

## Article - postprint

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Evans, Chris D.; Thomas, David N. 2016. **Controls on the processing and fate of terrestrially-derived organic carbon in aquatic ecosystems: synthesis of special issue** [in special issue: Carbon cycling in aquatic ecosystems] *Aquatic Sciences*, 78 (3). 415-418. [10.1007/s00027-016-0470-7](https://doi.org/10.1007/s00027-016-0470-7)

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## **Controls on the processing and fate of terrestrially-derived organic carbon in aquatic ecosystems:**

### **Synthesis of special issue**

*Chris D. Evans and David N. Thomas*

### **Introduction**

Organic matter export to aquatic ecosystems represents an important pathway of carbon loss from terrestrial ecosystems. In peatlands, for example, fluvial losses of dissolved organic carbon (DOC) and particulate organic carbon (POC) may offset, or even outweigh, the net flux of carbon dioxide (CO<sub>2</sub>) from the atmosphere to the peat surface (e.g. Billett et al., 2004). At a continental European scale, it has been estimated that lateral carbon losses, via rivers, are of a similar magnitude to net carbon accumulation in forests (Ciais et al., 2008). In some areas, these fluxes are thought to be changing. In the industrialised regions of Northern Europe and Eastern North America, DOC concentrations in many surface waters have risen consistently since the 1980s, often by a factor of two or more, as these ecosystems have recovered from the historic impacts of acid deposition (Monteith et al., 2007). In addition, fluvial carbon losses have in some cases been exacerbated by climatic and/or land-use disturbances such as drainage, intensive agriculture and urbanisation (e.g. Moore et al., 2013; Frank et al., 2014; Hulatt et al. 2014a; Butman et al., 2015).

The consequences of fluvial organic matter fluxes, as well as their subsequent influence on the global carbon cycle, depend on their form, reactivity and ultimate fate. For terrestrial ecosystems, export of DOC and POC represent both a loss of carbon and, if this carbon is subsequently converted to CO<sub>2</sub> or methane (CH<sub>4</sub>), an indirect pathway for greenhouse gas (GHG) emissions. In freshwater ecosystems, terrestrially-derived dissolved and particulate organic matter (DOM and POM) provide a substrate for heterotrophic production (Hulatt et al. 2014b; Bouvier and del Giorgio, 2002), and the breakdown of this organic matter can release inorganic carbon, nitrogen and phosphorous, affecting the stoichiometry of the aquatic environment and rates of autotrophic production.

Dissolved organic matter strongly influences the aquatic light regime, regulating the depth of photosynthetic activity and balance of photosynthesis versus respiration (e.g. von Einem and Granéli, 2010; Kritzberg et al., 2014), as well as the thermal regime and mixing of lakes (e.g. Forsius et al., 2010). By forming complexes with a range of metals such as iron, aluminium and mercury, DOM can influence their mobility, physicochemical reactivity and toxicity (e.g. Gensemer et al., 1999; Clements et al., 2008), whilst photochemical reactions involving DOM have been shown to accelerate the degradation of organic pollutants such as pharmaceuticals (e.g. Santoke et al., 2012). In drinking water supplies, high levels of both DOM and POM are problematic, decreasing the effectiveness of conventional treatment methods and adding to energy, chemical and infrastructure-related costs of water treatment. In addition, sedimentation of POM in reservoirs reduces storage capacity and may increase the requirement for reservoir drawdown and dredging (Labadz et al., 1991). Finally, increased DOC and POC export from terrestrial ecosystems may ultimately lead to increased organic matter fluxes to estuarine and coastal waters, affecting the biogeochemistry and ecology of marine ecosystems (e.g. Hansell et al., 2004; Bianchi, 2011).

It is now generally recognised that aquatic systems do not simply act as passive 'pipes' for organic carbon transport, but are active zones of organic matter transformation, CO<sub>2</sub> emissions and carbon

burial (e.g. Cole et al., 2007; Battin et al., 2009; Tranvik et al., 2009). Despite this high level understanding, fundamental data on the reactivity of terrestrial organic matter in aquatic systems are in some cases sparse, or weighted towards particular water types or regions (for example high-latitude lakes or large river systems). Small headwater streams represent a key interface between the terrestrial and aquatic systems, may represent a significant fraction of the total river surface area (Bishop et al., 2008), and potential hotspots of organic matter processing (Battin et al., 2008), yet data to enable the extent of this processing to be quantified are sparse (Cole et al., 2007; Lauerwald et al., 2012). More generally, we still lack a comprehensive understanding of the fate of terrestrially-derived organic matter in different forms (e.g. dissolved versus particulate), derived from different sources (e.g. peatland versus forest or farmland), subject to different transformation processes (e.g. photo- versus bio-degradation) and subject to multiple environmental controls (e.g. light, nutrients, water residence times). As a result, our capacity to incorporate these processes into management decisions, mechanistic models, global flux estimates and site or regional greenhouse gas balances remains limited.

### **Aims and scope**

This special issue brought together a complementary set of studies describing the range of processes affecting the cycling and fate of terrigenous organic matter within aquatic systems. It focuses in particular on headwater streams, and on peat catchments as major global sources of freshwater DOM (Aitkenhead and McDowell, 2000), and of POM in areas of peat erosion (Worrall et al., 2011). This work is placed within the wider context of processes occurring in forested and agricultural headwaters, larger river systems and estuaries, and in a policy context in relation to national-scale carbon fluxes, the treatment of organic matter in drinking water supplies and the inclusion of aquatic carbon in international greenhouse gas accounting.

### **The papers**

A number of the papers in the special issue apply experimental approaches to quantify the rates of, and controls on, key organic matter transformation processes. Moody and Worrall (2015) focus on the photochemical reactivity of peat-derived organic matter, based on field experiments carried out under ambient light and dark conditions, and show high rates of light-mediated DOC loss, which are used to estimate total CO<sub>2</sub> emissions from UK rivers resulting from organic matter degradation. Jones et al. (2015a) compare rates of DOM photodegradation, and the fate of the carbon released, for a range of water types under controlled light exposure levels, using <sup>14</sup>C-labelled DOC as a tracer, with their results showing high photodegradation rates for peat-derived DOM. Stutter and Cains (2015) compare the biodegradability of DOM from a similar range of water types using a micro-plate respiration system, and show faster biodegradation rates for DOM draining forested and agricultural catchments, as well as apparent 'priming' of bed sediment decomposition by DOM in the water column. Ekström et al. (2015) quantify both the photo- and bio-degradability of DOM obtained from forest soils within an acidity manipulation experiment, and conclude that the magnitude and relative importance of these two processes is changing over time in response to rising DOC concentrations and declining acid deposition. Goulsbra et al. (2015) focus on the reactivity of peat-derived POC during short-term incubation experiments and provide evidence of rapid conversion of this material to CO<sub>2</sub> within 24 hours of contact with streamwater, suggesting possible 'hot moments' of carbon

release from the fluvial system during storm events, as well as more gradual, microbially-mediated POC to DOC conversion.

A second group of papers places these experimental studies into a wider context. Palmer et al (2015) present reach-, confluence- and estuarine-scale measurements of DOC and POC from a number of UK catchments, and suggest that the rapid, physico-chemically mediated removal of organic matter is both spatially and temporally limited in these systems. Raike et al. (2015) describe long-term trends in both DOC and dissolved inorganic carbon (DIC) at the scale of large river catchments in Finland, and show a tendency towards increasing concentrations of both DOC and DIC since the 1990s. They suggest that DIC increases could in part be linked to the mineralisation of increasing amounts of terrestrially-derived DOC within lakes. Kaartokallio et al. (2015) further consider the influence of DOM quantity and quality on its bioavailability within three Finnish estuaries discharging to the Baltic Sea. They provide evidence the rate of estuarine bacterial biomass turnover is affected by the quality of terrigenous DOM input from different types of catchment, and that populations of bacteria along estuarine gradients are influenced by DOM quality in different ways.

Finally, three papers consider the wider implications of terrestrial organic matter export and aquatic processing for ecosystem carbon and greenhouse gas balances. Barry et al. (2015) incorporate a range of aquatic carbon fluxes into the carbon balances of two Irish agricultural grasslands on organic soil. They demonstrate that these fluxes represent a quantitatively important carbon loss pathway at both sites, but further suggest that the conversion of these carbon losses to gaseous CO<sub>2</sub> emissions may be limited by the high solubility of inorganic carbon in high-pH waters, and by low measured biodegradability of DOM. Jones et al. (2015b) examine the fate of both POC and DOC through the drinking water treatment process at a number of water treatment plants, and combine these results with data on energy and chemical use to estimate overall greenhouse gas emissions associated with the removal of organic matter during treatment. In the final paper, Evans et al. (2015) describe, and expand on, a methodology developed to account for waterborne greenhouse gas emissions from peatlands as part of the Intergovernmental Panel on Climate Change Wetland Supplement (IPCC, 2014). This is the first time that aquatic greenhouse gas emissions have been included in international emissions reporting mechanisms such as the Kyoto Protocol.

## **Conclusions**

The work presented in this Special Issue provides new insights into the processing of terrestrially-derived organic matter within aquatic ecosystems. It emphasises the highly photo-reactive nature of DOM exported from peatlands, and the potential reactivity of all forms of terrigenous organic matter within headwater streams. It highlights the role of headwaters as the beginning of a continuum of terrestrial organic matter processing that extends through rivers, lakes, reservoirs and the drinking water supply system into estuaries and coastal waters, which collectively determine the fate of this organic matter, its ecological impacts, and its contribution to CO<sub>2</sub> fluxes. As a contribution to the growing body of literature in this field, it represents a further step towards the integration of freshwaters in studies of the global carbon cycle, and the incorporation of aquatic carbon in policy and land-management decisions, greenhouse gas emissions inventories, and earth system models.

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