn	Lake	59°86'0 N 15°18'2 E	0.26	15.4	7.6	65	0.61	10.7	4.2	0.59	9.3	0.30
19	Svartjärn	Lake	59°53'2 N 15°15'2 E	0.04	14.6	6.0	71	1.65	15.2	6.6	1.30	13.8	0.28
20	Lilla Sångaren	Lake	59°54'0 N 15°23'2 E	1.18	15.7	7.3	47	0.40	6.5	1.6	0.96	6.0	0.26
21	Ljustjärn	Lake	59°55'3 N 15°26'5 E	4.3	16.7	7.3	22	0.48	3.3	0.5	0.59	7.6	0.23
22	Hedströmmen	River	59°58'5 N 15°24'3 E	0.0028	15.4	6.8	22	0.31	8.3	2.6	0.79	6.8	0.26
23	Oppsveten	Lake	60°01'0 N 15°28'0 E	0.43	16.1	6.7	27	0.51	16.1	6.1	2.41	8.1	0.41
24	Bjursjöbäcken	Stream	60°02'5 N 15°32'2 E	0.0136	14.3	7.1	37	0.48	12.6	3.9	0.96	12.2	0.39
25	Lötsjön	Lake	59°51'4 N 17°56'3 E	4.4	15.9	8.2	212	2.86	11.3	0.6	15.28	24.8	1.12
26	Sävjaån	Stream	59°59'3 N 17°51'4 E	0.0068	10.9	7.2	279	0.58	22.7	6.0	0.67	77.3	1.07
27	Stensjön	Lake	60°02'3 N 17°48'5 E	0.06	15.1	7.2	162	0.93	18.6	4.7	3.07	20.3	0.76
28	Fyrisån	River	59°54'4 N 17°33'3 E	0.0062	15.1	7.8	384	0.57	16.4	2.3	1.15	39.8	1.10
29	Siggeforasjön	Lake	59°58'4 N 17°09'1 E	0.49	16.7	7.2	44	0.51	15.5	4.1	0.81	9.3	0.51
30	Ramsjön	Lake	59°50'9 N 17°12'5 E	1.41	18.3	7.5	109	0.51	23.8	7.8	2.78	29.1	0.76

*Note.* ID = identification; WRT = water retention time; POC = particulate organic carbon; DOC = dissolved organic carbon; Chla = chlorophyll-*a*; TN = total nitrogen; TP = total phosphorus.

(<http://vattenwebb.smhi.se/modelarea/>; 14 November 2017) were used to obtain discharge during the month preceding the sampling date. For all sites, the total area of the catchment was also obtained from the HYPE database.

The WRT is defined as the length of the stream reach (*L*; m) divided by the water velocity (*v*). Accordingly, for site 8, with a known reach length and velocity, we directly obtained WRT as *L/v*. Then we obtained the length of the catchment upstream (*Lc*) assuming it had the height of an isosceles triangle with known area, obtaining similar RTT as Raymond et al. (2016) for similar stream orders. Then WRT was obtained as RTT/*Lc*.

### 2.3. Laboratory Incubations

The degradation of DOC and POC was determined during laboratory incubations. We incubated 1 L unfiltered water from each site in triplicates in acid-washed 1-L glass bottles (headspace of ca. 140 ml) for 7 days at 13 °C in the dark. The bottles were closed to the atmosphere during incubations, and oxygen concentrations did not fall below 2 mg/L in the bottles. This threshold is well above the limiting oxygen concentrations described for different bacterial species (Stolper et al., 2010, and references therein). An incubation time of 1 week has been shown to lead to significant changes in the DOC as well as POC pool for suspended particles (Richardson et al., 2013). The bottles were turned on a roller device (Shanks & Edmondson, 1989) at about 2 rpm to avoid the settling of particles and the establishment of gradients in the water. The water was sampled directly before (one mixed sample; same water that was also taken for the characterization of the original samples, see section 2.1) and after the incubation period (one sample each from triplicate

incubations) and subsequently filtered over precombusted GF/F filters (pore size  $\sim 0.7 \mu\text{m}$ ; Whatman), and DOC, fluorescence/absorbance of DOM, and DOM molecular composition via Orbitrap mass spectrometry were measured in the filtrate. POC and C:N were determined from the particles on the filter.

#### 2.4. DOC, TN, and TP

From each sampling site and each incubated bottle at the end of the incubation, DOC concentrations were analyzed on filtered water (precombusted GF/F, Whatman) and measured on a total carbon analyzer (Sievers M9 Laboratory Analyzer, GE Analytical Instruments, Boulder, Colorado, USA). The accuracy is given as  $<1\%$  relative standard deviation, and the precision is  $\pm 2\%$  (GE Analytical Instruments, Boulder, Colorado, USA). The concentrations of TN in the water were measured by a TN analyzer (Shimadzu TOC-L/TNM-L, Kyoto, Japan). TP concentrations were measured photometrically (UV/Vis Spectrometer Lambda 40; Perkin Elmer; Waltham, Massachusetts, USA) using the molybdenum-blue method (Menzel & Corwin, 1969) after oxidative hydrolysis with potassium persulfate in a slightly acidic solution at high temperature and high pressure in an autoclave.

#### 2.5. Chlorophyll-*a* Concentration

The suspended materials for the chlorophyll-*a* analyses were collected on glass microfiber filters (precombusted GF/F, Whatman) from duplicate original samples immediately after shaking and frozen at  $-20^\circ\text{C}$  after filtration. The chlorophyll-*a* was extracted from the filter by leaching with ethanol for 5 min at  $75^\circ\text{C}$ . Subsequently, the chlorophyll-*a* contents were determined with a spectrophotometer (UV/Vis Spectrometer Lambda 40; Perkin Elmer; Waltham, Massachusetts, USA), following standard guidelines (ISO10260, 1992; Jespersen & Christoffersen, 1987). A conversion factor of  $40 \mu\text{g C } (\mu\text{g chlorophyll-}a)^{-1}$  (Banse, 1977) was applied to roughly estimate the phytoplankton C content. This value was used to compare the contribution of phytoplankton C to the POC pool in the samples.

#### 2.6. Absorbance and Fluorescence of DOM

The absorbance and fluorescence of DOM were measured on filtered water (precombusted GF/F, Whatman). Absorbance spectra were measured from 200 to 600 nm, at 1-nm intervals using a Lambda 40 UV-visible spectrophotometer (Perkin Elmer, Waltham, USA). Samples were measured in a 1-cm quartz cuvette, and Milli-Q was used as blank. In samples with absorbance at 200 nm  $> 2$ , we used a 0.5-cm quartz cuvette. From these data, absorbance at 420 nm ( $\text{color}_{420}$ ) was retrieved and the specific UV absorbance (SUVA) at 254 nm, an indicator of the aromaticity of DOM, was calculated based on the absorbance at 254 nm divided by the respective DOC concentration (Weishaar et al., 2003). Excitation-emission matrices (EEMs) from lake water samples were obtained using a fluorescence spectrophotometer (SPEX FluoroMax-2, Horiba Jobin Yvon) with a 1-cm quartz cuvette (or 0.5 cm if absorbance at 200 nm  $> 2$ ). The excitation wavelengths spanned from 250 to 445 nm, with an increment of 5 nm, while emission wavelengths ranged from 300 to 600 nm at increments of 4 nm. Excitation and emission slit widths were set to 5 nm, and the integration time was 0.1 s. All EEMs were blank-subtracted using the EEM of a Milli-Q blank run on the same day, corrected for instrument biases and inner filter effects, and normalized to Raman units using the FDOMcorr toolbox for MATLAB (Murphy et al., 2010).

The EEMs were used to calculate three previously established indices: humification index (HIX = ratio of areas under the emission curve at 435–480 and 300–345 nm plus 435–480 nm at an excitation wavelength of 254 nm; Ohno, 2002), fluorescence index (FI = emission intensity at 470 nm divided with that of 520 nm at 370 nm excitation; Cory & McKnight, 2005), and freshness index ( $\beta:\alpha$  = ratio of emission intensity at 380 nm and maximum intensity between 420 and 435 nm at an excitation wavelength of 310 nm; Parlanti et al., 2000). The indices give information about the degree of humification (higher HIX = more humified material), source (a gradient from microbial [FI  $\approx 1.8$ ] to terrestrial [FI  $\approx 1.3$ ]), and biological origin (high freshness index = larger contribution of freshly produced DOM), respectively.

#### 2.7. Ion Chromatography (Anions, Cations, and Organic Acids)

The samples for measurements of anions, cations, and organic acids were filtered over  $0.2\text{-}\mu\text{m}$  Acrodisc Supor hydrophilic polyethersulfone membranes (Pall Laboratory, Port Washington, New York, USA) and stored frozen. Anions ( $\text{F}^-$ ,  $\text{Cl}^-$ , and  $\text{SO}_4^{2-}$ ), cations ( $\text{Na}^+$ ,  $\text{K}^+$ , and  $\text{Ca}^{2+}$ ), and organic acids (acetate and glycolate) were analyzed on a Metrohm IC system (883 Basic IC Plus and 919 Autosampler Plus).

## 2.8. Mass Spectrometry—Orbitrap

The composition of organic matter was measured after solid-phase extraction by Orbitrap mass spectrometry (Hawkes et al., 2016). Solid-phase extraction was conducted as described in Dittmar et al. (2008), with the slight modification that sorbent conditioning and washing was done with 0.1% formic, not hydrochloric, acid. This was done to minimize chloride ions before mass spectrometry.

The extracted samples were diluted with water and methanol to a final concentration of 30 mg/L in 50:50 methanol:water and were loaded into an Agilent 1100 HPLC autosampler. 50  $\mu$ L of sample was taken and injected into a flow of 50% methanol:water that was pumped at 10  $\mu$ L/min, pumping straight to the ESI source (−3 kV). Data were acquired for a total of 12 min, the sample coming through between 2 and 8 min, and the rest of the time used to achieve a blank baseline. Finally, 150 scans were averaged (typically between 3 and 7.5 min) and the data ( $m/z$ , intensity, and peak resolution) were exported to Microsoft Excel, where in-house routines were used to remove noise, align samples, and assign formulas up to  $C_{40}H_{80}O_{40}N_2S_1$  under the conditions  $0.3 \geq H/C \geq 2$ ,  $O/C \leq 1$ . Formulas were only assigned with a mass error  $<1$  ppm ( $m/\Delta m$ ).  $^{13}C$  isotopologues and rare cases of double assignments were removed from consideration.

Typically, 2,000 peaks were assigned to each sample. For this study, the data were reduced to one metric, the weight-averaged H:C ratio ( $H/C_{wa}$ ). This was calculated as  $\frac{\sum(H \times I)}{\sum I}$ , where  $H:C$  is the atomic ratio of each formula detected in a sample and  $I$  is the intensity of the corresponding peak. This measure reduces the complex data into a metric that summarizes the extent of saturation of double bonds in the organic mixture, where 2 would indicate complete saturation.

## 2.9. POM, POC, and C:N

Samples for POM/POC/C:N were collected on glass microfiber filters (precombusted GF/F, Whatman). We took three replicate filters from samples that were thoroughly shaken to homogenize the sample before decantation at the start of the incubation in the laboratory and one filter from each bottle at the end of the incubation. The samples were vacuum filtered, and the vacuum pressure never exceeded 200 mbar. After filtration, the samples were stored frozen at  $-20^\circ C$  until analysis. The filters were freeze-dried prior to analysis and subsequently acidified with 3% HCl in order to eliminate any particulate inorganic C (see Nieuwenhuize et al., 1994) and again dried in a desiccator. POM was calculated from the weight of the filter determined on an ultra-microbalance (Sartorius, Göttingen, Germany) after filtering and drying minus the empty weight. Subsequently, the organic C (POC) and N (PN) contents were analyzed on an elemental combustion system (Costech Instruments, Cernusco s/Nav., Italy). Therefore, the C:N ratio refers to the ratio POC:PN. The precision of C and N was determined from analysis of ethylenediaminetetraacetic acid (LECO Corporation; Saint Joseph, MI, USA) with a known C and N content. The average C and N content deviated by 1.2% for C and 1.4% for N from the certified sample ( $n = 28$ ).

## 2.10. Bacterial Abundance and Production

Total bacterial cell numbers were determined by epifluorescence microscopy after staining with SYBR Gold (Invitrogen, Darmstadt, Germany; Shibata et al., 2006). One to two milliliters of formaldehyde fixed (2% final concentration) sample was filtered onto a 0.2- $\mu$ m polycarbonate filter (Whatman, Dassel, Germany) and embedded in Citifluor (CITIFLUOR AF1, Science Services, München, Germany) with a SYBR Gold staining solution diluted to 1:1000 (Molecular Probes, Eugene, Oregon, USA). At least 50 bacterial cells per field were counted in 10 fields of view (minimum of 500 cells per filter) with an epifluorescence microscope at 1000 $\times$  magnification (Nikon Eclipse E600, Tokyo, Japan). Bacterial production was determined via incorporation of  $^3H$ -leucine into the protein fraction using the protocol of Smith and Azam (1992). Unfiltered water (1.7 ml) was incubated with  $L$ - $^3H$ -leucine (Perkman Elmer, Waltham, Massachusetts, USA, specific activity 161 Ci/mmol) for 1 hr at the field site at a final concentration of 100 nmol/L. Three replicates and one blank were measured for each sampling site. Net disintegrations per minute were converted to pmol leucine  $L^{-1} d^{-1}$ , and an isotope dilution factor of 2 was applied (Kirchman, 1993). Bacterial carbon production ( $\mu g C L^{-1} d^{-1}$ ) was calculated, using the conversion factor of 0.86 (Simon & Azam, 1989).

## 2.11. Calculation of Decay Rates and Half-Lives of Organic C

Decay rates ( $k$ ) of the dissolved and particulate organic C pool ( $k_{DOC}$  and  $k_{POC}$ , respectively) for the 7-day incubations were calculated separately as  $OC(t) = OC_0 e^{-kt}$  with initial and final DOC or POC concentrations

considered as the organic C (OC) concentration measured at the beginning ( $OC_0$ ) and end of the incubation ( $OC(t)$ ), respectively. We evaluated only incubations with a net organic C loss as we aimed to obtain the decay rates of organic C along the continuum of inland waters. Therefore, incubations with an increase in organic C concentrations during incubations in which net production of organic C occurred (a total of two sample sites for DOC and four for POC) were discarded from further analysis. We assumed decay to be a first order process and selected an exponential decay model to obtain the degradation rates of POC and DOC. First order exponential decay models have been extensively applied to fractional loss in aquatic and terrestrial organic matter decay studies (Catalán et al., 2016; Olson, 1963) and we used the same decay model as Catalán et al. (2016) to be able to compare our results to that study. In a second step, we corrected  $k$  for the in situ water temperature of the sampling site, using the Arrhenius equation with a temperature coefficient of 2 according to Catalán et al. (2016). More details can be found in Catalán et al. (2016).

### 2.12. Statistics

The results are presented as mean  $\pm$  standard error unless stated otherwise. Some data showed nonnormal distribution, and thus, we ran nonparametric tests. In order to test qualitative changes during the incubations comparing dependent samples before and after treatment, we performed Wilcoxon signed-rank tests (Wilcoxon, 1945). Differences between DOC and POC decay rates and half-lives were tested with the Wilcoxon sum-rank test appropriate for independent group comparisons (Wilcoxon, 1945). Additionally, we tested relationships between WRT (as a surrogate for the aging of a water sample along the aquatic continuum) and proxies for terrestrial sources of DOM (i.e., aromaticity as SUVA) and POM (i.e., C:N) with simple linear regressions. The WRT was  $\log_{10}(x + 1)$ -transformed to account for the wide range of WRTs. The statistical analyses were done in R 3.2.2.

Multivariate linear regressions by means of partial least squares projections (PLS regression; Eriksson et al., 2001) were conducted to determine the major environmental factors explaining the patterns in DOC and POC degradation. We used the relative changes in DOC and POC in relation to their single initial concentrations (in %; data shown in Figure 3a) as dependent (Y) variables and a set of environmental factors (summarized in Table 2; data in the supporting information S1) as independent (X) variables. Both POC and DOC degradation rates were arcsine transformed, and whenever environmental data did not fulfil the recommendations (see Eriksson et al., 2001), they were  $\log_{10}$  or  $\log_{10}(x + 1)$  transformed. Data were mean-centered and scaled to unit variance prior to analysis. The results of the PLS regression are shown as loadings plots, which reveal the importance of the independent variables in explaining the dependent variables. The importance of independent variables is summarized as the variable influence on projections (VIPs) and values  $>1$  are considered as highly influential, between 1 and 0.7 as moderately influential and  $<0.7$  as less influential. Model evaluation was done on the basis of the  $R^2Y$  and  $Q^2$  values.  $R^2Y$  is comparable to  $R^2$  for simple linear regressions and reveals the explained variance of X by Y (goodness of fit) and  $Q^2$  the predicted variation of the model. The closer and higher both values, the more robust is the model. In addition, permutation tests (100 times permutations) were run to assess the explained variance that can be attributed to random chance for both models. The original  $R^2Y$  and  $Q^2$  provided an estimate of the background correlation due to chance for the Y variable. Small background correlations indicate a stable model (Eriksson et al., 2001). The PLS regressions were performed in SIMCA 14 (Umetrics, Umeå, Sweden).

## 3. Results

### 3.1. Quantity and Quality of Organic C Along the Aquatic Continuum

The DOC concentrations were higher than POC concentrations at all sites. Concentrations of DOC tended to decrease toward longer WRTs with concentrations ranging from 30.4 to 37.3 mg/L in the peat samples, from 2.7 to 24.2 mg/L in running waters, and from 3.0 to 23.8 mg/L in the lakes (Table 1 and Figure 2a). Percentages of POC were  $9 \pm 1\%$  of the TOC pool, and POC concentrations ranged from 1.5 to 21.4 mg/L (one extreme value of sample site 10) in peat surface waters, 0.3 to 5.6 mg/L in running waters, and 0.2 to 2.9 mg/L in lakes (Table 1 and Figure 2a). The FI and SUVA showed a strong terrestrial influence, especially in the peats, running waters, and lake samples with short WRTs (Figures 2b and 2c). The strong terrestrial influence was obvious from the FI, which was below 1.4 in most samples, and reaching higher values only in some lakes with WRT  $>1$  year (Figure 2b). Similar tendencies were visible in the SUVA (Figure 2c). Most SUVA values were

**Table 2**  
*Environmental Variables for Partial Least Squares (PLS) Regressions*

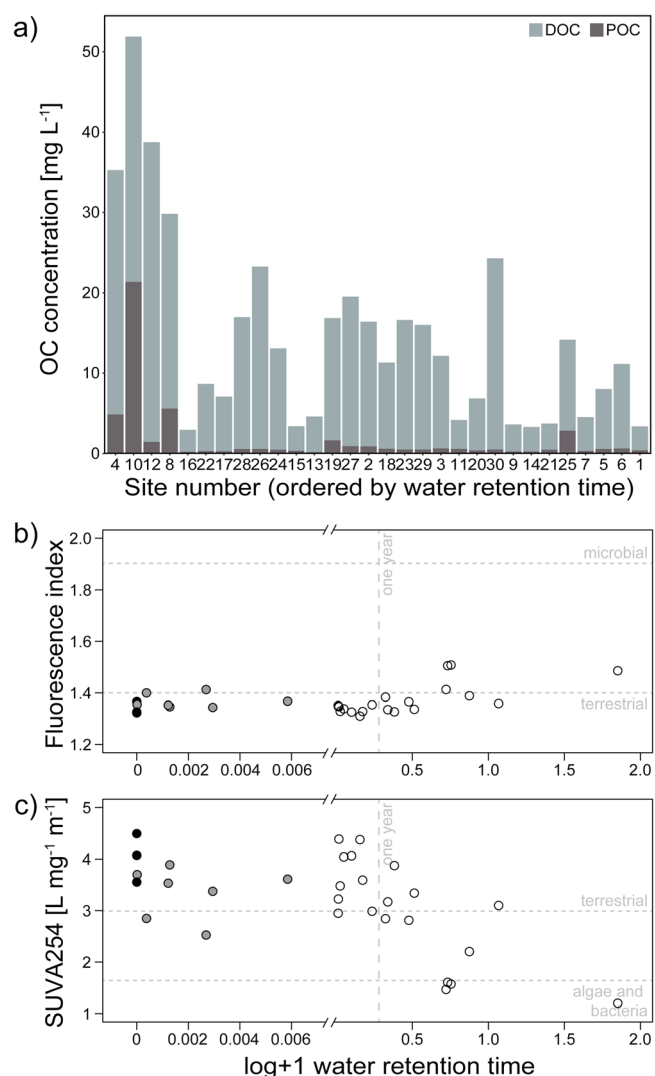
Parameter	Abbreviation	Transformed?	VIP values of POC degradation model	VIP values of DOC degradation model
Relative dissolved organic carbon degradation (in percent) compared to original dissolved organic carbon pool	DOC degr	Arcsine		
Relative particulate organic carbon degradation (in percent) compared to original particulate organic carbon pool	POC degr	Arcsine		
Particulate nitrogen concentration	PN	Log10	1.67	1.34
Water temperature	T	no	1.52	1.28
Particulate organic carbon concentration	POC	Log10	1.50	1.42
Fluoride concentration	F	Log10	1.43	1.48
Total nitrogen	TN	Log10	1.29	1.06
Total phosphorus	TP	Log10	1.28	1.06
Acetate and glycolate concentration	Acetate/glycolate	Log10	1.25	0.74
Particulate organic matter concentration	POM	Log10	1.20	1.25
Chlorophyll- <i>a</i>	Chl <sub>a</sub>	Log10	1.18	1.02
Absorbance at 420 nm	Color <sub>420</sub>	Log10	1.03	1.32
Dissolved organic carbon concentration	DOC	Log10	1.03	1.33
Calcium concentration	Ca	Log10	1.01	0.45
Sodium concentration	Na	Log10	1.00	0.79
Chloride concentration	Cl	Log10	0.92	0.42
pH	pH	no	0.79	0.69
Sulfate concentration	SO <sub>4</sub>	Log10 (x + 1)	0.78	0.67
Bacterial abundance	Bacabun	Log10	0.72	0.95
Fluorescence index	FI	no	0.71	0.97
Carbon to nitrogen ratio of particles	CN	no	0.66	0.88
Potassium concentration	K	Log10	0.58	0.34
Oxygen concentration	O <sub>2</sub>	Log10	0.53	0.77
Bacterial carbon production	BCP	Log10	0.45	0.96
Specific UV absorbance at 254 nm	SUVA	no	0.40	0.99
Freshness index	FRESH	no	0.35	0.73
Humification index	HIX	Log10	0.26	1.04
Water retention time	WRT	Log10 (x + 1)	0.20	0.83

Note. VIP = variable influence on projections; POC = particulate organic carbon; DOC = dissolved organic carbon.

$>3 \text{ L mg}^{-1} \text{ m}^{-1}$  with the exception of two running water sites (16 and 28) and in 45% of the lake samples with most of them with WRTs  $>1$  year. When testing the terrestrial character of the DOM pool (using SUVA as the proxy for the terrestrial influence) along the aquatic continuum, we found a significant decrease with longer WRTs (linear regression of SUVA with  $\log_{10}(x + 1)$  WRT for lakes:  $y = -1.48x + 3.63$ ,  $R^2 = 0.50$ ,  $p < 0.001$ ). The change in the qualitative parameters (spectroscopic indices and  $\text{H:C}_{\text{WA}}$  from Orbitrap mass spectrometry) was prominent at WRTs  $>1$  year. HIX, for example, was lower in samples with WRTs  $>1$  year (Figure 3a), and, accordingly,  $\text{H:C}_{\text{WA}}$  increased at longer WRTs (Figure 3b). The C:N ratios of the particulate pool showed a significant decrease with the lowest values in the samples with WRT  $>1$  year, but the explained variance was very low (linear regression of C:N and  $\log_{10}(x + 1)$  WRT:  $y = -1.92x + 8.78$ ,  $R^2 = 0.13$ ,  $p = 0.027$ ). The C:N ratios ranged between 4.6 to 13.6 (Figure 3c) and the majority of the C:N ratios were in a similar range as the peat samples (peats ranged between 8.0 to 11.5). The phytoplankton C content calculated from chlorophyll-*a* concentration showed minor contributions to the standing stock of the POC pool with  $9.7 \pm 1.2\%$  (range: 0.7–21.8%) in the sampled boreal freshwaters (Figure 3d).

### 3.2. Loss of DOC and POC and Qualitative Changes During Incubations

The DOC and POC decreased in almost all the incubations (except in some replicates of sites 19 and 25 for DOC and 10, 12, 24, and 26 for POC; Figure 4a). The samples that did not show a net decrease in all replicates were excluded from further analyses (average calculations and PLS) but are still displayed in the figures. During incubations,  $2.3 \pm 0.3\%$  (range, 0.3–6.2%) of DOC was lost (Figure 4a). The average DOC change is less than the precision of the TOC analyzer and should be taken with care. However, we keep all values for further analyses to not overestimate the losses of DOC along the aquatic continuum. In contrast to the low DOC loss,



**Figure 2.** Quantity of organic C pools and quantitative parameters of DOM along the boreal aquatic continuum (increasing WRTs). Organic C concentrations showing dissolved organic carbon (in gray) and particulate organic carbon (black; in mg/L) from peats (4, 10, and 12) to running waters (8, 16, 22, 17, 28, 26, and 24) and then lakes ordered by water retention time (a). Fluorescence index (FI) (b) and SUVA (in  $\text{L mg}^{-1} \text{m}^{-1}$ ) (c) are shown on a continuous scale of water retention times ( $\log_{10}(x + 1)$  scaled). The retention time corresponding to 1 year is labeled with a gray dotted line. Peat samples are displayed in black, running waters in gray and lakes in white.

a much larger fraction of POC was lost (Figure 4a;  $27.8 \pm 2.3\%$ , range 4.3–47.7%). Although the concentration of DOC was generally substantially higher than the POC concentrations, the two pools contributed equally to TOC loss, due to the higher relative loss of POC (Figure 4b). Hence, the average relative decrease in DOC from the TOC pool was  $2.1 \pm 0.3\%$  and ranged from 0.3% to 5.4% (Figure 4b). POC contributed on average  $2.4 \pm 0.5\%$  to the TOC pool loss and had a similar range compared to DOC from 0.2% to 5.4% (Figure 4b).

To assess qualitative changes in DOC and POC during incubation and across the WRT gradient, we measured spectroscopic indices,  $\text{H:C}_{\text{WAV}}$  and the C:N of POC. All spectroscopic indices showed similar results, and hereby, we display and discuss HIX as an example of these indices (Figure 3a). The HIX and  $\text{H:C}_{\text{WAV}}$  (Figure 3b) did not reveal significant changes in DOM during the incubations (Wilcoxon signed-rank test; HIX:  $W = 251$ ,  $r$  [effect size] =  $-0.067$ , and  $p = 0.715$ ;  $\text{H:C}_{\text{WAV}}$ :  $W = 282$ ,  $r$  [effect size] =  $-0.182$ , and  $p = 0.318$ ). In contrast, the C:N ratios of the POM changed significantly (Wilcoxon signed-rank test;  $W = 42$ ,  $r$  [effect size] =  $-0.777$ , and  $p < 0.001$ ) and increased during all incubations except in sites 15 and 16 (Figure 3c).

### 3.3. Organic C Degradation Rates

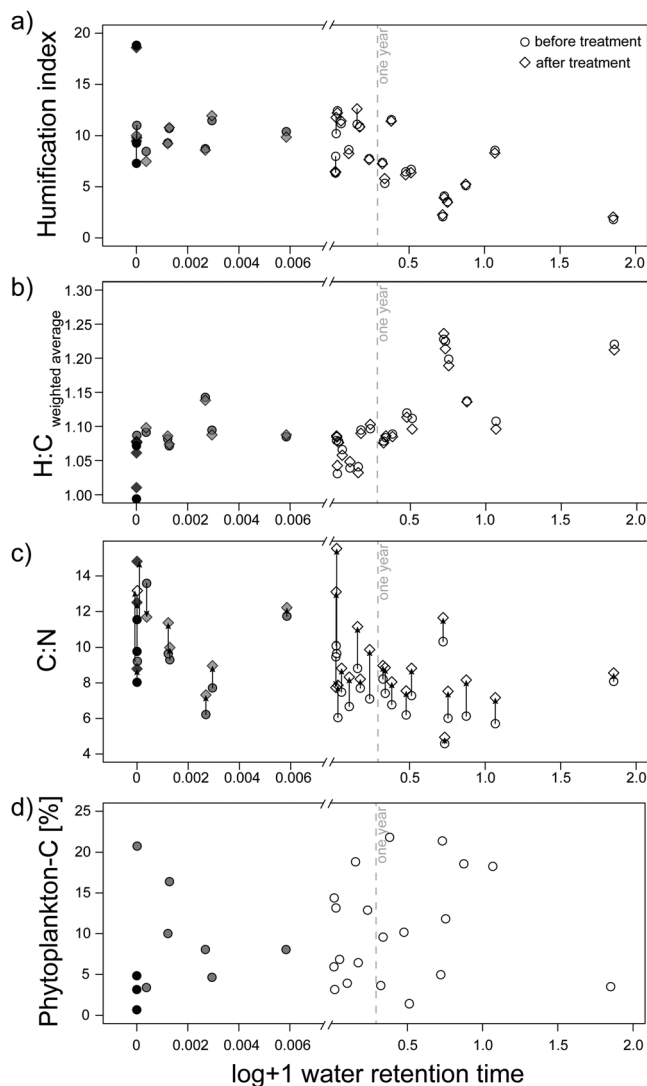
In order to assess degradation rates and compare half-lives of DOC and POC, we calculated decay rates during incubations with an exponential decay model. Decay rates ( $k$ ) as well as half-lives of DOC and POC were significantly different (Wilcoxon rank-sum test;  $k$ :  $W = 2$ ,  $r$  [effect size] =  $-1.068$ , and  $p < 0.001$ ; half-lives:  $W = 726$ ,  $r$  [effect size] =  $-1.068$ , and  $p < 0.001$ ). The POC was degraded at faster rates ( $21.4 \pm 2.2 \text{ year}^{-1}$ ) compared to DOC ( $1.4 \pm 0.2 \text{ year}^{-1}$ ) and the average half-life of POC was  $17 \pm 3$  days and compared to DOC ( $328 \pm 54$  days) about 20 times lower (Figure 5).

### 3.4. Environmental Factors Correlating With DOC and POC Degradation

We identified factors explaining the variability of DOC and POC degradation separately via PLS models (Figure 6 and Table 2). The DOC degradation model extracted one significant component from the data matrix that explained 37% of the variance in DOC degradation. Model predictability and the permutation test revealed that the model is valid ( $Q^2 = 0.15$ ; background correlation in  $R^2Y = 0.29$ ). The loadings plot and VIP values showed that the particulate pool (POC, PN, and POM), nutrients (TN and TP) as well as fluoride, chlorophyll-*a*, DOC concentrations, water temperature, and spectroscopic proxies of terrestrial DOC sources ( $\text{color}_{420}$  and HIX) were the most important variables,

explaining the observed variance in DOC degradation (Figure 6a). Thereby, particle-related parameters (POC, PN, and POM) correlated positively whereas DOC and indicators of terrestrial DOC origin showed a negative correlation with DOC degradation.

The POC degradation model also extracted one significant component, which explained 27% of the variance (Figure 6b). Model predictability was lower for the POC degradation model ( $Q^2 = 0.12$ ), and background correlation was higher ( $R^2Y = 0.37$ ) compared to the DOC degradation model. Similar to the DOC model, particle-related parameters (POC, PN, and POM) and nutrients (TN and TP) were highly influential factors together with chlorophyll-*a*, acetate and glycolate concentration, fluoride, DOC and  $\text{color}_{420}$ , water temperature, and sodium and calcium concentrations, which was confirmed by high VIP numbers (Table 2 and Figure 6b).



**Figure 3.** Qualitative changes in the dissolved fraction: humification index (a), weight averaged H:C ratio (b), the particulate fraction: C:N ratio (c), and the phytoplankton carbon content of POC (d) before (circles) and after (diamond) the incubations over WRT ( $\log_{10}(x+1)$  scaled). The retention time of 1 year is labeled with a gray dotted line. Peat samples are displayed in black, running waters in gray, and lakes are white.

## 4. Discussion

Throughout the aquatic continuum, at WRTs ranging from days or less to several years, POC showed more extensive degradation than DOC. Within the time of incubation, we also found significant qualitative changes in POM, but not in DOM. The POM along the continuum was predominantly of terrestrial origin, as indicated by C:N ratios in the range of the wetland peats (Figure 3c; Bianchi & Canuel, 2011), and low contributions of phytoplankton C (Figure 3d), highlighting the undervalued role of particle transformations in inland waters for the terrestrial C cycle.

### 4.1. Higher Turnover of Particulate Pool

The loss of TOC was similar via loss of POC or DOC during incubations (mean 2.1% [DOC] and 2.4% [POC]; Figure 4b), but one third of the POC pool was lost in 7 days (Figure 4a). Consequently, those experiments suggest that the POC pool in freshwaters has the potential to be transformed much faster than the DOC. Accordingly, the half-lives of POC are about 20 times shorter than DOC half-lives (Figure 5). Corresponding loss rates of suspended stream POC were found in a North American stream, where turnover times of  $\sim 10$  days were determined with similar methods (Richardson et al., 2013). However, our calculations of half-lives have to be taken with care as they are based on only two measurement points. Future studies should thus consider measuring over longer time scales ( $>1$  week to several months) and sampling more time points.

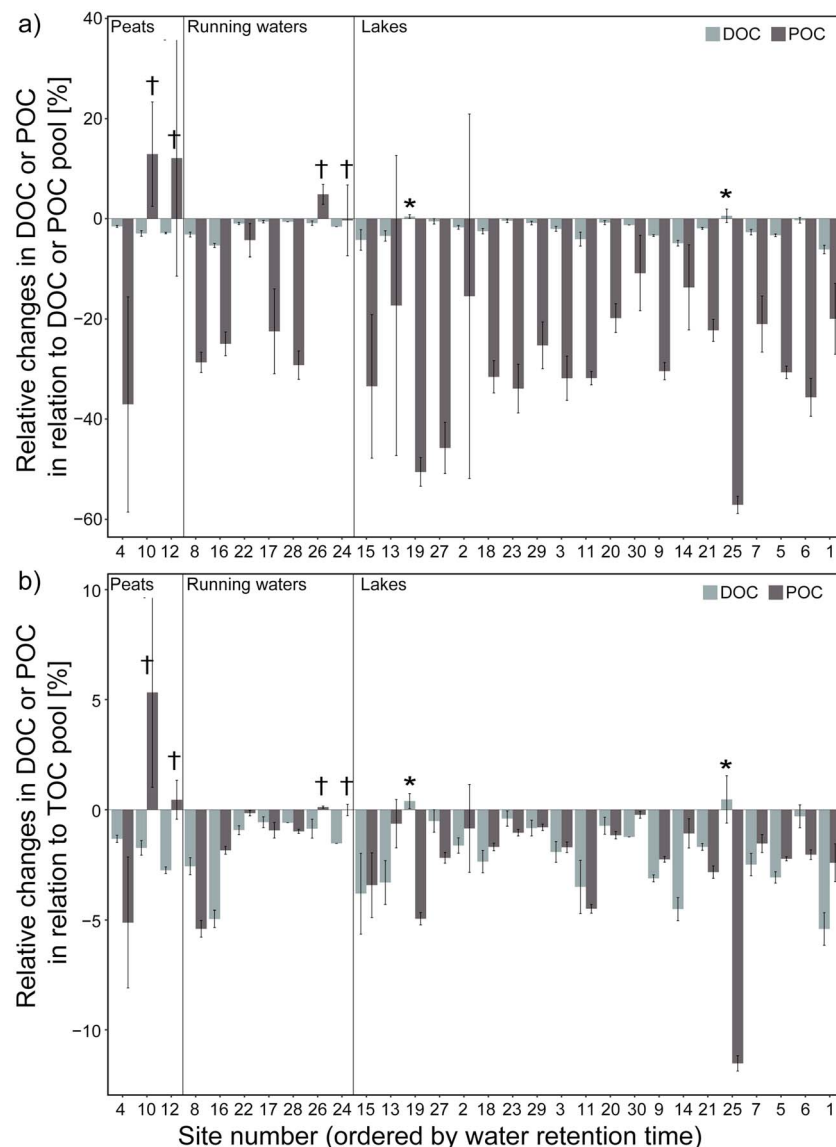
Interestingly, half-lives of terrestrial litter (Figure 5) collected by Catalán et al. (2016) were longer than the half-lives that we calculated for suspended POC. This suggests that the POC degraded in the boreal inland waters, although being largely of terrestrial origin, degrades faster than fresh terrestrial organic matter that has escaped hydraulic transport, or is not yet mobilized. Hence, either the conditions in the water promote faster decay or the organic matter differs in quality between the terrestrial and aquatic habitats. Previous studies on the dynamics of particles in inland waters revealed higher enzymatic rates (Grossart & Simon, 1998; Proia et al., 2016; Worm & Søndergaard, 1998) as well as bacterial activities (Grossart & Ploug, 2000, 2001) on particles. However, those rates were mostly measured on algal aggregates (e.g., diatoms, Grossart & Ploug, 2001). In this study, we used samples from headwater systems such as wetland peats or small streams that typically have a high terrestrial influence (Karlsson et al., 2003; Webster & Meyer, 1997), suggesting that

particles of terrestrial origin are also sites of intense organic C processing, and highlighting the importance of POC transformations for C processing in transit through the aquatic systems.

We cannot exclude that POC was also lost via dissolution. However, only in two samples we detected a net increase in DOC concentration. Dissolution of POC has been shown to be mainly relevant under high irradiances, while only a minor amount of carbon is released to the water column in the dark (Pisani et al., 2011). Moreover, DOC half-lives in our incubations were comparable to the ones reported in a meta-analysis by Catalán et al. (2016; Catalán bioassays, Figure 5) that were calculated with the same approach as our data, suggesting that our observed decreases both in POC and DOC are mainly caused by microbial mineralization of organic C. Therefore, relocation of organic carbon between the two pools (flocculation or dissolution) did, at most, play a minor role during the dark incubations in this study.

### 4.2. Consequences for C Cycling in Boreal Freshwaters

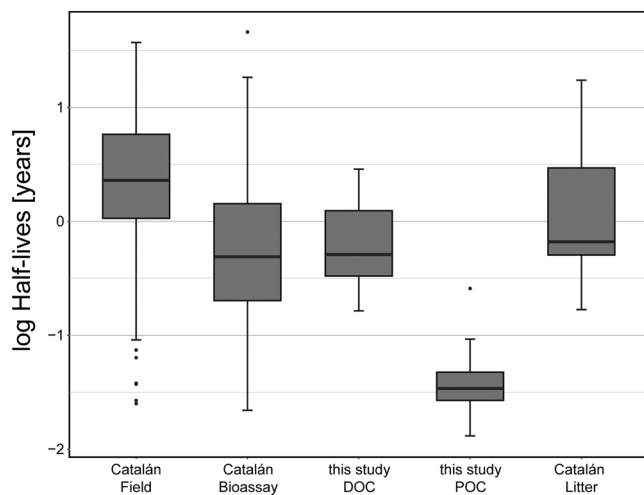
Inland waters of boreal regions are characterized by high influence of terrestrial organic C to both DOC (Kothawala et al., 2014) and organic matter settling to the sediments (von Wachenfeldt & Tranvik, 2008).



**Figure 4.** Relative quantitative changes in the pools of dissolved (DOC, in gray) and particulate organic carbon (POC, in black) after the incubation related to the initial concentrations of DOC and POC (a) and total organic carbon (DOC + POC) after the incubation (b) (mean  $\pm$  SE). Negative values indicate a decrease after the incubation (i.e., degradation) and \* mark sites with increases in DOC and † sites with increases in POC. Site numbers are ordered according to WRT as in Figure 2a.

Accordingly, we found a strong terrestrial signature in both these fractions (Figures 2 and 3). However, with increasing WRT, we found not only decreasing concentrations of DOC and POC but also decreasing terrestrial characteristics, as indicated by SUVA and  $H:C_{WA}$  (Figures 2a and 2c). This suggests gradual loss of organic C during residence time in surface waters, as previously demonstrated (Catalán et al., 2016; Molot & Dillon, 1996), as well as selective loss of terrestrially derived DOC (Kellerman et al., 2014; Weyhenmeyer et al., 2012). Interestingly, the loss of DOC and POC from the TOC pool during incubation were on similar levels downstream the aquatic continuum (Figure 4b) but the POC pool was quantitatively much smaller than the DOC pool (Figure 5). Consequently, to sustain the similar share of POC and DOC despite the much shorter half-life of POC, there has to be a continuous and substantial replenishment of POC along the aquatic continuum (conceptually pictured in Figure 7).

Organic particles can be introduced and formed by several processes including terrestrial inputs from the surroundings (Cole et al., 2006). However, POC input from terrestrial boundaries is mostly driven by hydrology



**Figure 5.** Half-lives of different pools or incubations of organic C. For comparison of the DOC and POC half-lives from this study, we used the data set collected and published in Catalán et al. (2016). “Catalán Field” represents half-lives calculated from field surveys and “Catalán Bioassay” from laboratory incubations across a diverse set of freshwater systems (streams, rivers, lakes, and reservoirs). “Catalán Litter” includes data additionally collected from litter degradation experiments in terrestrial sites and as a comparison to other particulate pools for POC. Boxplots show median (line), 25th and 75th percentiles (box) and 5th and 95th percentiles (whiskers). Half-lives (in years) are log<sub>10</sub> transformed.

and major import of POC into freshwaters occurs sporadically during heavy rain events and storms (Dhillon & Inamdar, 2013; Lee et al., 2016) or during leaf litter fall (Richardson et al., 2010). Moreover, replenishment of the POC via direct transport from land into lakes with WRT in the order of years is hard to reconcile with the persistence of a POC pool with a half-life in the order of weeks, as found in this study. Another source of POC is algal production (Hatten et al., 2012). However, stable isotope data have confirmed that POC in north-temperate and boreal systems is mainly dominated by terrestrial organic matter (Karlsson et al., 2003; Wilkinson et al., 2013). Our data support these results. The phytoplankton fraction of the POC pool, as indicated by its chlorophyll concentration is <25% across all systems (Figure 3d). However, we cannot exclude that POC produced by algae contributes to the replenishment of the POC pool, as this pool is often attributed to a high bioavailability (Kritzberg et al., 2004) and might not be detectable in the samples due to the faster turnover. Accordingly, we found lower C:N ratios in the lakes with long WRT, suggesting that POC replenishment via algal production might be significant at the higher end of our WRT gradient (Figure 3c). Resuspension from sediments has also been shown to be an important source of water column POC (Yang et al., 2016). Finally, flocculation and adsorption of DOC onto particles can contribute to a constant replenishment of POC (Bauer & Bianchi, 2011; von Wachenfeldt & Tranvik, 2008). This is in accordance with previous studies that highlight the role of terrestrial DOC as a precursor of POC in boreal lakes (von Wachenfeldt & Tranvik, 2008). Flocculation and sorption have been suggested to be important factors in the transforma-

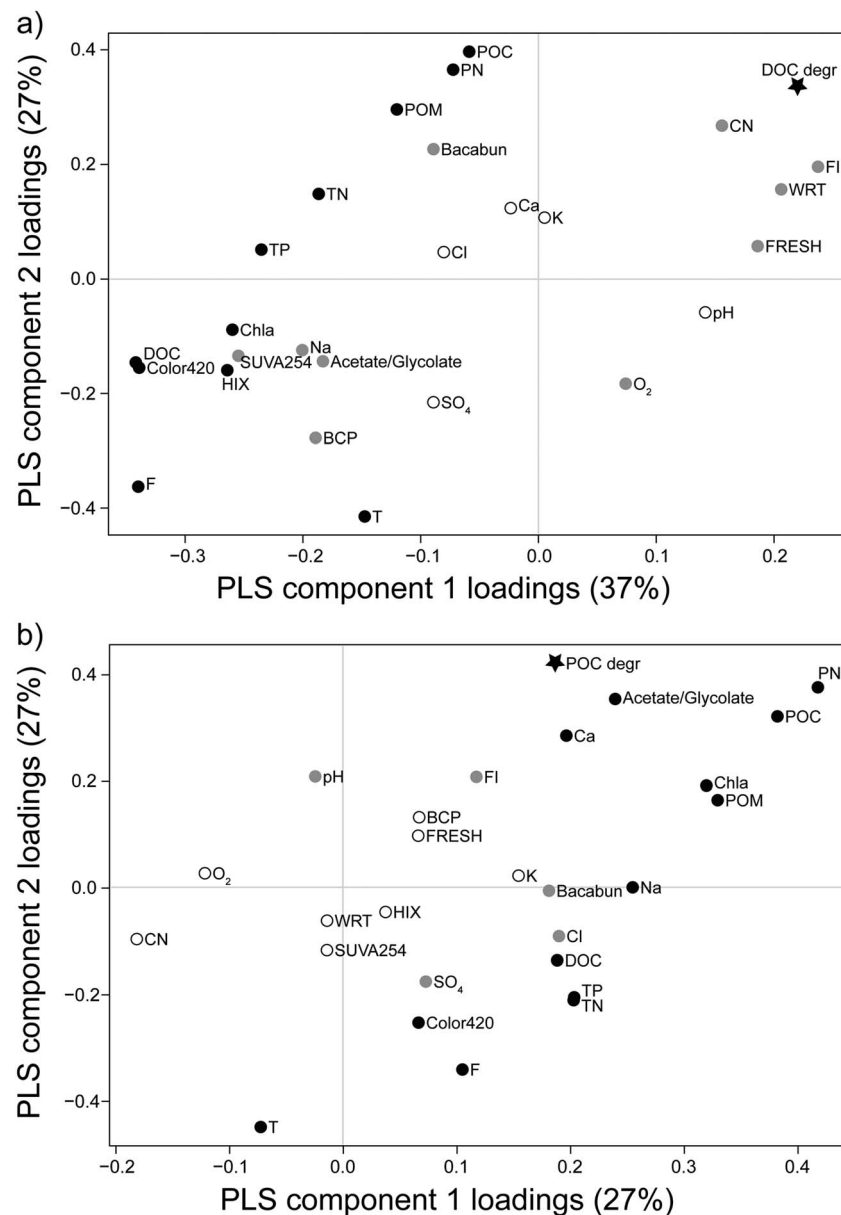
tion of organic C, especially in streams and lakes with low WRTs (Evans et al., 2017). In the natural environment, the flocculation of the dissolved pool can be triggered by, for example, light as shown in a previous study in boreal lakes (von Wachenfeldt et al., 2008). The transition of DOC into POC allows a higher microbial degradation rate and, thus, a more rapid cycling of the organic C due to higher biomasses of the microorganisms and higher enzyme activities (Grossart & Ploug, 2000; Grossart & Simon, 1998). Assuming that POC replenishment is entirely due to transformation of DOC, between 0.02% and 2.06% of the DOC pool per day across all sites would flocculate or adsorb onto particles to sustain observed POC concentrations. This is similar to the range found by von Wachenfeldt and Tranvik (2008).

#### 4.3. Low Microbial Influence on DOM Quality at Short WRTs

Despite a decrease in DOC during our incubations, we did not measure any qualitative changes as revealed by the optical descriptors and H:C<sub>WA</sub> (Figures 3a and 3b). Although other incubation experiments have detected molecular changes during short-term microbial incubations (Seidel et al., 2015), the effects of microbial degradation on DOM quality might be minor compared with the ones of other transformation processes such as photodegradation (Cao et al., 2016; Chen & Jaffé, 2014). Interestingly, our field data show clear changes in qualitative indices in lakes with WRTs longer than 1 year. Thus, transformation of DOM linked to microbial degradation might be conspicuous at longer time scales than the ones in our 7-day incubations from boreal inland waters. Also, the influence of primary production with a production of fresh autochthonous organic matter might play a greater role than degradation of existing DOM in increasing the H:C<sub>WA</sub> and decreasing SUVA in the lakes with longer WRT. In streams and rivers with short WRT, the input of fresh materials from terrestrial surroundings is likely the major driver of qualitative changes. Despite the fact that Mediterranean rivers can actively process DOC (Casas-Ruiz et al., 2017), in boreal stream networks, in-stream processes seem to have only minor effects on DOM composition (Kothawala et al., 2015).

#### 4.4. Qualitative Changes in the Particulate Pool

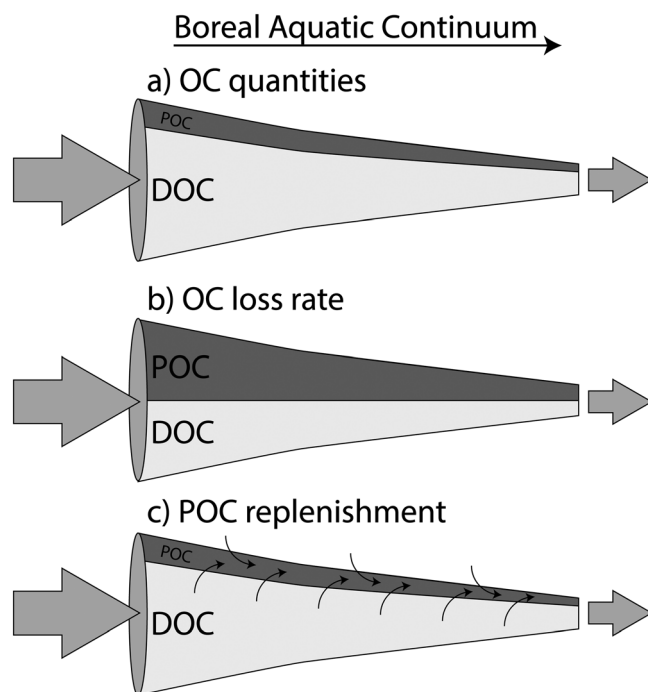
The quality of the POM pool changed during the dark incubations, which is likely due to microbial activity. The C:N ratios mostly increased (Figure 3c); thus, more N was lost in relation to C. Microbial degradation of particles was rapid throughout the WRT continuum (Figure 4a), which suggests that the microbes on the



**Figure 6.** Loadings plots of the PLS analyses. The graphs show the correlation structures of the independent (X) variables (circles) and DOC degradation (percent decrease of initial DOC pool) (a) as well as POC degradation (percent decrease of initial POC pool) (b). Black circles are highly influential variables ( $VIP > 1$ ), gray circles are moderately influential ( $VIP 1-0.7$ ) and blank circles are less influential variables ( $VIP > 0.7$ ). The dependent variables are displayed as black stars and abbreviations of variables are explained in Table 2. PLS = partial least squares; DOC = dissolved organic carbon; POC = particulate organic carbon; WRT = water retention time; Chla = chlorophyll-*a*; TN = total nitrogen; TP = total phosphorus; BCP = bacterial carbon production; bacabun = bacterial abundance; CN = carbon to nitrogen ratio of particles.

particles were not severely limited by nutrients, especially N. Their nutrient demand may have been covered on those particles, contributing to high turnover rates.

C:N ratios are often used as an indicator of the degree of terrestrial origin of the particulate pool, with higher ratios indicating the influence of vascular plants mostly coming from the terrestrial surroundings (Luo et al., 2016; Meyers, 1994). During our dark incubation experiments, the C:N ratio of the particles was pushed toward a more “terrestrial” signal, which is consistent with selective loss of the autochthonous fraction. This is supported by studies on the turnover of sediment organic matter, which was fueled by autochthonous C, resulting in a preferential burial of allochthonous organic matter (Gudas et al., 2012;



**Figure 7.** Conceptual figure of terrestrial DOC and POC and their transformations along the boreal aquatic continuum. POC and DOC concentrations decrease along the aquatic continuum (=active pipe) (a). Their contributions to the loss of TOC (=mostly mineralization) are comparable but decrease along the aquatic continuum (this study for POC and Catalán et al., 2016, for DOC) (b) but when comparing them to their own pool, POC shows much higher losses. To keep up the measured POC concentrations, we suggest a constant replenishment of organic C in the particle fraction that might come from the DOC pool (c). DOC = dissolved organic C; POC = particulate organic C; TOC = total organic C.

Guillemette et al., 2017). However, this points toward an interesting contrast between DOM and POM. DOM has been found to selectively lose terrestrial organic matter during the transport from land to sea (Seidel et al., 2015; Weyhenmeyer et al., 2012), in contrast to the selective retention of terrestrial characteristics in POM that our experimental results suggest and that has been found for boreal sediments (Guillemette et al., 2017).

#### 4.5. Environmental Factors Related to DOC Versus POC Degradation Patterns

Both DOC and POC degradation were significantly correlated with POC, PN, and POM concentrations. The fact that DOC degradation correlates with the same parameters as POC degradation comes as a surprise and might be due to the influence of particle-associated microbes that release enzymes into the water, supporting the degradation of DOC (Grossart & Ploug, 2000; Proia et al., 2016). Those results further support the importance of particle-related processes for the turnover of both DOC and POC and thus their key role for organic C cycling in aquatic systems.

Interestingly, WRT was not significantly correlated with either POC or DOC degradation (Figure 6), thus not supporting the pattern described in Catalán et al. (2016) for the aging process of organic C with longer WRTs. However, these authors had a much larger data set and the PLS model for POC degradation only explained 28% of the variability, and DOC degradation is explained by 37% with our environmental data set (Figure 6). Thus, it seems more difficult to explain and therefore predict POC degradation based on these data. Yang et al. (2016) also mention a large unexplained uncertainty for POC concentrations in running waters in the coterminous United States, indicating complex interactions among multiple factors regulating POC concentrations in freshwaters. Thus, more studies focusing on particle input, composition, and turnover are needed

to better understand transformations of not only POC but also organic matter in general, and the role of aging processes on the particles along the aquatic continuum.

## 5. Conclusions

We show that POC, although occurring at much lower concentrations than DOC, has the potential to be similarly important for the loss of organic C along the continuum of boreal waters, from wetland peats via running waters to lakes with multiyear WRTs. The consistently high POC loss rate along with the sustained concentrations of POC in inland waters requires replenishment of the POC pool. This could possibly be supplied via flocculation and sorption of terrestrial DOC, which is lost along the aquatic continuum. We suggest that the inland water “active pipe” (Cole et al., 2007) to a substantial extent might work via the transformation of terrestrial DOC into POC (Figure 7). The organic carbon in the water column of inland waters has a remarkably short half-life, compared to other compartments, including sediments, and terrestrial and marine environments (Catalán et al., 2016). Here we suggest that, although the inland water organic carbon is generally dominated by DOC, its rapid loss to a large extent might occur via POC formation and degradation by the particle-associated microbial community (Figures 5 and 7).

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