

Preservation of organic carbon during active fluvial transport and particle abrasion

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ABSTRACT

Oxidation of particulate organic carbon (POC) during fluvial transit releases CO₂ to the atmosphere and can influence global climate. Field data show large POC oxidation fluxes in lowland rivers; however, it is unclear if POC losses occur predominantly during in-river transport, where POC is in continual motion within an aerated environment, or during transient storage in floodplains, which may be anoxic. Determination of the locus of POC oxidation in lowland rivers is needed to develop process-based models to predict POC losses, constrain carbon budgets, and unravel links between climate and erosion. However, sediment exchange between rivers and floodplains makes differentiating POC oxidation during in-river transport from oxidation during floodplain storage difficult. Here, we isolated in-river POC oxidation using flume experiments transporting petrogenic and biospheric POC without floodplain storage. Our experiments showed solid phase POC losses of 0%–10% over ~10³ km of fluvial transport, compared to ~7% to >50% losses observed in rivers over similar distances. The production of dissolved organic carbon (DOC) and dissolved rhenium (a proxy for petrogenic POC oxidation) was consistent with small POC losses, and replicate experiments in static water tanks gave similar results. Our results show that fluvial sediment transport, particle abrasion, and turbulent mixing have a minimal role on POC oxidation, and they suggest that POC losses may accrue primarily in floodplain storage.

INTRODUCTION

Fluvial transport of particulate organic carbon (POC), including discrete particles and mineral-associated organic matter >0.2 μm, represents the primary source of terrestrial organic carbon to the ocean (e.g., Blair and Aller, 2012; Bianchi et al., 2018). The fate of POC during fluvial transit affects atmospheric CO₂ levels over a range of time scales (e.g., Cole et al., 2007; Galy et al., 2007). Transient storage of CO₂ sequestered via photosynthesis in soils and biomass (biospheric POC [POC_{bio}]) represents a short-term (<10⁵ yr) carbon sink (Berner, 1999). POC_{bio} mobilized by and transported through rivers can be oxidized and returned to the atmosphere as CO₂ (Richey et al., 2002; Mayorga et al., 2005), or it can be deliv-

ered to depositional basins to create a long-term (>10⁶ yr) carbon sink (e.g., France-Lanord and Derry, 1997). This CO₂ sink may be offset by CO₂ released from oxidation of fossilized organic carbon within sedimentary rock (petrogenic organic carbon [POC_{petro}]) during exhumation and transport (e.g., Bouchez et al., 2010).

POC losses during fluvial transit can be substantial. Amazon basin POC_{bio} and POC_{petro} oxidation fluxes are approximately an order of magnitude larger than their respective fluxes to the ocean (Richey et al., 2002; Bouchez et al., 2010). Similarly, ~50% of POC_{bio} and POC_{petro} is lost during transit across the Ganges alluvial basin (Galy et al., 2008a, 2008b). Despite large POC loss in transit, the location of POC oxidation is poorly constrained, as existing measurements cannot distinguish between oxidation during in-river transport and oxidation during

floodplain storage. In rivers, fluid turbulence mixes and aerates water, which may enhance oxidation rates (Hartnett et al., 1998). Increases in sediment specific surface area (SSA) from particle abrasion (e.g., Attal and Lave, 2009) can further expose mineral-associated POC_{bio} and POC_{petro} to oxidative conditions, while also creating fine particles onto which discrete POC particles can attach. However, the time POC spends in river transport is small relative to floodplain storage (e.g., Torres et al., 2017). Intermittent floodplain storage may allow sufficient time for POC oxidation (Arora et al., 2016; Scheingross et al., 2018; Wan et al., 2018), but POC can be subject to anoxic conditions, which may promote POC preservation (Boye et al., 2017). The relative rates of POC oxidation during in-river transport versus floodplain storage are key unknowns that are needed to develop a process-level understanding of source-to-sink POC transfer and oxidation.

Determining the location of POC oxidation in lowland basins is complicated by tributary mixing (Galy et al., 2008b; Bouchez et al., 2010) and exchange of POC between rivers and floodplains (Torres et al., 2017). An alternative approach is to isolate in-river POC oxidation via laboratory experiments. Analog experiments have fundamentally advanced river mechanics (Paola et al., 2009); however, flume experiments have yet to explore POC oxidation. Existing laboratory investigations of organic carbon oxidation (e.g., Chang and Berner, 1999; Druhan et al., 2014) were not designed to simulate the physical processes present in natural rivers (e.g., particle abrasion).

Here, we evaluated POC oxidation in flume experiments transporting carbon-rich sediment

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without floodplain storage. Our experiments showed minimal evidence for POC oxidation despite $\sim 10^3$ km of fluvial transport, a distance similar to the lowland portion of the Ganges River. These results are consistent with the idea that POC oxidation in lowland basins occurs primarily during floodplain storage (e.g., Torres et al., 2017), such that the rate and magnitude of CO_2 evasion in alluvial basins are likely set by POC residence time in aerobic floodplains.

EXPERIMENTAL AND ANALYTICAL METHODS

In lowland basins, water chemistry, microbial communities, and other factors can influence POC oxidation (e.g., Battin et al., 2009; Druhan et al., 2014; Bianchi et al., 2018). Here, we sought to isolate the influence of sediment transport, particle abrasion, and liquid-water leaching on POC oxidation using annular flumes that recirculated sediment, and static tanks (control experiments) where POC was subject to liquid-water leaching without sediment transport (Fig. 1). Our experiments used natural sediments with native microbial communities and a 1:10 mix of deionized to tap water with major-element concentrations similar to rivers in the Gaillardet et al. (1999) database (Fig. DR1 in the GSA Data Repository¹).

Turbulent, subcritical flow in flume experiments (Reynolds number $>10^4$, Froude number ~ 0.2) produced dynamical scaling with natural rivers (Paola et al., 2009), with depth-averaged velocities of $\sim 0.25 \pm 0.02$ m/s (Table DR1) similar to near-bed flow velocities in rivers (e.g., Smith and McLean, 1977). Visual observations in a transparent flume showed continual motion of sediment without static deposits. Experiments

ran 6–8 wk at room temperature, producing suspended sediment transport distances of ~ 900 – 1200 km (see the Data Repository; Table DR2).

We performed nine experiments with four distinct materials (Table DR2). To explore the role of crystallographic order on $\text{POC}_{\text{petro}}$ oxidation, we used lignite and shale containing relatively amorphous and more crystalline carbon, respectively, collected from outcrop. To explore POC_{bio} oxidation, we collected sediment from a headwater stream underlain by volcanic rock (Smith and Roe, 2015) with well-described POC composition (Lookout Creek, Oregon; Smith, 2013) and a lowland river with negligible $\text{POC}_{\text{petro}}$ quantities (Rio Bermejo, Argentina) where preliminary data show significant POC oxidation during floodplain storage (Scheingross et al., 2018). In select experiments, we varied sediment size to examine the role of sediment abrasion on POC oxidation (see the Data Repository). Blank experiments (one experiment without sediment and one experiment transporting combusted quartz sand) showed no systematic changes in dissolved organic carbon (DOC) and returned POC concentrations below the measurement detection limit (Table DR3; Fig. DR2).

We assessed solid and dissolved load changes using sediment samples at the start and end of experiments and 5–10 water samples per experiment (see the Data Repository). Solid samples were analyzed for POC content (C_{org} , wt%) and stable carbon isotopes ($\delta^{13}\text{C}_{\text{org}}$, ‰) following carbonate removal using elemental analysis isotope ratio mass spectrometry. Particle size distribution was analyzed using a laser diffraction particle size analyzer or via optical images. We analyzed water samples for DOC concentration using liquid-chromatography organic carbon detection, major ions by ion chromatography and inductively coupled plasma–optical emission spectrometry (ICP-OES), and dissolved rhenium concentration, a $\text{POC}_{\text{petro}}$ oxidation proxy (Dalai et al., 2002; Jaffe et al., 2002; Hilton et al., 2014), using ICP–mass spectrometry (see the Data Repository).

EVALUATING POC OXIDATION

POC loss during fluvial transit includes direct CO_2 and DOC production. We assume that any organic carbon loss from the solid phase represents long-term POC oxidation because terrestrial DOC is rapidly oxidized in the ocean (Bianchi, 2011). We quantified POC oxidation by measuring the percent change in POC mass over an experiment; i.e.,

$$\Delta M_c = \left[(M_{c_{\text{final}}} - M_{c_{\text{initial}}}) / M_{c_{\text{initial}}} \right] \times 100, \quad (1)$$

where $M_{c_{\text{initial}}}$ and $M_{c_{\text{final}}}$ are the solid POC mass at the experiment start and end, respectively (see the Data Repository). Solid load POC loss yields negative ΔM_c values.

DOC measurements provided additional POC loss constraints. We calculate the percentage mass fraction, f , of organic carbon transferred from the solid to the dissolved load as

$$f_{\text{DOC}} = (M_{\text{DOC}_{\text{max}}} / M_{c_{\text{initial}}}) \times 100, \quad (2)$$

where $M_{\text{DOC}_{\text{max}}}$ is the maximum DOC mass in the experiment (see the Data Repository). Using $M_{\text{DOC}_{\text{max}}}$ gives an upper limit on f_{DOC} ; however, f_{DOC} is a minimum constraint on total POC loss because DOC can be oxidized during the experiment.

Rhenium is typically associated with organic matter in sedimentary rocks and upon oxidation forms a soluble anion within the dissolved load, therefore $\text{POC}_{\text{petro}}$ oxidation during the experiment could result in increased dissolved rhenium concentrations (Hilton et al., 2014). We express the mass of rhenium transferred from the solid to dissolved load as

$$f_{\text{Re}} = (M_{\text{diss}_{\text{Re}}} / M_{\text{Re}_{\text{initial}}}) \times 100, \quad (3)$$

where $M_{\text{diss}_{\text{Re}}}$ is the accumulated dissolved rhenium mass during the experiment, and $M_{\text{Re}_{\text{initial}}}$ is the initial rhenium mass in the

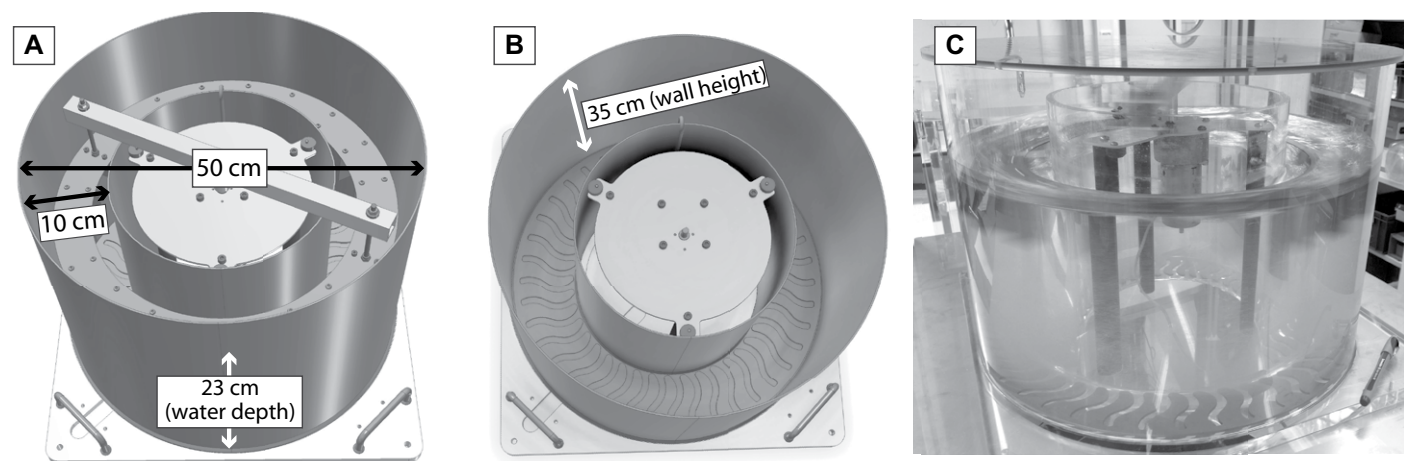


Figure 1. Schematic diagrams of stainless-steel flumes with (A) and without (B) paddle wheel visible. (C) Acrylic, but otherwise identical, flume used to monitor hydrodynamics with visible sediment transport.

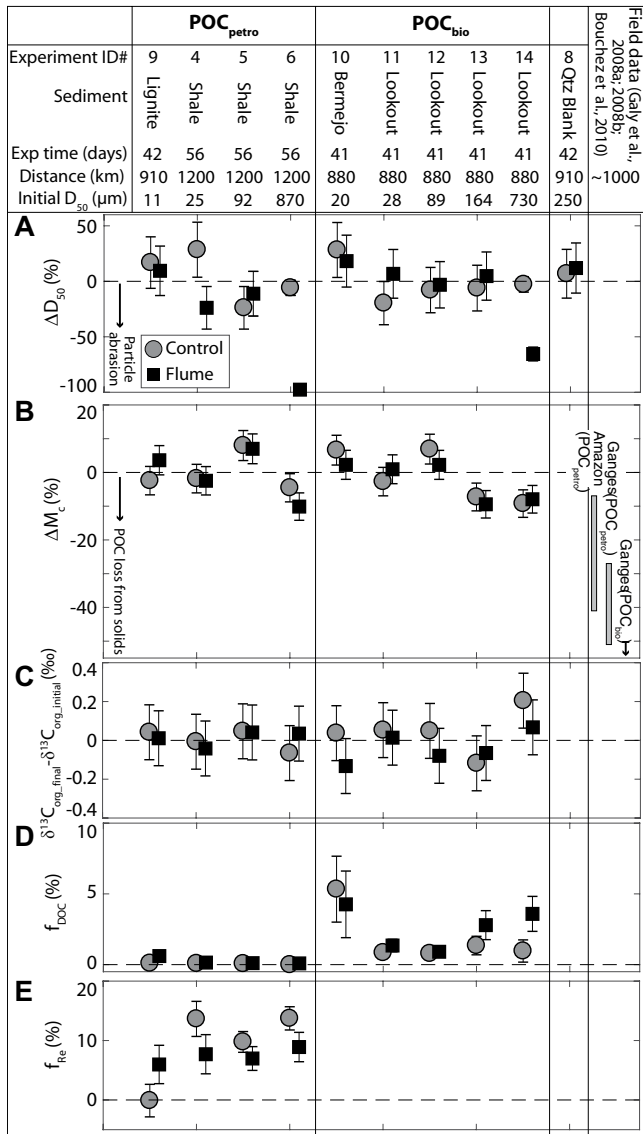


Figure 2. Comparison of (A) changes in median grain size ($\Delta D_{50} = [(D_{50, \text{final}} - D_{50, \text{initial}})/D_{50, \text{initial}}] \times 100$, where $D_{50, \text{final}}$ and $D_{50, \text{initial}}$ are median grain sizes at start and end of experiments, respectively); (B) changes in solid load particulate organic carbon (POC; ΔM_c , Equation 1); (C) $\delta^{13}\text{C}_{\text{org}}$; (D) changes in dissolved organic carbon (DOC; f_{DOC} , Equation 2); and (E) changes in dissolved rhenium (f_{Re} , Equation 3). Black squares and gray circles distinguish between flume experiments with sediment transport and static control tanks, respectively. Gray bars and black arrow in panel B show field estimates for POC oxidation over ~1000 km transport in the Amazon and Ganges Rivers (see the Data Repository [see footnote 1] for details). $\delta^{13}\text{C}_{\text{org, initial}}$ and $\delta^{13}\text{C}_{\text{org, final}}$ are $\delta^{13}\text{C}_{\text{org}}$ values at the start and end of experiments, respectively. Negative ΔD_{50} values indicate particle abrasion, and reduced ΔD_{50} uncertainty for experiments 6 and 14 is due to use of optical measurements permissible on larger grain sizes (see the Data Repository). Error bars represent the propagated uncertainty of measurements as reported in the Data Repository, and are smaller than symbols where not shown.

within error of or greater than ΔM_c values, consistent with f_{Re} as an upper limit on POC_{petro} loss.

Flume experiments showed no systematic difference in ΔM_c , f_{DOC} , and f_{Re} compared to their associated control runs (Fig. 2). For cases with initial median grain diameter (D_{50}) >500 μm, fluvial transport caused particle abrasion in flume experiments (Fig. 2A), yet flume and (abrasion-free) control ΔM_c and f_{Re} values were within error of each other or indicated increased oxidation in the control run, suggesting a minimal influence of particle abrasion on POC loss (Figs. 2B and 2E). Experiments with D_{50} <500 μm showed no detectable abrasion, likely due to viscous damping of particle impacts (Joseph et al., 2001; see also Table DR2 herein).

POTENTIAL FOR POC OXIDATION DURING IN-RIVER TRANSPORT

The lack of systematic differences between the flume and control experiments suggests an insignificant role of sediment transport and particle abrasion in POC oxidation relative to liquid-water leaching. The largest POC mass losses ($-8\% < \Delta M_c < -10\%$) occurred for $D_{50} > 125$ μm (Fig. 2), suggesting that POC oxidation rates may be more strongly influenced by whether and how carbon is bound to sediment, rather than by sediment transport processes or size (a proxy for particle SSA; Blair and Aller, 2012).

The maximum POC_{petro} losses measured in our experiments ($\Delta M_c = -10\% \pm 4\%$ and the proxy $f_{\text{Re}} = 14\% \pm 3\%$; Fig. 2) were small compared to field measurements in lowland river systems. Reported POC_{petro} losses between the mountain front and coastline in the Amazon and Ganges Rivers range from 7% to 50% per 10^3 km (see the Data Repository), i.e., up to ~5 times larger than the maximum POC_{petro} loss in our experiments (Fig. 2). These losses occur despite the prevalence of graphitized POC_{petro} in the Ganges, which is likely more resistant to oxidation than the disordered lignite used here (Galy et al., 2008a).

Experimental POC_{bio} losses were up to $\Delta M_c = -9\% \pm 4\%$. Unlike POC_{petro}, which is typically sourced from bedrock in river headwaters, POC_{bio} is sourced from vegetation and soils along the entire river length. The Amazon (Mayorga et al., 2005; Aufdenkampe et al., 2007) and Ganges (Galy et al., 2008b) basins show replacement of headwater-derived POC with floodplain POC_{bio}. Galy et al. (2008b) suggested >50% replacement of POC_{bio} over ~900 km of fluvial transit along the Ganges, i.e., ~5 times greater than the maximum loss in our experiments. We interpret the lack of large differences between flume and control experiments and consistently low POC losses indicated by multiple measurements (ΔM_c , f_{DOC} , and the proxy f_{Re}) to suggest that the physical processes in river systems, i.e., turbulent fluid mixing, sediment transport, and particle abrasion, cannot produce the magnitude

sediment (see the Data Repository). For $M_{\text{diss, Re}}$, we excluded an initial rapid period of change of ion concentrations (Re, SO_4^{2-} , and Ca^{2+}), which is inconsistent with the lowland river processes we sought to model (see the Data Repository). The value of f_{Re} provides a maximum constraint on POC_{petro} loss due to contributions of dissolved rhenium from non-POC_{petro} phases (Horan et al., 2019).

EXPERIMENTAL RESULTS

POC loss in all runs was small relative to measurement uncertainty. ΔM_c never fell below $-10.1\% \pm 4.1\%$ and $-9.4\% \pm 4.1\%$ in POC_{petro} and POC_{bio} experiments, respectively, and the majority of experiments showed no POC loss (ΔM_c greater than or within error of 0%; Fig. 2B). $\Delta M_c > 0\%$ (POC gain) was likely due to solid sample heterogeneity. Shifts in $\delta^{13}\text{C}_{\text{org}}$ values were typically within analytical uncertainty, suggesting that POC stable isotope composition stayed constant during sediment transport (Fig. 2C).

DOC production was small; f_{DOC} never exceeded 0.6% and 5.3% in the POC_{petro} and POC_{bio} experiments, respectively (Fig. 2D). For POC_{petro} experiments, after some initial water-sediment interactions over <1 d, shale experiments showed negligible DOC production relative to the background levels, while the lignite flume experiment showed steady DOC decrease (Fig. DR3; Table DR4). The POC_{bio} experiments showed a sharp DOC increase during the first 24 h, and then the rate slowed (Fig. DR3).

In the POC_{petro} experiments, dissolved ion concentrations increased rapidly over the first 1–4 d and then slowed (Fig. DR4). We attribute this either to ion exchange (Sayles and Mangelsdorf, 1979), leaching of ions from freshly exposed defect sites (White and Brantley, 2003), and/or rapid carbonate dissolution (Chou et al., 1989) forced by the interaction of fresh rock with a fluid out of equilibrium with the sediment. Accounting for this (see the Data Repository) yielded f_{Re} values that varied from <0% to $14\% \pm 3\%$ (Fig. 2E). These changes tended to be

of POC oxidation observed over equivalent transport distances in natural river systems.

IS POC OXIDIZED DURING IN-RIVER TRANSPORT OR DURING FLOODPLAIN STORAGE?

Despite field measurements showing large POC losses in lowland rivers (Richey et al., 2002; Galy et al., 2008a, 2008b; Bouchez et al., 2010), it remains unknown whether POC oxidation occurs primarily during active river transport or transient floodplain storage. In nine laboratory experiments, we observed no significant influence of $\sim 10^3$ km of sediment transport on POC oxidation. POC mass loss never exceeded $\Delta M_c = -10\%$, and 6 of 9 experiments showed POC loss within error of zero or POC gain, compared to field observations of 7% to >50% POC loss over equivalent transport distances. Chemical and biological processes not systematically explored in our experiments may increase POC oxidation during in-river transport (e.g., Battin et al., 2009; Druhan et al., 2014) and are potential avenues for future experimental study. However, by using sediment with native microbial communities and water similar in chemical composition to major rivers, our results suggest that POC oxidation in lowland rivers occurs primarily during floodplain storage rather than during in-river transport. POC oxidation is likely sensitive to the residence time of sediment in floodplains, as well as the rate and depth of burial, local redox conditions, and microbial dynamics, variables that should be systematically explored in future experiments, along with field work to build mechanistic models of POC transport and oxidation.

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