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Carbon isotopic characterisation of dissolved organic matter during water treatment

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ABSTRACT

Water treatment is a series of physio-chemical processes to aid organic matter (OM) removal, which helps to minimise the formation of potentially carcinogenic disinfection by-products and microbial regrowth. Changes in OM character through the treatment processes can provide insight into the treatment efficiency, but radiogenic isotopic characterisation techniques have yet to be applied. Here, we show for the first time that analysis of ^{13}C and ^{14}C of dissolved organic carbon (DOC) effectively characterises dissolved OM through a water treatment works. At the sites investigated: post-clarification, DOC becomes isotopically lighter, due to an increased proportion of relatively hydrophilic DOC. Filtration adds 'old' ^{14}C-DOC from abrasion of the filter media, whilst the use of activated carbon adds 'young' ^{14}C-DOC, most likely from the presence of biofilms. Overall, carbon isotopes provide clear evidence for the first time that new sources of organic carbon are added within the treatment processes, and that treated water is isotopically lighter and typically younger in ^{14}C-DOC age than untreated water. We anticipate our findings will precipitate real-time monitoring of treatment performance using stable carbon isotopes, with associated improvements in energy and carbon footprint (e.g. isotopic analysis used as triggers for filter washing and activated carbon regeneration) and public health benefits resulting from improved carbon removal.

1. Introduction

Provision of a sufficient volume of appropriately treated water, free from potentially harmful chemical and microbiological contaminants is a fundamental requirement for human life. Treatment of surface water sources for potable supply routinely comprises a series of physical, chemical and biological processes designed to remove impurities to produce product water in line with local legislative standards. Whilst exact design and operational details will be determined by raw water quality characteristics, a surface water treatment works (WTW) will generally involve screening of gross solids, coagulant addition to precipitate and destabilise negatively charged colloidal matter, slow mixing to encourage collision and agglomeration of destabilised particles into flocs, settlement of flocs in a clarification stage, granular media filtration,

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adsorption of pesticide, natural organic matter, and taste- and odour-causing molecules, and disinfection of residual micro-organisms. However, the use of chlorine, ozone, or chlorine dioxide as a disinfectant in water rich in organic matter (OM) can lead to the occurrence of potentially carcinogenic disinfection by-products (DBPs). Consequently, water companies must manage the competing needs of biological and chemical compliance; *i.e.* the risk of DBP toxicity must be weighed against the certainty that water that has not been disinfected can cause illness and even death.

Water and wastewater treatment are resource-intensive processes; latest figures suggest that the global annual treatment of 1166 $km^3\ yr^{-1}$ for domestic and industrial use (30% of total global abstraction) uses approximately 1,420,030 GW h of energy and emits 1.21 Pg $CO_2e\ yr^{-1}$. (UNESCO, 2009; EPRI, 2002; WaterUK, 2010); see Fig. 1. This is equivalent to approximately 3.6% of annual anthropogenic carbon emissions, and is 45% of the total carbon that is transported, mineralised and buried in inland waters (Battin et al., 2009). The energy consumption of the water industry has increased significantly in the last 30 years, primarily in response to tightened legislation and regulation surrounding treatment of raw water and the discharge of final effluent from sewage treatment works to watercourses, and the corresponding increase of more energy-intensive processes (Reiling et al., 2009). Furthermore, many water companies have targeted themselves with carbon neutrality in the forthcoming 25 years. Thus, the urgent drive for more sustainable solutions and process improvements to existing solutions is clear. The use of chemicals in water treatment is widespread, being required for coagulation, as a flocculant aid, for pH correction and for disinfection (0.074 tonnes of chemical per megalitre (10^6 L) of drinking water produced (WaterUK, 2010)). This further compounds the urgent need for identification and elimination of process inefficiencies. Carbon accounting at the unit process level can help facilitate the development of new carbon-efficient technologies.

Pressure on water resources has led to the need to utilise sources with higher organic matter concentrations. Organic matter removal at WTWs is necessary, yet complex, and occurs in the clarification, filtration and adsorption stages of treatment. There is a significant body of research characterising 'raw' water organic matter (for example, see Gjessing et al., 1999), and considering the implications for treatment and removal (Rizzo et al., 2004; Kim and Yu, 2005; Fearing et al., 2004). However, our understanding of the composition and stability of aquatic dissolved organic matter which is being removed during the water treatment process has undergone rapid revision in recent years, with our current understanding that organic carbon is processed in-stream, both by biological and physiochemical processes. Key evidence was provided by the observation that riverine DOC is relatively 'young' in radiocarbon age (Raymond and Bauer, 2001); a finding that has since been replicated in numerous rivers (Evans et al., 2007; Mayorga et al., 2005; Benner et al., 2004). Rivers are now seen as organic carbon processors, with the microbial loop utilising previously unavailable soil carbon (Ward et al., 2013). Riverine DOC is therefore repeatedly reprocessed and 'young' in radiocarbon age by the time it reaches the oceans (Battin et al., 2009), with recent evidence that bacterial and microbial processing can commence within minutes (Pollard and Ducklow, 2011). In rivers draining peatlands, observations of rising dissolved organic carbon concentrations have created concerns that those stores are beginning to destabilise, with an associated increase in DOC entering reservoirs (Freeman et al., 2001, 2004). Consequently,

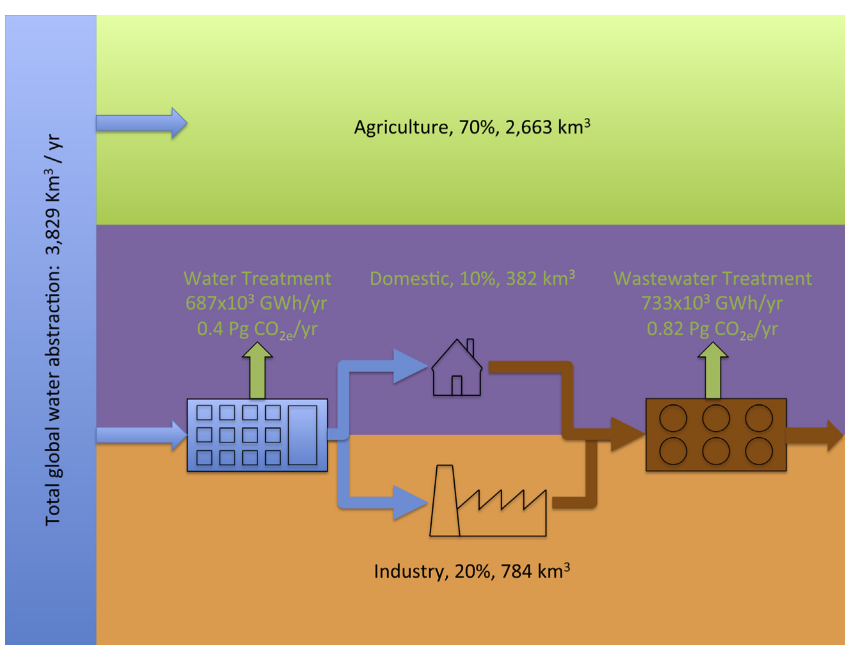

Fig. 1 – Energy consumption and CO_{2e} emissions associated with global water and wastewater treatment. (Abstraction figures from UNESCO (2009). Energy calculations based on 48.22% surface water at 0.371 kWh/m^3, 48.23% groundwater at 5 kWh/m^3 and 3.55% desalination at 5 kWh/m (EPRI, 2002; WaterUK, 2010). CO_{2e} emissions based on 0.34 and 0.7 tonnes/Ml for water and wastewater treatment respectively (WaterUK, 2010).