



Aged riverine particulate organic carbon in four UK catchments



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HIGHLIGHTS

- Particulate organic radiocarbon values reflect mixtures of sources and processes.
- Similar radiocarbon values were obtained for 7 ruraly-dominated UK rivers.
- The 7 rivers export particulate organic carbon with an average age of 680 years.
- Mining and industrial activity result in the export of still older carbon.

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ABSTRACT

The riverine transport of particulate organic matter (POM) is a significant flux in the carbon cycle, and affects macronutrients and contaminants. We used radiocarbon to characterise POM at 9 riverine sites of four UK catchments (Avon, Conwy, Dee, Ribble) over a one-year period. High-discharge samples were collected on three or four occasions at each site. Suspended particulate matter (SPM) was obtained by centrifugation, and the samples were analysed for carbon isotopes. Concentrations of SPM and SPM organic carbon (OC) contents were also determined, and were found to have a significant negative correlation. For the 7 rivers draining predominantly rural catchments, PO¹⁴C values, expressed as percent modern carbon absolute (pMC), varied little among samplings at each site, and there was no significant difference in the average values among the sites. The overall average PO¹⁴C value for the 7 sites of 91.2 pMC corresponded to an average age of 680 ¹⁴C years, but this value arises from the mixing of differently-aged components, and therefore significant amounts of organic matter older than the average value are present in the samples. Although topsoil erosion is probably the major source of the riverine POM, the average PO¹⁴C value is appreciably lower than topsoil values (which are typically 100 pMC). This is most likely explained by inputs of older subsoil OC from bank erosion, or the preferential loss of high-¹⁴C topsoil organic matter by mineralisation during riverine transport. The significantly lower average PO¹⁴C of samples from the River Calder (76.6 pMC), can be ascribed to components containing little or no radiocarbon, derived either from industrial sources or historical coal mining, and this effect is also seen in the River Ribble, downstream of its confluence with the Calder. At the global scale, the results significantly expand available information for PO¹⁴C in rivers draining catchments with low erosion rates.

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Abbreviations: AMS, accelerator mass spectrometry; NRCF, NERC Radiocarbon Facility; OC, organic carbon; pMC, percent modern carbon absolute; POC, particulate organic carbon; POM, particulate organic matter; PO¹⁴C, particulate organic radiocarbon; SPM, suspended particulate matter; [SPM], concentration of SPM.

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1. Introduction

Particulate organic matter (POM) transported by rivers is defined as organic matter that does not pass a filter with sub-micron pore size, and mainly comprises allochthonous inputs from plant litter, soils and wastes, autochthonous phytoplankton and macrophyte debris, and in situ production from dissolved organic matter (Ittekkot and Laane, 1991). It plays a significant role in the carbon cycle, being a loss from the terrestrial environment, a source of C to the atmosphere due to decomposition during transit, and ultimately a gain to the marine

environment (Raymond and Bauer, 2001; Rosenheim and Galy, 2012; Worrall et al., 2014). Particulate organic carbon (POC) accounts for approximately 50% of the riverine global organic carbon export of 0.4 Pg a^{-1} (Schlünz and Schneider, 2000; Aufdenkampe et al., 2011). Particulate organic matter also governs the transport of the macronutrients nitrogen and phosphorus (Meybeck, 1982; Walling, 2005), metals (Tipping et al., 1997) and organic contaminants (Foster et al., 2000). To understand and quantify these POM-associated processes, and thereby predict how they might respond to changes in land use, climate and other environmental drivers, we need to delineate the sources of POM in different systems.

Because POM is part of suspended particulate matter (SPM), information about sources can be obtained from more general investigations into SPM, which is known to comprise a mixture of terrestrial material derived from both surface and sub-surface materials including bedrock and mineral soil (Blair et al., 2003). The SPM entering the river systems is generally the result of physical weathering and physical disturbance through anthropogenic activity. Sediment sourcing techniques, including mineralogy (Klages and Hsieh, 1975), chemistry (Gaillardet et al., 1999), magnetism (Gruszowski et al., 2003) and radionuclides (Walling and Woodward, 1992; Lu et al., 2014; Rosenheim and Galy, 2012) identify sources of SPM as being primarily topsoil and sub-surface material including erosion of exposed banks. Walling (2013) summarised sediment source information on 84 UK rivers, and showed that on average topsoil was the largest contributor of SPM, with relative contributions from surface material and channel banks contributing between 50% to 99% and <50% respectively.

The organic matter components of SPM have been studied by a variety of analytical techniques and with stable isotopes and element ratios (da Cunha et al., 2000; Onstad et al., 2000; Kendall et al., 2001; Higuera et al., 2014) to obtain information on molecular structure and insight into the materials from which the POM is derived. Determination of the radiocarbon content of POM provides further information, not only on sources but also apparent age. Radiocarbon gives a measure of the time elapsed since C fixation into plants from the atmosphere, providing an understanding of the residence time of plant-derived C and losses of C through mechanisms including leaching and erosion (Trumbore, 2009). Naturally occurring atmospheric ^{14}C can be used for the measure of C turnover on the centennial and millennial timescale due to the natural radioactive decay process, while “bomb carbon” originating from atmospheric weapons testing in the mid-20th century, which almost doubled the atmospheric ^{14}C levels (Hua et al., 2013), provides information on decadal timescales. Radiocarbon has been used in studies of POM in rivers in North and South America (Raymond and Bauer, 2001; Bouchez et al., 2014), Asia (Hilton et al., 2008; Rosenheim and Galy, 2012), Europe (Cathalot et al., 2013; Megens et al., 2001), and Africa (Marwick et al., 2015). From their recent comprehensive review and data collation, Marwick et al. (2015) showed that at the global scale the median age of riverine POC is approximately 1800 years, and that SPM in highly-eroding catchments is depleted in PO^{14}C and low in OC (organic carbon).

Marwick et al. (2015) also commented that there remain large regions for which no data are available, and their results show while there are many PO^{14}C data for catchments with high sediment loads, there are relatively few for riverine PO^{14}C in temperate, low-erosion European catchments and no known data for catchments of this kind in the UK. In the wider context, UK data may be of interest because UK soils tend to be rich in carbon, which may lead to differences from global averages. Therefore understanding UK sources should improve our ability to model the terrestrial-freshwater C cycle, and its role in transferring carbon to the atmosphere and ocean. To this end, we carried out a programme of sampling and analysis to determine the radiocarbon contents of POM from four differing UK catchments. Since SPM concentrations, and therefore POM transport, are elevated at high flow, where possible we focused our sample collection on high-discharge events. To

interpret the results, we made use of the extensive soil radiocarbon data available for the UK.

2. Methods

2.1. Field sites

Table 1 provides information on the individual rivers and their catchments, and their locations are shown in Fig. 1. Data on discharge and rainfall were taken from the National River Flow Archive (NRFA) (<http://www.ceh.ac.uk/data/nrfa/> accessed January 2015) and Met Office (<http://www.metoffice.gov.uk/industry/data/commercial/rainfall> accessed March 2015) respectively. Catchment areas were obtained from the CEH River and Catchment Query and Extraction Layer (Racquel) (<http://wlwater.ceh.ac.uk/racquel/> accessed March 2015). Geological information was provided by the British Geological Society online map (<http://mapapps.bgs.ac.uk/geologyofbritain/home.html>, accessed March 2015). Soil types for England and Wales were obtained using soil maps (scale 1:250 000). For the Dee catchments, soil information was obtained from The James Hutton Institute online soils map (http://sifss.hutton.ac.uk/SSKIB_Stats.php accessed March 2015). Land use data were obtained from the website <http://digimap.edina.ac.uk/> (accessed March 2015).

The Ribble catchment is situated in north-west England and has a population density of 989 persons km^{-2} . Two major sub catchments, the Rivers Hodder and Calder drain from the north and south of the catchment respectively. Unlike other tributaries of the Ribble, the Calder catchment contains extensive conurbations including Burnley and Blackburn, with a history of industrial and mining activity. The upper parts of the catchment are responsive to rainfall, exhibiting a flashy flow regime.

The River Conwy is one of the major drainage systems in North Wales. The catchment has a population density of 49 persons km^{-2} . The topography is largely mountainous, giving a high river response during storm events.

Situated in the south of England, the Hampshire Avon catchment has a population density of 108 persons km^{-2} . The catchment is largely groundwater dominated due to the presence of chalk aquifers. Thus, the system at the tidal limit does not significantly respond during rainfall events.

The River Dee catchment is situated in the north-east of Scotland. The catchment is sparsely populated above the tidal limit, with a population density of 4 persons km^{-2} . The tributary of the River Gairn is situated in the upper western reaches of the catchment. The upper mountainous areas respond rapidly to rainfall and snowmelt, producing a flashy flow regime.

2.2. River sampling and analysis

To minimise risk of carbon contamination, all equipment used during sampling and processing was new or acid-washed and all samples were managed in a radiocarbon tracer-free laboratory. Water samples (5 L) were collected in high-density polyethylene containers from the tidal limit of the four main catchments, and additional upstream samples were taken in the Ribble and Dee catchments (Fig. 1). For the Ribble, Conwy and Dee sites samples were collected during high-flow events. For the Avon, which has a much less flashy flow regime, they were taken at regular intervals throughout one year. High flow events were identified from daily river levels measured at gauging stations near the sampling sites, and made available on-line by the Environment Agency of England and Wales and the Scottish Environment Protection Agency. During the period October 2013–October 2014, four samples were taken for most of the sites, but only three each at the Gairn and Dee A sites. Additional 500 or 1000 mL samples were collected for the determination of SPM concentration and the carbon content of the SPM.

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