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Fate of leaf litter deposits and impacts on oxygen availability in bank filtration column studies

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ABSTRACT

Degradation of particulate organic carbon (POC) such as leaf litter might deplete dissolved oxygen within the upper layers of bank filtration, an efficient and robust barrier for pathogens and for various organic micro-pollutants (OMP) in water supply systems worldwide. The degradation of OMP during bank filtration depends on the redox conditions. The present study aimed at identifying the impacts and fates of different local leaves on the oxygen consumption and the possible biological degradation of indicator OMP. Oxygen concentrations initially decreased within the columns from around 8 mg/L in the influent to low concentrations indicating extensive consumption within a short travel distance. Still a substantial oxygen consumption was observed after 250 days. OMP concentrations were not significantly affected by the microbial processes. A layer of calcium carbonate crystallites was observed on the POC layer. Some leaf fragments appeared to be persistant towards degradation and the carbon content relative to nitrogen and sulfur contents decreased within 250 days. The results demonstrate that trees at bank filtration sites might have a strong long-term impact on the subsurface redox conditions.

1. Introduction

Many organic micro-pollutants (OMP) including pharmaceuticals, pesticides, industrial and household chemicals persist in wastewater treatment plants (Reemtsma et al., 2010). Possible harmful effects of OMP on the aquatic environment and consumers of drinking water due to life-long exposure are anticipated but not fully resolved, especially for recently developed chemicals (Cunningham et al., 2006).

Bank filtration is an established sustainable and very cost- and energy-efficient option for water treatment (Tufenkji et al., 2002). Therein, natural attenuation processes efficiently eliminate particles, microorganisms, natural organic matter and partially OMP (Grunheid et al., 2005; Massmann et al., 2007; Schmidt et al., 2007; Wiese et al., 2011). In bank filtration, redox conditions play a crucial role in the removal of pollutants and they are mainly influenced by the quantity of carbon substrates (Rauch-Williams et al., 2010) and microbial processes (Schmidt et al., 2007; von Rohr et al., 2014). Organic carbon (OC) in both dissolved (DOC) and particulate form (POC) is a main energy (electron) and nutrient source for microorganisms (Grunheid et al., 2005). Accumulated POC inclusions in bank filtration material contribute to a significant long-term oxygen consumption (Filter et al., 2017).

The OC in aquatic systems stems from autochthonous producers (e.g. primary producers such as algae) or from allochthonous terrestrial producers such as surrounding trees (compare Figs. S1 and S2 in the supporting information). Leaves remarkably contribute to POC inputs into surface water, they have an impact on the food web and influence the structure and function of aquatic ecosystems (Petersen and Cummins, 1974; Meyer et al., 1998; Scharnweber et al., 2014). During decomposition of fallen leaves, aquatic and terrestrial hyphomycetes play an important role (Medeiros et al., 2009; Schönborn and Risse-Buhl, 2013). Leaves fallen into surface water are degraded with extracellular enzymes and hyphae or larger mycelia (Schwoerbel and Brendelberger, 2013). The carbohydrate structures are broken down, the leaves become softer and detritus components are better degradable by benthic macro-invertebrates. However, the impact of terrestrial POC on oxygen availability in the subsequent bank filter is not well understood.

The objectives of the present investigations were to identify the impacts of different leaves (as important source of POC) on 1) conversions within the leaf layer, 2) long-term oxygen consumption in such a thin layer of leaf deposits, 3) the elimination of selected indicator OMP during biological processes within the leaf layer and 4) to characterize the residual leaf fragments after long-term column

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experiments.

2. Materials and methods

2.1. Leaves

Freshly fallen leaves of maple (*Acer platanoides*), birch (*Betula*), beech (*Fagus*), oak (*Quercus*), red oak (*Quercus rubra*) and willow (*Salix babylonica*) around Lake Tegel (Berlin) were collected in autumn. The leaves were dried at 50 °C to minimize thermal degradation. The compositions and leaching of DOC from these POC materials were specific for the different leaves (Bayarsaikhan et al., 2016) and thus different leaves were considered in the present study. To replace naturally occurring fragmentation by detritivorous invertebrates, that could not be simulated in the laboratory, the dried leaves were milled and two sieve fractions (63–125 µm, 125–200 µm) were combined (40% and 60%, respectively) separately for each tree. Leaf fragments smaller than 63 µm and larger than 200 µm were excluded due to partly insufficient mass contributions.

2.2. Long-term column experiments

Technical sand (0.8–1.2 mm) was heat treated at 550 °C to remove all organic material and microorganisms and 40 g were filled into Berlin tap water containing glass columns (37 mm inner diameter, 10.8 cm^2 cross-sectional area, 70 mm height) to obtain fully water saturated sand layers. The prepared leaf materials (500 mg dry weight (at 50 °C) each, corresponding with 46.3 mg/cm^2) were suspended in 50 mL tap water and subsequently filtered in the packed columns to generate POC filter cakes on top of the sand as schematically illustrated in Fig. 1. However, the penetration depth of the leaf fragments into the sand layer could not be quantified. An identical reference columns was operated without a layer of leaf material. The columns were covered with aluminum foil to exclude photo-chemical or photo-trophic effects. Local Berlin tap water (background DOC of ca. 4.5 mg/L, pH of 7.5) originating from bank filtration was used as influent. Calculated volumes of an OMP stock solution containing 10 mg/L benzotriazole, diclofenac and carbamazepine were added to the tap water to obtain concentrations of 1 µg/L. The water was stored in covered but open to atmosphere stainless steel tanks at ambient temperature and no changes of water quality parameters (e.g. dissolved oxygen, pH, DOC) within the tank were observed. The experiments were conducted at ambient temperature around 20 °C. Peristaltic pumps were used to maintain fluxes of approximate 6 mL/h or 14.4 cm/d. The volume flows were controlled by weighing the complete collected effluents.

2.3. Analyses

Dissolved oxygen (DO) was quantified at the column effluents with flow-through cells attached to optodes (OXY-4, PreSens, Germany) with ca. 20 min required to reach a stable value. Oxygen consumptions by POC were calculated by subtracting the measured oxygen concentrations in effluents of POC containing columns from the oxygen concentration in the effluent of the reference column.

Dissolved organic carbon was characterized by size-exclusion chromatography with continuous organic carbon detection (LC-OCD, DOC-Labor Huber, Germany) (Haberkamp et al., 2007) with 1 mL sample volume. OMP concentrations were analyzed with high performance liquid chromatography coupled with tandem mass spectrometry (HPLC/MS-MS) using a TSQ Vantage (Thermo Fisher Scientific, USA) according to an analytical method described previously (Hellauer et al., 2017) with 100 µL sample volume.

Carbon, sulfur and nitrogen contents of the leaves were determined before and after column operation using a thermo-catalytic analyzer (type Vario EL III, Elementar, Germany). Selected leaf samples (original material and aged samples from columns) were sputtered with gold to ensure electrical conductivity and analyzed by field emission scanning electron microscopy (SEM, Hitachi S4000) equipped with energy dispersive X-ray spectroscopy (EDX, SAMx) for elemental analyses.

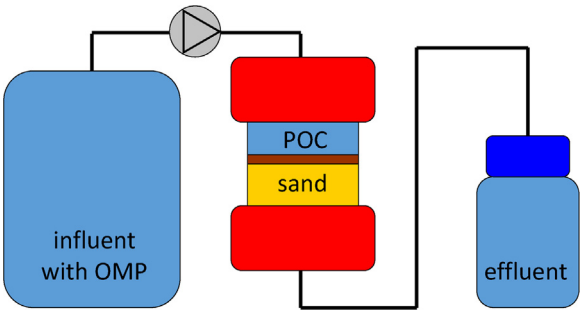

Fig. 1. Schematic illustration of the experimental set-up (the columns were protected from light with aluminum foil).

3. Results and discussion

3.1. Oxygen consumption

Throughout the experiment duration, the effluent oxygen concentrations in the reference column without leaf fragments were between 8 and 10 mg/L as indicated in Fig. 2. Initially, no microorganisms can be expected due to the heat treatment of the sand. The concentration of assimilable and thus oxygen requiring DOC in the influent water was negligible. The other columns revealed a strong decrease of the oxygen concentrations that indicated biological activity of indigenous bacteria on the leaf and in the influent water. Until day 50, low concentrations of less than 3 mg/L occurred in the effluents of all columns with POC depositions. During the run time, the oxygen concentrations in the column effluents increased with time, probably due to depletion of easily degradable organic matter. The consumption was high in all columns with POC deposition but revealed remarkable variation between the individual leaves. Calculated consumptions are provided in Table S1 in the Electronic Supplementary material.

The highest oxygen consumption was observed in the columns with maple and red oak. Probably, these leaves contain higher fractions of more easily degradable organic matter. The oxygen consumption can be attributed to microbial respiration that partly caused almost complete oxygen depletion and a shift to anoxic conditions. As the leaf fragments were mainly deposited as filter cake, the aerobic biodegradation probably occurred within a few millimeters and a very short contact time. However, initial leachates might also be degraded in the underlying sand layer. Another field study confirmed that the highest rates of oxygen consumption occur with a higher microbial activity (Diem et al., 2013). For example, Denecke (1997) referred the oxygen consumption of 35–45% to the biodegradation of POC and DOC during bank filtration at the Rhine in summer and Hoffmann and Gunkel (2011) reported similar observations. In this study, oxygen consumption of more than 8 mg/L were observed, corresponding with 100% consumption. However, under field conditions with lower temperatures, lower bioactivity can be expected. In the present study, POC is assumed to be the primary electron donor for microbial respiration and hence for oxygen consumption. The low infiltration rates induced a higher consumption of oxygen (Hendel, 1999).

Overall, a thin layer of leaf fragments caused large oxygen consumption, which might contribute to anoxic or anaerobic conditions. Thus, the input of terrestrial allochthonous POC might lead to quick consumption of dissolved oxygen in the upper bank by aerobic degradation.

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