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## Evidence for anthropogenic $^{14}\text{C}$ -enrichment in estuarine waters adjacent to the North Sea

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[1] The isotopic composition ( $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$ ) of high molecular weight dissolved organic carbon (HMW DOC) was studied in the Tyne and Tweed estuaries, NE England. Despite significant removal of terrigenous HMW DOC in the low salinity regions ( $S < 15$ ),  $\Delta^{14}\text{C}$  remained modern with little variation around 115‰. This lack of apparent age discrimination was attributed to either non-oxidative removal or the absence of a significant proportion of old refractory C in the HMW DOC pool. At  $S > 15$ , we observed seaward increases in  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$ . With no documented local  $^{14}\text{C}$  inputs, we attributed non-bomb related  $^{14}\text{C}$ -enrichment at  $S > 15$  to a possible ‘lingering effect’ of distal anthropogenic sources in near-coastal North Sea HMW DOC. Given the global distribution of potential sources, we propose that anthropogenic  $^{14}\text{C}$  should be considered in assigning ages of DOC pools in near-coastal waters and suggest its possible use as a tracer for DOC transformations. **Citation:** Ahad, J. M. E., R. S. Ganeshram, R. G. M. Spencer, G. Uher, P. Gulliver, and C. L. Bryant (2006), Evidence for anthropogenic  $^{14}\text{C}$ -enrichment in estuarine waters adjacent to the North Sea, *Geophys. Res. Lett.*, 33, L08608, doi:10.1029/2006GL025991.

### 1. Introduction

[2] The  $^{14}\text{C}$  content of estuarine and near-coastal dissolved organic carbon (DOC) may be affected by aged riverine DOC, modern DOC which includes  $^{14}\text{C}$  from atmospheric nuclear weapons testing (post-AD 1955), and radioactive discharges from biomedical and nuclear fuel cycle related establishments. However, understanding the relative contributions of these inputs to  $^{14}\text{C}$ -DOC in near-coastal waters remains challenging. While some rivers are dominated by DOC with apparent radiocarbon ages of 100s to 1000s of years, most rivers discharging into the Atlantic Ocean have freshwater  $\Delta^{14}\text{C}$ -DOC signatures suggestive of modern, decadal-old DOC [Raymond and Bauer, 2001a, 2001b]. This modern  $\Delta^{14}\text{C}$ -DOC signature is believed to contain both labile and refractory components. Preferential microbial utilization of the younger, more labile DOC

fraction during estuarine transport may therefore lower the  $\Delta^{14}\text{C}$  of bulk DOC, resulting in the net export of  $^{14}\text{C}$ -depleted, degraded terrigenous DOC into the ocean [Raymond and Bauer, 2001a; 2001b].

[3] The  $\Delta^{14}\text{C}$  of atmospheric  $\text{CO}_2$  has been declining steadily after reaching a maximum in the N Hemisphere of  $\sim 1000$ ‰ at the height of nuclear weapons tests in the mid 1960s [Nydal, 1998]. Consequently,  $\Delta^{14}\text{C}$  signatures of terrestrially fixed C that are significantly above the modern atmospheric  $\Delta^{14}\text{C}$ - $\text{CO}_2$  value [ $\sim 100$ ‰ in AD 1998; Levin and Kromer, 2004] but  $< 1000$ ‰ can be attributed to a sample age between AD 1955 and the present [Mayorga et al., 2005; Raymond and Bauer, 2001a]. Weapons testing related  $^{14}\text{C}$ -enrichment in surface ocean dissolved inorganic carbon (DIC) did not reach atmospheric levels because of atmospheric exchange and mixing with the large reservoir of  $^{14}\text{C}$ -depleted deep ocean DIC. As a result, surface ocean  $\Delta^{14}\text{C}$ -DIC reached bomb  $^{14}\text{C}$  maxima of only  $\sim 270$ ‰ and have since declined to modern values  $< 100$ ‰ [Nydal, 1998].

[4] In some coastal areas, however, recurring anthropogenic  $^{14}\text{C}$  releases may lead to  $\Delta^{14}\text{C}$  values which are significantly higher than the modern background signal. In this study we report elevated  $\Delta^{14}\text{C}$  signatures in high molecular weight (HMW) DOC collected in two North Sea estuaries, the Tyne and Tweed (NE England), with no previously documented sources of local anthropogenic  $^{14}\text{C}$  inputs. We discuss the behavior of DOC during estuarine mixing and potential sources and consequences of this  $^{14}\text{C}$ -enrichment for coastal C cycling research.

### 2. Study Sites and Methods

[5] The Tyne and Tweed (Figure 1) are mesotidal estuaries receiving humic-rich water from forested and moorland catchments at mean discharge rates of  $48 \text{ m}^3 \text{ s}^{-1}$  and  $84 \text{ m}^3 \text{ s}^{-1}$ , respectively. The area around the Tyne estuary is densely populated. In contrast, the Tweed drains a rural, sparsely populated catchment dominated by agricultural activity. Near-surface water from a submersible pump (1–2 m depth) was collected into 25 L carboys on single day excursions to the Tyne (Jul-02, Mar-03, Jul-03) and Tweed (Jul-03, Dec-03) estuaries. Water samples were sequentially filtered through  $0.7 \mu\text{m}$  glass fibre (Whatman, GF/F) and  $0.2 \mu\text{m}$  capsule filters (Whatman, Polycap 75 TF) to remove particulates, algae and most bacteria. HMW DOC was 30-fold concentrated via tangential-flow ultrafiltration (UF) using a cassette filter system with stainless steel impeller pump (Millipore, Pellicon-2) and regenerated cellulose membrane (Millipore, Cat. P2PLACC05). The molecular size cut-off (90% retention) of the membrane was determined at 15.5 kDa. Mass balances indicated DOC recovery

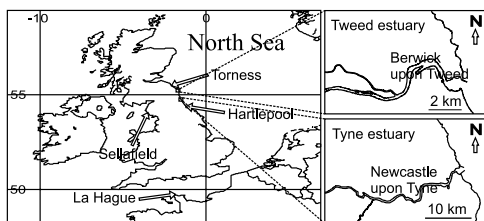
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**Figure 1.** Map of the North Sea showing the location of the Tyne and Tweed estuaries and several known or potential sources of anthropogenic  $^{14}\text{C}$  discharge.

of 90.2–107.5%, signifying minimal loss or contamination. The HMW component comprised  $\sim 3$ –5% and  $\sim 20$ –50% of the total DOC in coastal North Sea and riverine samples, respectively. Aliquots for HMW DOC analysis were acidified (pH 2), stored frozen in 20 ml glass vials and measured on a Shimadzu TOC-5000 analyzer after shaking thoroughly. The coefficient of variance of replicates was better than  $\pm 2\%$ .

[6] After UF, HMW DOC samples were diafiltered with Milli-Q water to remove salts, lyophilized, homogenized, re-wetted, decarbonated with HCl, and dried prior to isotopic analysis.  $\delta^{13}\text{C}$  signatures were determined using a Carlo Erba NA2500 elemental analyzer coupled to a Micromass PRISM III isotope ratio mass spectrometer. Based on replicate sample analyses the  $1\sigma$  precision for  $\delta^{13}\text{C}$  values was  $\pm 0.3\text{‰}$ . Radiocarbon analyses were carried out at the SUERC AMS facility (5MV NEC). Samples were combusted to  $\text{CO}_2$  at  $900^\circ\text{C}$  using CuO for 8 h in sealed quartz tubes or at  $600^\circ\text{C}$  using high purity  $\text{O}_2$  for 4 h. In the case of the latter, the resultant gases were then passed through a Pt/Ag furnace at  $900^\circ\text{C}$ .  $\text{CO}_2$  was cryogenically separated and converted to graphite targets by Fe/Zn reduction [Slota *et al.*, 1987]. Standard material of known ages added to ultrafiltered Milli-Q water and subjected to the sample protocol described above (lyophilization, etc.) showed minimal  $^{14}\text{C}$  contamination. Replicate analyses of riverine end-members, salinity ( $S \leq 0.2$ ), from the Tyne (Mar-03) and Tweed (Dec-03) indicated a  $1\sigma$  precision for  $\Delta^{14}\text{C}$  values [Stuiver and Polach, 1977] of  $\pm 7\text{‰}$ . For further details on the HMW DOC component and a complete list of data reported here please refer to the auxiliary material<sup>1</sup>.

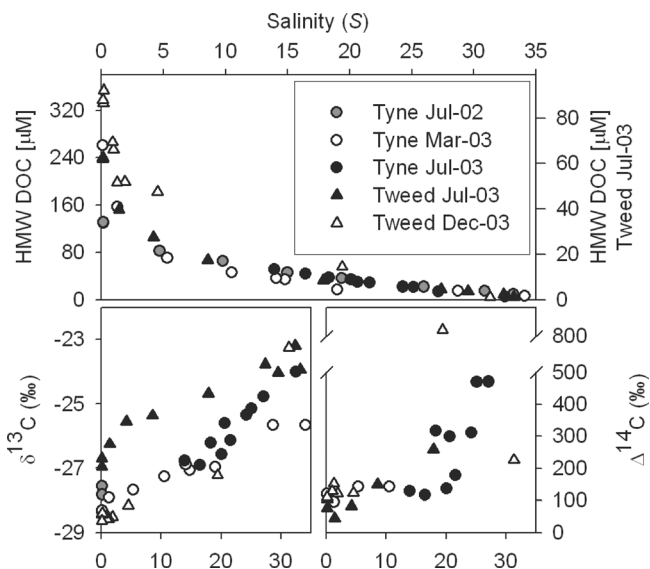
### 3. Results and Discussion

[7] HMW DOC concentrations varied by more than two orders of magnitude, ranging from 5.2–261.1  $\mu\text{M}$  and 1.3–354.0  $\mu\text{M}$  in the Tyne and Tweed estuaries, respectively (Figure 2). All concentration profiles were concavely shaped at low  $S$  ( $< 10$ ) and showed an approximately linear decrease at  $S > 10$ , indicating  $\sim 30$ –70% removal of terrigenous HMW DOC in the upper reaches of both estuaries. This was reflected in the total DOC as losses of up to  $\sim 10\%$ . Estuarine DOC removal was previously attributed to flocculation [Sholkovitz *et al.*, 1978], adsorption onto suspended particulates [Uher *et al.*, 2001], and bacterial and photochemical oxidation [Amon and Benner, 1996]. Regardless of the mechanisms responsible, our data

clearly indicate significant, non-conservative reductions in terrigenous HMW DOC, favoring the predominance of marine over terrestrial HMW DOC in near-coastal waters. In contrast, low molecular weight (LMW) DOC showed more or less conservative mixing behavior in these estuaries (auxiliary material). This suggests that the reactivity of terrestrial organic matter (OM) during estuarine mixing decreases along a size continuum from HMW to LMW DOC, as inferred in open ocean settings [Skoog and Benner, 1997; Loh *et al.*, 2004].

[8] The prevalence of North Sea HMW DOC in the outer Tyne and Tweed estuaries is substantiated by axial profiles of  $\delta^{13}\text{C}$  (Figure 2). All transects showed progressive  $^{13}\text{C}$ -enrichment with increasing  $S$ , consistent with mixing between peat- and moorland-derived HMW DOC (River Tyne:  $\delta^{13}\text{C} = -27.6$  to  $-28.4\text{‰}$ ; River Tweed:  $\delta^{13}\text{C} = -26.7$  to  $-28.6\text{‰}$ ) and heavier marine-derived HMW DOC. The  $\delta^{13}\text{C}$  of marine end-members ( $-25.7\text{‰}$ , Tyne Mar-03;  $-24.0\text{‰}$ , Tyne Jul-03;  $-23.9\text{‰}$ , Tweed Jul-03;  $-23.3\text{‰}$ , Tweed Dec-03) fell toward the lower end of the range reported for North Sea DOC [ $-18$  to  $-24\text{‰}$ ; Le Clercq *et al.*, 1997].

[9] The  $\Delta^{14}\text{C}$  of riverine HMW DOC ( $S \leq 0.2$ ) ranged between 76–121 $\text{‰}$  (Figure 2). Our data therefore suggest that UK rivers draining peat and moorland catchments are a source of modern HMW DOC, as reported for various DOC-rich Arctic [Benner *et al.*, 2004] and Amazonian [Mayorga *et al.*, 2005] rivers. Despite substantial HMW DOC removal  $\Delta^{14}\text{C}$  showed little variation ( $115 \pm 29\text{‰}$ ,  $n = 16$ ) at low to mid  $S$  ( $< 15$ ). This is in contrast to the behavior of  $\Delta^{14}\text{C}$  of total DOC reported in the York River estuary (Virginia, USA) by Raymond and Bauer [2001a]. In this case a downriver decrease in  $\Delta^{14}\text{C}$ -DOC of  $\sim 200\text{‰}$  across the low  $S$  range was attributed to the preferential utilization of younger, more labile fractions during bacterial degradation. This contrasting behavior of the two size classes of DOC again illustrates the size dependency of age and reactivity for various C pools. The higher size fraction of



**Figure 2.** Concentrations [ $\mu\text{M}$ ],  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  (‰) of HMW DOC in the Tyne and Tweed estuaries plotted versus salinity ( $S$ ).

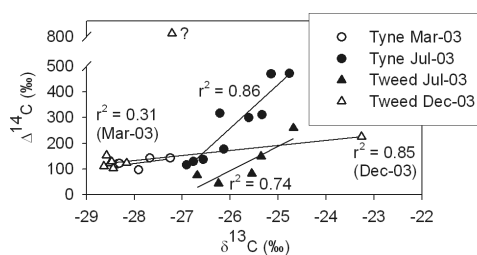
<sup>1</sup>Auxiliary material is available at <ftp://ftp.agu.org/apend/g/l/2006gl025991>.

terigenous DOC considered here is likely to be much more homogenous in age than total DOC examined elsewhere, and thus may have contained only minor amounts of older C, masking any age discrimination effects in the residual fraction. It is likely that the older components were contained mainly within the LMW DOC pool, as was observed in DOC from the open Atlantic and Pacific oceans [Loh *et al.*, 2004]. It is also possible that the large removals of HMW DOC found in the Tyne and Tweed were caused mainly by non-oxidative processes such as flocculation and adsorption, which may not discriminate against age.

[10]  $\Delta^{14}\text{C}$  of HMW DOC broadly increased toward the North Sea (Figure 2), reaching values of up to 471‰ (Tyne) and 811‰ (Tweed) at mid to high  $S$  ( $>15$ ). These  $\Delta^{14}\text{C}$  values were much greater than those previously reported ( $-200$  to  $\sim 100$ ‰) for DOC sampled across similar salinities in other temperate estuaries, and were contrary to the general trend of decreasing  $\Delta^{14}\text{C}$  with increasing  $S$  attributed to mixing with older marine DOC or the preferential utilization of younger DOC fractions [Guo and Santschi, 1997; Raymond and Bauer, 2001a]. Adopting the following two lines of reasoning, our estuarine  $\Delta^{14}\text{C}$  data may be attributed to anthropogenic  $^{14}\text{C}$  inputs into coastal North Sea waters that are unrelated to nuclear weapons testing. Firstly, the values of our near-coastal HMW DOC in most cases were significantly higher than the peak weapons testing DIC values in the N Atlantic and cannot be explained either by modern DIC or potential bomb  $^{14}\text{C}$  memory effects. Secondly, estuarine HMW DOC removal in conjunction with seaward increases in  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  precludes a riverine origin of  $^{14}\text{C}$ -enriched HMW DOC in the near-coastal North Sea.

[11] To confirm the marine origin of  $^{14}\text{C}$ -enrichment, we plotted  $\delta^{13}\text{C}$  against  $\Delta^{14}\text{C}$  (Figure 3). These plots yielded statistically significant positive linear relationships during the Jul-03 surveys to the Tyne ( $r^2 = 0.86$ ,  $n = 9$ ) and Tweed ( $r^2 = 0.74$ ,  $n = 5$ ), indicating mixing between a  $^{13}\text{C}$ - and  $^{14}\text{C}$ -enriched (marine) and a  $^{13}\text{C}$ -depleted modern (terrestrial) end-member. A weaker correlation was obtained for Mar-03 ( $r^2 = 0.32$ ,  $n = 5$ ), perhaps due to the low  $S$  bias for these samples. The Dec-03 Tweed survey is subject to one anomalously high  $\Delta^{14}\text{C}$  value (811‰,  $S = 19.38$ ; denoted by question mark) which has not been included in the regression ( $r^2 = 0.85$ ,  $n = 7$ ) plotted in Figure 3. But given that the low  $S$  samples during this survey did not exhibit any significant  $^{14}\text{C}$ -enrichment, it is unlikely that terrigenous OM contributed to this sample's elevated  $\Delta^{14}\text{C}$ . Ignoring this one anomalous value, the mixing trends shown in Figure 3 collectively indicate a marine origin of  $^{14}\text{C}$ -enriched HMW DOC.

[12] The largest documented source of anthropogenic  $^{14}\text{C}$  to the marine environment in the UK is the Sellafield nuclear fuel reprocessing plant (Cumbria, NW England; Figure 1), whose discharges are primarily in the form of DIC [Cook *et al.*, 2004]. As a result of this  $^{14}\text{C}$ -enriched DIC [ $\Delta^{14}\text{C} \sim 18000$ ‰; Cook *et al.*, 2004] entering the food chain via algal photosynthesis, significantly elevated  $^{14}\text{C}$  levels (100s to 1000s  $\text{Bq kg}^{-1}\text{C}$ ) were reported for particulate organic carbon (POC) and marine biota collected from the NE Irish Sea [Cook *et al.*, 1998, 1995]. Sellafield discharge spreads along the NW British coast and reaches the waters off eastern England after transport times of 3–5 a



**Figure 3.**  $\delta^{13}\text{C}$  and  $\Delta^{14}\text{C}$  (‰) of HMW DOC in the Tyne and Tweed estuaries plotted with their linear regressions. The one anomalously  $^{14}\text{C}$ -enriched sample (?) is not included in the Dec-03 regression.

[Otto *et al.*, 1990]. It may therefore be plausible to assume a diluted, Sellafield-derived  $^{14}\text{C}$  contribution to the near-coastal North Sea.

[13] An additional source of anthropogenic  $^{14}\text{C}$  into the North Sea is discharge from nuclear reprocessing at the La Hague plant in Normandy, France (Figure 1). This discharge enters the North Sea via the Straits of Dover and reaches SE England after 1–4 a [Otto *et al.*, 1990]. La Hague discharge was suggested as the likely source for  $^{14}\text{C}$ -enriched POC sampled off the Dutch coast in 1994 [Megens *et al.*, 2001], consistent with recent reports of elevated  $\Delta^{14}\text{C}$  (up to 700‰) in DIC and POC collected near La Hague [Douville *et al.*, 2004]. Localized but recurring  $^{14}\text{C}$ -enrichments have also been found in DIC (450–496‰), mussels (2534–4048‰), and seaweed (320–339‰) collected near the Hartlepool advanced gas-cooled reactor (AGR) nuclear power station, located  $\sim 40$  km south of the Tyne estuary (Figure 1), in 1998/9 surveys [Gulliver *et al.*, 2004]. Similarly, samples collected near Torness AGR, a reactor with a similar design to Hartlepool AGR and located  $\sim 35$  km NW of the Tweed (Figure 1), also showed enhanced  $^{14}\text{C}$  activities, albeit only marginally above modern levels [Gulliver *et al.*, 2004].

[14] Most anthropogenic  $^{14}\text{C}$ -enrichments previously reported in the literature occurred in the vicinity of known or potential sources. One notable exception is the value of 210‰ reported for HMW DOC sampled at the Frisian Front (southern North Sea) in 1996 [van Heemst, 2000]. The  $^{14}\text{C}$ -enrichments of marine HMW DOC observed by us occurred in two distinct estuaries with no documented  $^{14}\text{C}$  releases over two seasons and hence may not be the sole result of a local, single point source. Furthermore, POC collected during these surveys was significantly older [ $\Delta^{14}\text{C} = -465$  to  $-71$ ‰; Ahad, 2005], suggesting that DOC release from the local POC pool was not the source of elevated  $^{14}\text{C}$  in HMW DOC. This consideration and the mixing behavior in Figure 3 allude to an external marine source for enhanced  $^{14}\text{C}$  activities.

[15] One feature of the elevated  $^{14}\text{C}$  found in samples near Hartlepool and in the Irish Sea is the general pattern of greater enrichment in marine biota compared to DIC. An exception is seaweed, in which lower  $^{14}\text{C}$  activities can be attributed to its partial incorporation of atmospheric  $\text{CO}_2$  [Cook *et al.*, 2004, 1998]. Higher  $^{14}\text{C}$  activities in OM relative to concurrent DIC may be explained by the 'lingering effect' of the  $^{14}\text{C}$  signal once it enters the food chain. Filter feeders such as mussels attain elevated  $\Delta^{14}\text{C}$  from

their food source (i.e., planktonic detritus), maintaining this signal through episodic <sup>14</sup>C-enriched inorganic releases [Cook et al., 1998]. In contrast, the <sup>14</sup>C-enriched signal in the DIC is lost fairly quickly by mixing and dilution with the large pool of older marine DIC and via atmospheric equilibration.

[16] This 'lingering effect' of anthropogenic <sup>14</sup>C may also explain the <sup>14</sup>C-enrichment in marine HMW DOC encountered in the Tyne and Tweed estuaries. Firstly, unlike POC, which can settle out, DOC in general can be advected over larger distances. Recent estimates indicate that the turnover time of the bulk of HMW DOC is likely to be in the order of a few decades [Loh et al., 2004], although some labile components produced during spring phytoplankton blooms can break down much more rapidly [e.g., Amon and Benner, 1994]. The anthropogenic <sup>14</sup>C signal could therefore remain over these time scales on entering the HMW DOC pool. Secondly, this signal is less prone to dilution by mixing with aged HMW DOC from deeper waters because HMW DOC concentrations typically exhibit decline with depth [e.g., Loh et al., 2004]. This combination of factors – propensity to advect, decadal turnover time, less susceptibility to dilution – may allow anthropogenic <sup>14</sup>C incorporated in HMW DOC to travel relatively long distances away from the initial source.

[17] In the North Sea for instance, Sellafield discharge can reach the mouth of the Tyne and Tweed estuaries in 3–5 a [Otto et al., 1990], which is shorter than the presumed turnover time of the bulk of marine HMW DOC. The advection time is expected to be even shorter for other potential <sup>14</sup>C sources due to their relative proximity (Figure 1). Therefore, it is conceivable that the coastal North Sea HMW DOC pool integrated its <sup>14</sup>C signal from multiple non-bomb sources. On entering the Tyne and Tweed estuaries, this signal remained elevated above background <sup>14</sup>C levels despite incurring partial losses by mixing and turnover during advection. Our finding that the HMW DOC pool may act as the vector for transport of anthropogenic <sup>14</sup>C to areas distal from its initial source has broad implications. The diffuse signal resulting from anthropogenic <sup>14</sup>C releases may lead to an underestimation of DOC ages far from the point of discharge. It may also be possible to exploit elevated <sup>14</sup>C activities in affected environments as a tracer for C transfer between various DOC size fractions.

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