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Key Points:
- At anoxic conditions, the degradation of terrestrial organic carbon was reduced by about 50% compared to oxic conditions
- Organic carbon from aquatic production degraded extensively and similarly in different simulated depositional environments
- Production of methane from terrestrial organic carbon was only relevant at anoxic freshwater conditions

Supporting Information:
- Supporting Information S1

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Reduced Mineralization of Terrestrial OC in Anoxic Sediment Suggests Enhanced Burial Efficiency in Reservoirs Compared to Other Depositional Environments

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Abstract Freshwater reservoirs are important sites of organic carbon (OC) burial, but the extent to which reservoir OC burial is a new anthropogenic carbon sink is currently unclear. While burial of aquatic OC (by, e.g., phytoplankton) in reservoirs may count as a new C sink, the burial of terrestrial OC in reservoirs constitutes a new C sink only if the burial is more efficient in reservoirs than in other depositional environments. We carried out incubation experiments that mimicked the environmental conditions of different depositional environments along the land-sea continuum (oxic and anoxic freshwater, oxic and anoxic seawater, oxic river bedload, and atmosphere-exposed floodplain) to investigate whether reservoirs bury OC more efficiently compared to other depositional environments. For sediment OC predominantly of terrestrial origin, OC degradation rates were significantly lower, by a factor of 2, at anoxic freshwater and saltwater conditions compared to oxic freshwater and saltwater, river, and floodplain conditions. However, the transformation of predominantly terrestrial OC to methane was one order of magnitude higher in anoxic freshwater than at other conditions. For sediment OC predominantly of aquatic origin, OC degradation rates were uniformly high at all conditions, implying equally low burial efficiency of aquatic OC (76% C loss in 57 days). Since anoxia is more common in reservoirs than in the coastal ocean, these results suggest that reservoirs are a depositional environment in which terrestrial OC is prone to become buried at higher efficiency than in the ocean but where also the terrestrial OC most efficiently is transformed to methane.

Plain Language Summary The widespread construction of dams disrupts the transport of sediment particles by rivers and traps sediment in reservoirs. The sediment contains organic carbon (OC) that can be degraded into greenhouse gas (carbon dioxide and methane) or buried in the sediment. In the absence of dams, sediment would be deposited in other environments, such as rivers, floodplains, or sea. In our study, we investigated through incubation experiments if the construction of reservoirs creates an environment that stores sediment OC more efficiently than other environments. We found that OC from terrestrial origin is buried more efficiently in anoxic conditions than in oxic conditions. Slowing down of water and high primary production makes reservoirs prone to anoxia, implying that they may be sites of highly efficient OC burial. However, anoxic reservoir conditions were also characterized by higher methane emissions than other environments. OC from aquatic origin was degraded extensively in all studied conditions. Our results suggest that the ~50% of the terrestrial OC that escapes degradation if it gets trapped in an anoxic reservoir may be accounted as a new carbon sink; however, whole-system studies that account for site-specific environmental conditions are necessary to calculate the magnitude of this effect.

1. Introduction

Freshwater reservoirs, like all inland waters, play an important role in the global carbon cycle both as carbon (C) sinks, due to C burial in their sediment, and as C sources, due to emission of the greenhouse gasses carbon dioxide (CO_2) and methane (CH_4) to the atmosphere (Deemer et al., 2016; Mendonça et al., 2017; Prairie et al., 2017). Freshwater reservoirs are responsible for about 40% of the global inland water organic carbon (OC) burial (Mendonça et al., 2017) but in how far reservoir OC burial is accountable as a new, anthropogenic carbon sink is currently unresolved (Prairie et al., 2017).

If the OC buried in reservoirs originates from within-reservoir CO_2 fixation by aquatic plants (phytoplankton and macrophytes), it may be accounted as a new C sink resulting directly from the creation of the

reservoir. On the other hand, if the buried OC is of terrestrial origin, burial in reservoirs may just be regarded as displaced burial, that is, OC that would have been buried in other depositional environments (alluvial soils, floodplains, natural lakes, and coastal ocean) is buried in a reservoir instead, and therefore, reservoir burial of terrestrial OC may not be accounted as a new C sink (Prairie et al., 2017). However, this reasoning is not valid if the OC is buried more efficiently in reservoirs than in other depositional environments: in that case, the additional fraction of buried terrestrial OC qualifies as a new, anthropogenic C sink. There are indications that reservoirs may be characterized by a high OC burial efficiency (OC BE; Mendonça et al., 2016; Sobek et al., 2012), defined as the ratio between OC burial (i.e., OC that stays in the sediment over periods of decades or longer) and OC deposition (OC that settles onto the sediment surface). However, there are no systematic cross-system comparisons of OC BE in different depositional environments. The annual C accumulation in reservoirs is ~0.06 Pg C/year (Mendonça et al., 2017), but it is currently not possible to estimate how much of the accumulated OC in reservoir sediments can be accounted as an anthropogenic C sink due to differences in OC BE.

Reservoirs are also important sources of anthropogenic CH_4 emission to the atmosphere (Prairie et al., 2017). Deemer et al. (2016) found that the areal CH_4 emission rate from reservoirs is higher than from natural lakes, ponds, rivers, and wetlands and increases with reservoir productivity. The bottom water and sediment of reservoirs is frequently low or free of dissolved oxygen, particularly in tropical and eutrophic systems, owing to the combination of thermal stratification of the water column and high sedimentation rates supplying organic matter to microbial respiration in the sediment (Fearnside & Pueyo, 2012; Prairie et al., 2017). Oxygen deficiency, in turn, promotes the production and release of CH_4 from sediments to the water column and the atmosphere. On the other hand, anoxia has also been shown to reduce OC degradation, and thereby promote OC burial, in both marine and freshwater sediment (Burdige, 2007; Carey et al., 2018; Sobek et al., 2009). Since river damming creates a depositional environment that is prone to develop oxygen-deficient bottom water, reservoir construction may enhance not only the C sink but also the overall extent of OC transformation into CH_4 in a river system. However, the dual role of reservoir sediments as OC sinks and CH_4 sources remains poorly understood (Prairie et al., 2017), and it is currently unclear in how far the efficiency of OC burial and CH_4 production is different in reservoirs than in other depositional environments.

We performed laboratory incubation experiments to test the hypotheses that (1) anoxic freshwater conditions are characterized by a lower sediment OC mineralization (implying more efficient OC burial) than conditions of other depositional environments along the land-sea aquatic continuum (river, floodplain and sea environments) and that (2) the extent of CH_4 production at anoxic freshwater conditions is higher than at the conditions of other depositional environments. Since the OC source has strong implications in terms of accounting of OC burial, these hypotheses were tested both for sediment containing predominantly terrestrial OC and for sediment containing predominantly aquatic OC.

2. Materials and Methods

2.1. Overview

We incubated sediment of an oligotrophic drinking water reservoir in conditions that mimic six different depositional environments (treatments) and measured their effect on the sediment OC mineralization rates (response). The treatments were manipulating the oxygenation regime of the medium (water or air) overlying the sediment, that is, oxygenated and anoxic freshwater (representing conditions in a reservoir or a natural lake), oxygenated and anoxic sea (representing different marine depositional environments), oxygenated freshwater with mixing of sediment (representing river bedload conditions), and oxygenated atmosphere (representing floodplain soil surface conditions; Table 1). The experiment was first run using homogenized natural (unamended) sediment to study the treatment effects on sediment that contains predominantly terrestrial OC and then run a second time after addition of aquatic OC (phytoplankton debris) to the sediment. For the unamended sediment, the mineralization rates were measured as dissolved inorganic carbon (DIC) and CH_4 production and O_2 consumption over time. For the sediment amended with aquatic OC, the total mass loss of carbon during the incubation was used as a measure of OC mineralization.

Table 1
Conditions at Which Sediment Mineralization Was Tested

Treatment	Medium above sediment	Oxygenation	Stirring regime	Average incubation length: Unamended sediment	Average incubation length: Amended sediment
Freshwater oxic	Artificial lake water	Oxic	Water column	26 hr (25–27)	34-day incubation after which dry C loss was measured
Freshwater anoxic	Artificial lake water	Anoxic	Water column	84 hr to 56 days	
Sea oxic	Artificial seawater	Oxic	Water column	32 hr (23–42)	
Sea anoxic	Artificial seawater	Anoxic	Water column	66 hr to 58 days	
River	Artificial lake water	Oxic	Water column and sediment surface	21 hr (2–24)	
Floodplain	Air	Oxic	No	13 hr (4–27)	

Note. All treatments were run with natural sediment containing predominantly terrestrial organic carbon and then repeated after amendment of the sediment with aquatic organic carbon.

2.2. Site Description and Sampling

Sediment for the experiment was sampled in the oligotrophic drinking water reservoir Chapeu D'Uvas (21°31.13′S, 43°37.34′W in Minas Gerais state, Brazil, with a mean total phosphorus of 12 μg/L and mean total nitrogen of 452 μg/L; Paranaiba et al., 2017). The sampling site was located at 8-m depth at the main river inflow area into the reservoir. Previous and other studies have not detected anoxic bottom water in this reservoir (data not shown), such that the sediment-water interface at the time of sampling most likely was oxygenated. The site was chosen in order to collect sediment that contains a high share of terrestrial OC and a low contribution of fresh aquatic OC. The sediment had a C:N ratio of 13, indicating that the sediment predominantly originated from terrestrial sources (Meyers & Ishiwatari, 1993), and 2.7% OC content. Sediment was sampled with a UWITEC gravity corer (total, 30 cores) into 60-cm-long PVCu tubes. About 5–10 cm of surface sediment was transferred into two 2.5-L PET bottles and stored at room temperature in darkness.

2.3. Sediment Preparation

Prior to the start of the experiment, sediment was sieved through a stainless steel sieve (3.7-mm pore size) to remove larger particles, and it was mixed until completely homogenous. For seawater treatments the sediment was spiked with an inoculum of Baltic Sea sediment and with an inoculum derived from reservoir sediment that was previously incubated under saline conditions (inoculum weight was 1.3% of the total sediment weight), in order to assure the presence of microbial communities adapted to saline conditions. For all treatments, 5 cm of sediment was placed on the bottom of sediment incubation tubes. Sediment for anoxic and floodplain treatments was transferred into oxygen-tight, extensively tested PVCu tubes (400 × 57 mm; Peter et al., 2016)). Sediment for oxic treatments was transferred into PC (400 × 54 mm) tubes that were previously used for oxic sediment incubations (Gudasz et al., 2010). Sediment tubes were equipped with top and bottom stoppers with triple and double O-rings for PVCu and PC cores respectively.

For unamended sediment incubations homogenized sediment from CDU was used without any additions. After incubations of unamended sediment, the unamended sediment was collected from incubation tubes into two beakers (Figure 1), one with all freshwater and one with all seawater treatments, and homogenized. Then, freeze-dried phytoplankton debris (5.9 g per 100 g of sediment, Superfruit organic spirulina powder) was added, and the sediment was homogenized again. This procedure lowered the C/N ratio of the sediment from 13.0 to 6.9, indicative of primarily aquatic OC (Meyers & Ishiwatari, 1993). The OC content of the amended sediment was 5.2% for the freshwater treatments and 5.3% for the sea treatments. For incubation, the amended sediment was distributed into PVCu tubes for anoxic treatments and PC tubes for oxic treatments, as described above.

2.4. Experimental Treatments

All treatments intend to mimic different depositional environments regarding their typical oxygenation regime and were therefore prepared to investigate the differences in sediment OC mineralization rates in response to different oxygenation regimes. We also include seawater treatments since the high

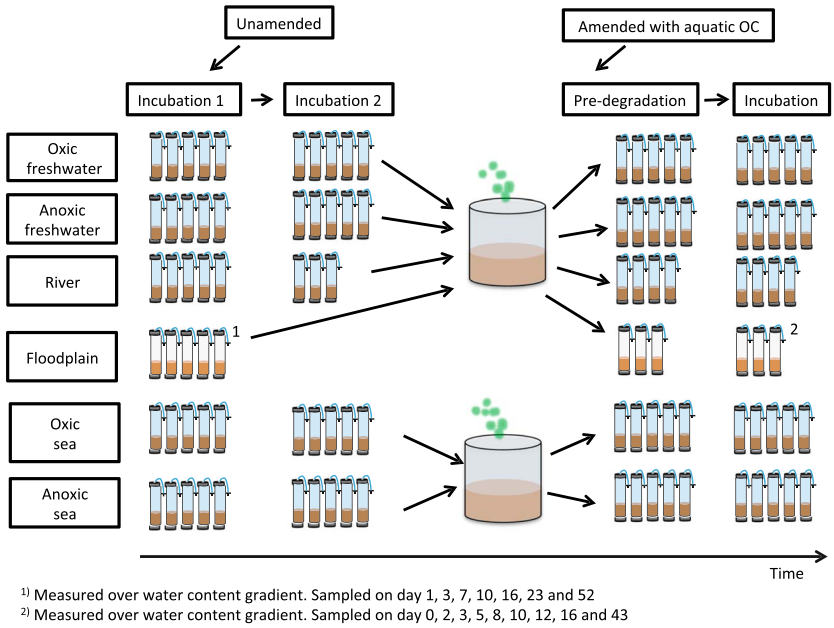

Figure 1. Scheme of the experimental setup.

concentration of sulfate in seawater may affect anaerobic mineralization pathways and thus CH_4 production and/or OC burial. The effects of other factors that can be important in the natural situation were not studied here. The treatments were oxic freshwater, anoxic freshwater, oxic seawater, anoxic seawater, river, and floodplain.

For all freshwater treatments, artificial lake water was prepared by dissolving minerals of macroconstituents and microconstituents in pure water. Exact amounts can be found in Bastviken, Persson, et al. (2004), but no OC was added to the water.

The freshwater oxic treatment was prepared by covering sediment with fully oxygenated artificial lake water (Table 1). The water column above the sediment was continuously gently mixed through the experiment not allowing sediment to be resuspended.

In the anoxic freshwater treatment, sediment was covered with artificial lake water, and prior to the experiment, the water overlaying the sediment was bubbled with N_2 gas until dissolved oxygen in the water column was below detection limit (0.1 mg/L). The anoxic freshwater treatment samples were kept sealed and anoxic throughout the experiment. The water column above the sediment was continuously gently mixed through the experiment in the same way as for the freshwater oxic treatment.

The river treatment was intended to mimic oxygenation conditions during bedload sediment transport in a river. The higher water current in a river as compared to a reservoir or a lake results in frequent deposition-resuspension cycles, which can affect OC mineralization (Rasmussen & Jorgensen, 1992). It was prepared by covering sediment with fully oxygenated artificial lake water, similar to the freshwater treatment, but in addition to mixing the water column, the river treatment was equipped with a magnet-based stirring device located at the surface of the sediment, resulting in the surface sediment being slightly resuspended in the water column. Since an anoxic water column is unlikely in rivers during bedload transport, no anoxic river treatment was included.

In oxic sea treatment, sediment was covered with artificial seawater (35‰) that was prepared with an aquarium salt mixture (Tropic Marin pharmaceutical grade sea salt). The water column above the sediment was gently mixed through the experiment.

The anoxic sea treatment was prepared by covering sediment with the same artificial seawater as the oxic sea treatment, but the water overlaying the sediment was bubbled with N_2 gas until dissolved oxygen in the water column was below detection limit (0.1 mg/L). The anoxic seawater treatment samples were kept

sealed and anoxic through the experiment. The water column above the sediment was gently mixed in the same way and intensity as for the other treatments.

The floodplain treatment mimics riverine sediment that is deposited on an inundated floodplain, followed by receding water and exposure to an oxygenated atmosphere. For this purpose, sediment was prepared by draining the overlaying water from the sediment cores and allowing the sediment to slowly dry at 21 °C with a gentle air circulation over the sediment driven by an air pump. The setup was identical to the other treatments except that sediment had 26 ± 2 cm of overlaying air instead of water. No anoxic floodplain treatment was included since sediment deposited on a soil surface is unlikely to experience anoxia as the water has receded and the soil dries up.

2.5. Incubation Experiments

Prior to the start of the incubations, all sediment cores were allowed to equilibrate for 20 days while being open to the atmosphere and with the water overlying the sediment being mixed with a magnet-based stirring device, in order to approach steady state between the sediment-water gas exchange and OC mineralization. All sediment cores were kept in the dark at 21 °C during both equilibration and incubation periods.

For each sediment incubation, the sediment cores were closed to the atmosphere to measure changes in the concentrations of dissolved oxygen (O_2), DIC, and dissolved CH_4 over time (in the beginning and end of each incubation) in the medium overlying the sediment (water or air). The length of the sediment incubation, that is, the period during which degradation rates were measured, varied from 2 hr to 5 days, depending on the degradation rates in each treatment (Table 1). Water above the sediment was continuously mixed with a magnet-based stirring device. The moment to finish the incubation was determined from the O_2 concentration decrease in oxic treatments. During the incubation, oxygen was continuously monitored in one to three sediment cores of each treatment, and the oxic incubations were stopped once oxygen concentration in the sediment overlaying water decreased by about 2 mg/L, in order to avoid the inhibition of aerobic microbes by oxygen deficiency (Rasmussen & Jorgensen, 1992). Oxygen was measured noninvasively in each core 5 cm above the sediment surface with optical sensors (PreSens) at the start and end of the incubations. Oxygen sensors were factory calibrated. Anoxic incubations were kept for 1 to 5 days to allow for analytically sufficient change in DIC concentration.

Water samples for measurement of concentrations of DIC and dissolved CH_4 were obtained from closed cores thorough pushing down the top stopper, which was equipped with an outlet port and connected to a 60-ml plastic syringe. For measurement of DIC, 9.5 ml of water was transferred into 20-ml syringe equipped with stopcock. There was 0.5 ml of 5% HCl was added to the water, leading to water pH < 3. There were 10 ml of N_2 was added as headspace to the sample. Samples were vigorously shaken for 30 s to allow equilibration of gasses between the headspace and the water. The headspace was injected into the gas chromatograph (GC) equipped with a nickel catalyst for converting CO_2 to CH_4 (GC FID, Agilent Technologies, 7890A GC system). Three replicates of each water sample were analyzed (analytical precision of replicate analysis was <5%). The DIC concentration in the samples was calculated from CO_2 peak areas and a calibration curve obtained from known DIC standards that were run in an identical setup. For measurements of CH_4, 10 ml of sample was transferred into a 20-ml syringe equipped with stopcock, and 10 ml of N_2 was added to the sample. The syringe was vigorously shaken for 30 s to allow equilibration of gasses between the headspace and water. The extracted headspace was injected into the GC calibrated with CH_4 standards of known concentrations. The DIC data for the oxygenated sea treatment did not allow for calculation of C mineralization rates due to high variability between replicates and incubation occasions, possibly related to small changes in DIC against the high background. Instead, C mineralization of the oxic sea treatment was calculated from O_2 consumption rates, using an apparent respiratory quotient of 1 (Sobek et al., 2017).

To measure CH_4 ebullition, the bubbles formed during incubation periods were collected from the incubation cores with a 10-ml syringe connected with a stopcock to the top stopper, at the end of each incubation. The volume of the bubble was read from the syringe gradation. The bubble gas was further diluted with nitrogen to obtain the amount of gas necessary for the GC autosampler and injected into the GC.

Incubations with unamended reservoir sediment were carried out twice on the same sediment cores.

In the floodplain treatment, mineralization rates were measured as the difference in CO_2 and CH_4 concentration in air above the sediment at the start and end of the incubations. For incubations, the cores were closed to the atmosphere, and samples of overlying air were extracted into 10-ml syringes in triplicates through an outlet port. The sediment overlaying air was mixed prior to sample extraction with a magnet-based stirring device placed into each sediment core. Extracted gas samples were injected into the GC for measurement of CO_2 and CH_4 the same day. The cores were incubated for 3 to 29 hr. Before each incubation, the water content in the floodplain treatments was measured gravimetrically. The floodplain treatments were incubated over a gradient of decreasing water content, seven times for the unamended sediment and nine times for the amended sediment.

After the experiment with the unamended sediment was finished, the sediment was collected, amended with phytoplankton debris, and again filled in incubation tubes (see above; Figure 1). Sediment cores amended with phytoplankton debris were allowed to predegrade for 20 days at equal conditions (anoxically) for all treatments before the start of the experiment. After the start of the experiment, due to extremely high mineralization and oxygen consumption rates, cores with oxic water columns (oxic freshwater, oxic sea, and river treatments) were continuously bubbled with air to avoid establishment of anoxia between incubations (the periods during which sediment degradation was measured). Incubations of amended sediment were carried out at three occasions: 20, 35, and 55 days after sediment was amended with aquatic OC. Incubation setup was identical to incubations of unamended sediment described above. When the sediment cores were closed to measure mineralization rates, they were not bubbled with air. Mineralization rates from only the last measurement occasion (55 days after amendment) are presented here, because mineralization rates at the two first measurement occasions (20 and 35 after amendment) were too high to be reliably quantified in our setup; the oxic treatments ran anoxic so rapidly that it was very difficult to retrieve mineralization rates at truly oxic conditions with the handling times entailed by the setup. This was also the reason for running the predegradation period anoxically, thereby ensuring equal conditions for all cores during predegradation. Incubations of amended floodplain sediment of were only carried out 55 days after amendment. Since the measurement of O_2 consumption as well as CO_2 and CH_4 production was not reliably possible during the amended experiment, OC mineralization in the amended experiment was determined as total OC mass loss over time since addition of aquatic OC (57 days).

Sediment amended with phytoplankton debris was dried at 60 °C after incubation. C and N content were measured on acidified samples with a Costech Elemental Analyzer.

2.6. Calculations and Statistics

Concentrations of CH_4 in the water were calculated according to Henry's law, where coefficients for CH_4 solubility in water at varying temperature and salinity were obtained from Wiesenburg and Guinasso (1979) and the ideal gas law. Temperature, water volume in syringes and atmospheric pressure were measured for every sample. Sediment mineralization rates (mg C $\cdot$ m^{-2} $\cdot$ hr^{-1}) were then calculated from concentrations and normalized for the area of sediment cores, water volume in the sediment cores, and incubation time, to be able to compare samples and treatments.

For amended sediment samples overall OC degradation in experiment was determined as the difference in C mass initially (directly after amendment of the natural sediment with phytoplankton debris) and at the end of the experiment. We assumed that the observed OC degradation is entirely attributable to the degradation of the added aquatic OC (Grasset et al., 2018).

The treatments were compared with analysis of variance (ANOVA), *t* test, or Wilcoxon test in case the data could not be assumed to be normally distributed. Shapiro-Wilk normality test and plots of model residuals were used to check if the residuals were normally distributed. Levene's test was used to check for the homogeneity of variance. All data analyses were performed in R 3.4.3 (R Core Team, 2017), and packages *car* (Fox & Weisberg, 2011) and *ggplot2* (Wickham, 2009).

3. Results

3.1. Unamended Sediment

In the unamended sediment, containing predominantly terrestrial OC, we found a significant difference in DIC production between all oxic treatments and all anoxic treatments (mean ± SD for oxic freshwater, sea

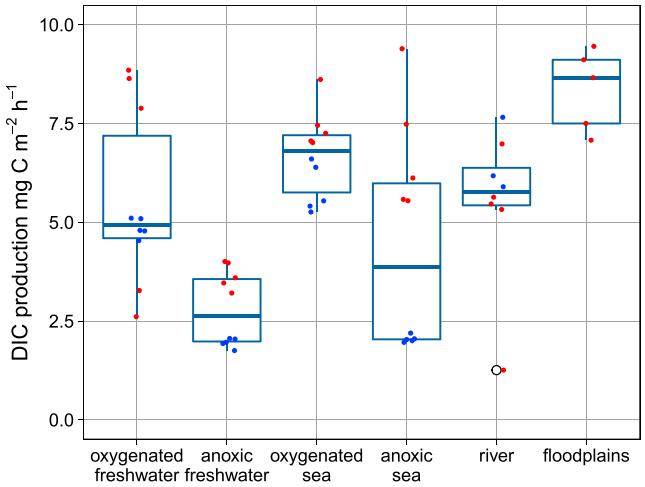

Figure 2. Dissolved inorganic carbon (DIC) production during degradation of unamended reservoir sediment at freshwater, sea, river, and fully water-saturated floodplain conditions. Boxplots show median value, 25th and 75th percentiles; the large circles represent outliers; the points indicate individual measurements. The red points are measurements from the first incubation, and the blue points are measurements from the second incubation.

and river: 5.9 ± 1.8 mg · m^{-2} · hr^{-1}; for anoxic freshwater and sea: 3.6 ± 2.1 mg C · m^{-2} · hr^{-1}; *t* test, $p < 0.001$; Figure 2). There were no significant differences between oxic treatments (freshwater, sea and river; ANOVA, $p = 0.3$) or between anoxic treatments (freshwater and sea; Wilcoxon test, $p = 0.2$). Unamended sediment exposed to floodplain conditions (i.e., atmosphere) displayed a gradient of CO_2 production over sediment water content (Figure S1a in the supporting information). Sediment fully saturated with water but exposed to the atmosphere produced slightly more CO_2 than sediment incubated with oxic overlaying water (8.4 ± 1.0 mg C · m^{-2} · hr^{-1}; significantly different than oxic freshwater, sea and river treatments combined, Wilcoxon test, $p = 0.02$; Figure 2). Floodplain sediment CO_2 production peaked at 33.6 ± 4.9 mg C · m^{-2} · hr^{-1} at 41% water content (Figure S1a) and was comparable to anoxic treatments only at very low water content (<6%).

CH_4 production from unamended sediment was clearly highest in the anoxic freshwater treatment (0.6 ± 0.1 mg C · m^{-2} · hr^{-1}), and it was significantly higher than in the anoxic sea treatment (0.04 ± 0.03 mg C · m^{-2} · hr^{-1}; Wilcoxon test, $p < 0.001$). CH_4 production was uniformly very low in all oxic treatments (0.01 ± 0.03 mg C · m^{-2} · hr^{-1}; Figure 3). In the floodplain treatment, sediment CH_4 production was very low (0.01 ± 0.01 mg C · m^{-2} · hr^{-1}) and not significantly different from other oxic treatments (Wilcoxon test, $p = 0.4$) if the sediment was saturated with water (water content >60%; Figure 3) or dry (water content <20%; Figure S1b). Also between 20 and 60% water content, the CH_4 production was relatively low (< 0.2 mg C · m^{-2} · hr^{-1}) and reached levels comparable to the anoxic freshwater treatment (1.34 and 0.88 mg C · m^{-2} hr^{-1}) only at two out of 35 measurement occasions. (Figure S1b). We did not observe CH_4 ebullition in any of the incubations of unamended sediment.

3.2. Sediment Amended With Aquatic OC

Sediment amended with aquatic OC had extremely high degradation rates. On average 76% of the added OC was lost through emission of CO_2 and CH_4 during a 57-day experiment, of which sediment was anoxically predegraded for 20 days and later incubated for 37 days (Figure 4). There were no significant differences in OC loss between the treatments by the end of the incubation (ANOVA, $p = 0.6$).

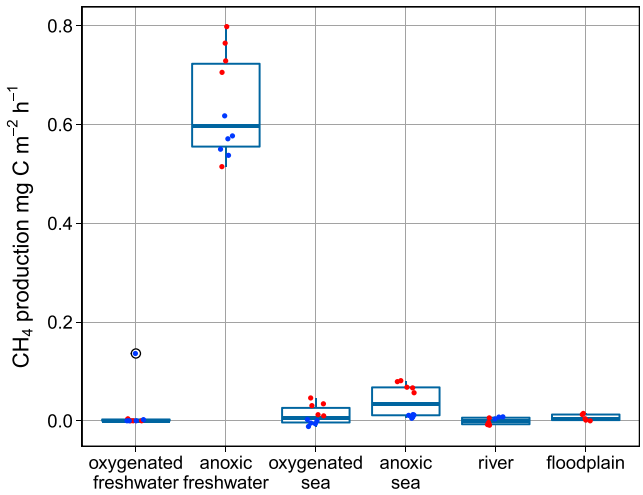

Figure 3. CH_4 production during degradation of unamended reservoir sediment at freshwater, sea, river, and fully saturated floodplain conditions. Boxplots show median value, and 25th and 75th percentiles, the large circles represent the outliers; the points represent individual measurements. The red points are measurements from the first incubation, and the blue points are measurements from the second incubation.

Amending the sediment with aquatic OC resulted in very high CH_4 production in all treatments. At the first measurement, which was carried out after 20 days of predegradation, CH_4 ebullition was so intense in all treatments that reliable quantification was not possible in our experimental setup. At the end of the experiment, 57 days after amendment with aquatic OC and when ~76% of the added aquatic OC was lost, CH_4 production rates were about 5 times higher than in the experiment with unamended sediment, and not significantly different between oxic and anoxic treatments (Wilcoxon test, $p = 0.6$; Figure S2a). In the floodplain treatment, at the end of the experiment, CH_4 production was highest at 50–60% water content and was comparable to CH_4 production in all other treatments (Figure S2b).

4. Discussion

4.1. Anoxia Leads to Highly Efficient Burial of Terrestrial OC

Our results suggest that along the land-sea continuum, terrestrial OC is buried most efficiently under anoxic conditions. OC degradation rates of an unamended sediment containing predominantly terrestrial OC were

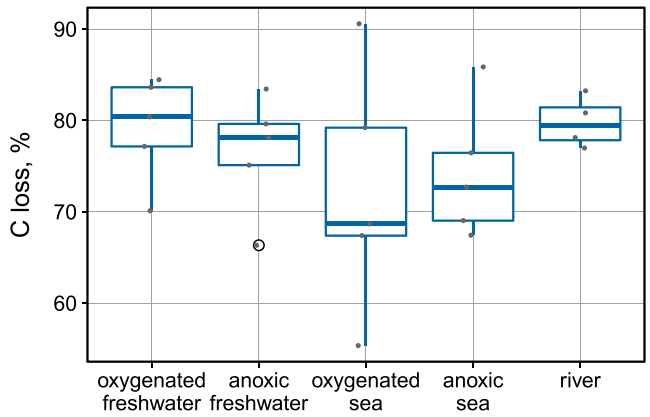

Figure 4. Loss of added aquatic organic carbon during the degradation of unamended sediment amended with aquatic organic carbon by the end of the 57-day experiment.

smaller by a factor of ~2 in anoxic treatments compared to oxic treatments (Figure 2). There was no statistical difference between the anoxic seawater and the anoxic freshwater treatments, indicating that terrestrial OC is equally efficiently buried at simulated marine and freshwater conditions, as long as the bottom water is anoxic.

However, during the first incubation occasion, the DIC production rate in the anoxic seawater treatment was comparable to the mean DIC production rates of the oxygenated treatments (Figure 2). In order to closer examine this result, we immediately performed a third incubation of the anoxic treatments (Figure S3), during which the anoxic seawater treatment returned a negative mean DIC production rate. Apparently, microbial activity was inhibited during the incubation, possibly related to the production of toxic H_2S during sulfate reduction at anoxic seawater conditions (Chen et al., 2008); in fact, the smell of H_2S was very strong when opening these incubations. Conversely, all three incubations of the anoxic freshwater treatment were similar to each other, and no inhibition of microbial degradation was apparent (Figure S3). This evidence suggests that the second incubation of the seawater treatment may already have been affected by inhibited microbial activity, and thus that the overall low mean DIC production rate of the anoxic seawater treatment may be an experimental artifact potentially caused by the accumulation of toxic H_2S. However, we found this evidence too weak to exclude the second incubation data of the seawater treatment, and therefore, we conservatively conclude that in our experiment, anoxia was found to result in significantly reduced DIC production rates of unamended sediment containing predominantly terrestrial OC, at both seawater and freshwater conditions.

Anoxia is not widespread in the sea, and we therefore argue that the anoxic sea treatment is not representative of a condition that terrestrial OC deposited at the seafloor is likely to experience. While some areas in the coastal ocean are characterized by very high sediment deposition and hypoxic conditions that result in very high OC burial efficiency (Leithold et al., 2016), e.g., the Bengal Fan (Galy et al., 2007), most of the coastal areas that receive a high input of terrestrial OC (Schlünz & Schneider, 2000) were not characterized as hypoxic (Diaz & Rosenberg, 2008). Instead, it has been shown that as terrestrial sediment is deposited in estuaries, sediment OC degradation rates can be very high because physical transport and marine benthic invertebrates rework the sediment, resulting in extensive oscillations in redox conditions (oxic-anoxic cycles), which overall increase the degradation of OC (Aller, 1994; Aller, 1998). Therefore, it is unlikely that the undisturbed anoxic sea conditions, as designed in our experiment, are widespread along coastal margins receiving terrestrial OC input. On the other hand, we argue that the anoxic freshwater treatment as designed in our experiment quite realistically mimics reservoir conditions. Many reservoirs, particularly those in warm climates or affected by eutrophication, are characterized by anoxic bottom water (Abril et al., 2005; Carey et al., 2018; Fearnside & Pueyo, 2012; Kemenes et al., 2007; Soares et al., 2008). Even if the bottom water contains dissolved oxygen, the frequently very high sediment deposition rates in reservoirs (Mendonça et al., 2017) result in very short sediment oxygen exposure times (Sobek et al., 2012), and therefore predominantly anaerobic and thus inefficient OC degradation (Sobek et al., 2009). The same reasoning applies to situations where terrestrial sediment is deposited in the river inflow areas of reservoirs: in spite of a shallow and oxygenated water column, the high sediment deposition rate renders a very short oxygen exposure time and thus virtually anoxic sediment (Sobek et al., 2012). We conclude that at a broad scale, anoxic freshwater environments, as can be found in lakes, reservoirs, and peatlands, are efficient in terrestrial OC burial and relatively widespread and therefore are an important carbon sink. Since river damming traps a large share (at least 25%) of the terrestrial sediment flux to the ocean (Vorosmarty et al., 2003), and since sediment deposited in reservoirs is prone to experience oxygen-deficient conditions, which in turn decreases the degradation rate of terrestrial sediment OC (Figure 1), we propose that the construction of reservoirs may enhance the overall burial of terrestrial OC.

Our experiment showed that anoxia reduced the sediment mineralization rates of predominantly terrestrial OC by about half compared to oxic treatments. Similar results have been reported for sediments from natural lakes (Isidorova et al., 2016; Peter et al., 2016). In the following, we give an example of the possible effect

anoxia could have on the OC burial efficiency in a Brazilian reservoir (Mascarenhas de Morais; (Mendonça et al., 2016)). In this reservoir, the OC BE at the observed conditions (oxic bottom water) was calculated by dividing mean OC burial (42 g C · m^{-2} · $year^{-1}$) with OC deposition (i.e., the sum of mean OC burial and mean oxic mineralization rate of 34 g C · m^{-2} · $year^{-1}$), resulting in a OC BE of 55%. If we assume that this reservoir had anoxic instead of oxic deep water, the mineralization rate would be reduced by about half (from 34 to 17 g C · m^{-2} · $year^{-1}$), thereby increasing its OC BE from 55 to 78% and consequently raise OC burial by ~40% (from 42 to 59 g C · m^{-2} · $year^{-1}$). This is a simplistic and hypothetical calculation, and the exact numbers will rely on the exact magnitude of burial, mineralization, and periods of anoxia at each specific reservoir. Nevertheless, this example illustrates that if terrestrial OC is deposited in reservoirs that experience anoxia, it may be buried at elevated efficiency. Future studies should address the potential increase of terrestrial OC burial in reservoirs due to anoxia at a larger scale, accounting for site-specific environmental conditions such as oxygenation, temperature (Yvon-Durocher et al., 2014) and OC sources (Grasset et al., 2018).

4.2. Transformation of OC to CH_4 in Unamended Sediment

Anoxic freshwater conditions were characterized not only by a high OC burial efficiency of predominantly terrestrial OC but also by higher CH_4 production from predominantly terrestrial OC compared to any other treatment (Figure 2). OC is transformed into CH_4 through anaerobic processes in subsurface sediment layers, as freshwater sediment is typically anoxic below a few millimeter depth (Sobek et al., 2009). All other depositional environments had similarly low CH_4 production rates. Most intensive CH_4 production occurs in anoxic environments with low sulfate concentration and high organic matter availability (Bastviken, Cole, et al., 2004). The anoxic seawater treatment was characterized by comparatively high sulfate concentration, which may explain the low CH_4 production. In reservoirs, where anoxia is widespread and high sedimentation rates often limit sediment oxygen exposure (Mendonça et al., 2017; Sobek et al., 2012), CH_4 emission per unit area has accordingly been shown to be very high compared to other freshwater environments (Deemer et al., 2016).

4.3. Aquatic OC Is Buried Equally Efficiently Across Depositional Environments

The OC degradation rates in sediment amended with phytoplankton debris were uniformly high in all treatments. About 76% of the added phytoplankton OC was lost by the end of the experiment (Figure 4); hence, the OC BE of aquatic OC was not related to the conditions of the simulated depositional environments. Similar oxic and anoxic mineralization rates were previously reported in sediments with a high share of labile organic matter, while in sediment containing more refractory organic matter, oxic mineralization rates were greater than anoxic mineralization rates (Bastviken et al., 2003; Hulthe et al., 1998). Since 76% of the added OC was lost already within 57 days, our results suggest that only a small part of phytoplankton OC could remain buried in the sediment of any environment at a longer time scale, in accordance with earlier studies (Burdige, 2007; Sobek et al., 2009). However, aquatic OC is commonly found to be buried at high rates in the sediment of eutrophic reservoirs (Downing et al., 2008). Even if our results indicate that the burial of aquatic OC is not more efficient in reservoir sediment, it seems that reservoirs may store a yet unknown amount of aquatic OC in their sediments. Future research should investigate how significant amounts of aquatic OC can be preserved in freshwater sediments over extended periods of time (Downing et al., 2008).

4.4. Transformation of Amended Sediment Aquatic OC to CH_4

Addition of phytoplankton debris resulted in very high CH_4 production rates in all treatments, as evidenced by high rates of both CH_4 diffusion and CH_4 ebullition. After amendment of the sediment with aquatic OC, the incubations bubbled so intensely that CH_4 ebullition could not be reliably quantified with our experimental setup. Even at the end of the 57-day experiment, when most (~76%) of the added aquatic OC had already been degraded, the CH_4 emission was still about 5 times higher than in the unamended sediment. This result suggests that an increase in aquatic biomass production through eutrophication may lead to increasing CH_4 emissions through ebullition and diffusion in all simulated depositional environments (Davidson et al., 2015; Deemer et al., 2016; Grasset et al., 2018). However, it needs to be noted that in the experiment, fresh dried phytoplankton debris was added to the entire sediment volume, which is an unrealistic scenario for the natural environment, where fresh matter is rather added to the sediment surface. In the

natural situation, by the moment aquatic OC reaches anoxic, methanogenic sediment layers, it will have been predegraded at oxic conditions in the water column and on the sediment surface, to a larger extent than in the experiment. Hence, the extremely high CH_4 production rates upon amendment of sediment with phytoplankton debris observed here may rather represent a worst-case scenario than reflect the natural situation. Nevertheless, the fact that >70% of the added phytoplankton OC was mineralized to CO_2 and CH_4 within 57 days illustrates that the primary fate of aquatic OC is degradation, and only a small fraction will end up being buried. Consequently, while eutrophication may increase OC sedimentation, our results suggest that the majority of the deposited OC may be recycled to CO_2 or transformed to CH_4.

5. Conclusions

The damming of rivers creates an anthropogenic depositional environment that traps a significant amount of the terrestrial sediment flux to the ocean and is prone to develop oxygen-deficient conditions. The experimental results reported here show that terrestrial OC is buried at higher efficiency at anoxic conditions. This suggests that river damming may enhance the magnitude of terrestrial OC burial and that the additionally buried OC in reservoirs may be accountable as an anthropogenic C sink. In the simplified simulations of depositional environments in the laboratory, anoxia resulted in a 50% decrease in mineralization of predominantly terrestrial OC, and this 50% fraction escaping mineralization may constitute additional C burial in an anoxic reservoir; however, studies at larger scale are needed to quantify this effect. Also, if terrestrial OC is deposited in permanently anoxic marine sediments, the OC BE will be similar to anoxic reservoirs, the experiment suggests. Hence, also environmental conditions in the recipient marine sediment will influence in how far the burial of terrestrial OC in individual reservoirs may be accountable as a new C sink. Aquatic OC, on the other hand, was degraded equally well independent of simulated conditions of depositional environments. Future studies should address the efficiency of OC burial in reservoirs and other depositional environments at the scale of ecosystems or river basins and address the dual role of freshwater sediments as C sink and CH_4 source.

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