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Fate of terrestrial carbon in the Scottish coastal environment

Final Report

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Each year approximately 400 - 430 x 10^{12}g of terrestrial organic carbon is transported from the continents via rivers to the global ocean. Yet it is estimated that only a very small fraction of the organic carbon dissolved in the ocean, or preserved in underlying sediments, seem to be of terrigenous origin, with about 10% of the riverine input of organic carbon (i.e. 43 x 10^{12}g) actually buried in shelf sediments each year. So, the question is where does the remaining 90% of the terrestrial organic carbon go?

Key findings

- Most terrestrial organic carbon is highly modified and/or removed via natural processes in estuaries and near-shore environments before reaching the ocean environment
- Shelf and coastal sediments are one of the most important sinks for carbon on earth Scottish estuaries (e.g. sea lochs) are net sinks for organic carbon, either through respiration back to CO_2 or by long term burial in deeper anoxic sediments, and only a relatively small fraction is exported further to the sea.
- Scottish estuaries are intense biogeochemical recycling zones for organic carbon, of both marine and terrestrial origin. Within the estuary the terrestrial organic carbon can be subject to multiple fates including aggregation, photo-degradation, respiration and burial.
- The residual sedimentary organic carbon that is not respired by the benthos will eventually be preserved (i.e. buried) in deeper anoxic sediments and hence removed from the marine carbon cycle over potentially geological timescales.
- Sea loch sediments are efficient in sequestering organic carbon, making them important long stores for carbon, comparing well to other important coastal 'blue carbon' sinks such as saltmarshes and sea-grass meadows.
- Climate change and land use (i.e. peatland drainage) are likely to increase the export of terrestrial particulate and dissolved organic carbon to Scottish estuaries in the future

Key words

Organic carbon, burial, carbon sequestration, respiration, sedimentation, sea loch, estuary, organic matter, aggregation, photo-degradation

1.0 INTRODUCTION

This paper responds to a CREW call down request submitted by Scottish Natural Heritage.

Fate of terrestrial carbon in the Scottish coastal environment enquiry
What does current evidence tell us about how much terrestrial carbon is lost when it reaches the marine environment (and how much is stored in marine ecosystems)?

A short review is required to provide a summary of current evidence on the nature and fate of carbon in Scotland's coastal system with specific attention to processes that control the change in stocks. In particular, the review should include any evidence about how carbon moving from the terrestrial ecosystem reaches the coastal environment, how much is retained in the coastal system, and how much is lost into the marine system.

1.1 Nature of carbon in Scottish coastal systems

Carbon in the aquatic environment (lakes, rivers and sea) can occur in both inorganic- and organic forms. Dissolved Inorganic Carbon (DIC) occurs as bicarbonate (HCO_3^-) and to a lesser extent, as carbonate (CO_3^{2-}) in natural waters. The majority of the DIC in the aquatic environment is supplied by carbon dioxide (CO_2) from the atmosphere diffusing into the hydrosphere, where it reacts with water to form bicarbonate and carbonate ions as per Equation 1:

$$CO_2(aq) + H_2O \leftrightarrow H_2CO_3(aq) \leftrightarrow H^+(aq) + HCO_3^-(aq) \leftrightarrow 2H^+ + CO_3^{2-}(aq) \qquad \text{(Eq. 1)}$$

This set of equilibrium reactions is referred to as the aquatic carbonate system (Libes, 2009). DIC can also be generated from respiration in bacteria and higher aquatic organisms, producing CO_2 which again can be incorporated into the carbonate system. Particulate inorganic carbon (PIC) is formed in the aquatic environment through biologically mediated production of calcium carbonate, for example in the exoskeletons of certain types of phytoplankton or in the shells of mollusks and bivalves.

The source of fluvial inorganic carbon (FIC) is mainly from weathering of carbonate rich bedrock (e.g. limestone rock). FIC is primarily transported as dissolved bicarbonate (HCO_3^-) or as carbonate mineral (for example, $CaCO_3(s)$). Once in the ocean environment, the dissolved bicarbonate is incorporated into the seawater carbonate system (see Equation 1 above).

Since inorganic carbon is already abundant in the ocean, the riverine input of HCO_3^- has little importance for overall production of organic matter through photosynthesis in the ocean (see Equation 2 below). However, riverine input of bicarbonate is an important contributor to the overall alkalinity of the ocean environment, i.e. how well the ocean can buffer changes in pH, as the net contribution of HCO_3^- from rivers will add to a net uptake of protons (H^+). This contribution is important for understanding recent climate-related phenomena such as ocean acidification (Raven et al., 2005), but lies outside the scope of this report. This report focuses on the transport and fate of terrestrial organic carbon in the coastal ocean environment.

Organic carbon (OC) in the aquatic environment is formed by photosynthesis, whereby microscopic algae (phytoplankton) and larger epiphytes, such as macro algae and sea grasses, take up DIC (mainly HCO_3^-), inorganic nutrients (NO_3^- and PO_4^{3-}) and water to convert it to organic matter with the help of light energy from the sun (Libes, 2009), according to the generic formula Equation 2:

$$106CO_2 + 16HNO_3 + H_3PO_4 + 122H_2O \rightarrow (CH_2O)_{106}(NH_3)_{16}H_3PO_4 + 138O_2 \qquad \text{(Eq. 2)}$$

Organic carbon can occur in a wide variety of forms in the aquatic environment, and together with nutrients such as nitrogen and phosphorus, comprises the main building blocks for the soft tissue in living organisms, both in aquatic- as well as terrestrial ecosystems. When organisms die and start to degrade they first form Particulate Organic Matter (POM; >0.5µm) and/or Dissolved Organic Matter (DOM; <0.5µm), which can further be divided into Particulate- and Dissolved Organic Carbon (POC and DOC) and Particulate- and Dissolved nutrients (PON/DON and POP/DOP) respectively as shown in Figure 1.

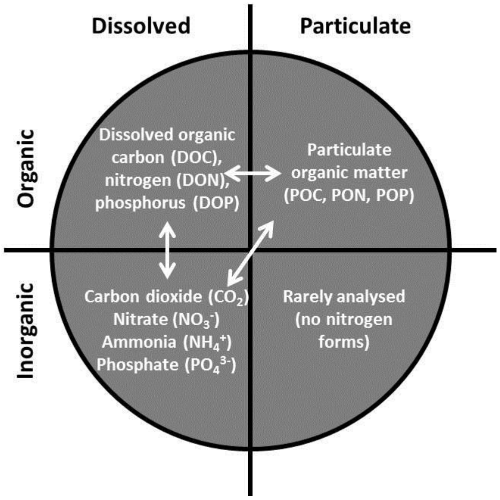

Figure 1. Different fractions of organic matter in aquatic environment and its inorganic components.
Note: Arrows indicate ways of transformation between different fractions (Williams et al. 2004).

Bacteria eventually completes the degradation process thereby converting the organic matter back to its original inorganic constituents, CO_2 and inorganic nutrients (NO_3^-, PO_4^{3-}), while consuming oxygen as part of their respiration (i.e. the reverse of Equation 2).

2.0 Riverine transport of terrestrial organic matter to the ocean

Each year approximately 400 - 430 x 10^{12}g of terrestrial organic carbon is transported from the continents via rivers to the global ocean (Hedges et al. ,1997; Schlünz and Schneider, 2000).

An estimated 63% of this terrestrial organic carbon is transported as dissolved organic carbon (DOC <0.5 µm) and the reminder (37%) as particulate organic carbon (POC >0.5 µm). The global fluvial discharge of DOC alone should be enough to sustain the turnover of the entire pool of organic carbon in the ocean. Likewise, the fluvial input of POC should be sufficient to supply all the organic carbon buried in marine sediments. Since riverine organic carbon typically consists of highly degraded and nitrogen poor terrestrial detritus, it might be expected to suffer minimal degradation in the ocean.

Yet only a very small fraction of the organic carbon dissolved in the ocean, or preserved in underlying sediments, seem to be of terrigenous origin (Hedges et al, 1997). Schlünz & Schneider (2000) estimate that 10% of the riverine input of organic carbon (i.e. 43 x 10^{12}g) is buried in shelf sediments each year. So, the question is where does the remaining 90% of the terrestrial organic carbon go? Recent studies suggest that this organic carbon is rapidly and effectively removed via natural processes in estuaries and near-shore environments, restricting export of terrestrial organic carbon to the open ocean (Borch et al. 2004).

3.0 Fate of terrestrial organic carbon in the ocean

Particulate and dissolved terrestrial organic carbon (and associated nutrients) are subject to substantial transformation and biogeochemical processing during fluvial transport as well as during estuarine circulation. This includes preferential degradation of labile carbon fractions by bacteria through respiration; physico-chemical and biological aggregation, photo-degradation and burial, as shown in Figure 2 below.

Depending on the type of terrestrial source for the organic matter (e.g. forest, peatland, soil) and the length and speed of the fluvial path as well as residence time in the estuary, the majority of this organic matter is highly modified and/or removed along the way towards the open ocean.

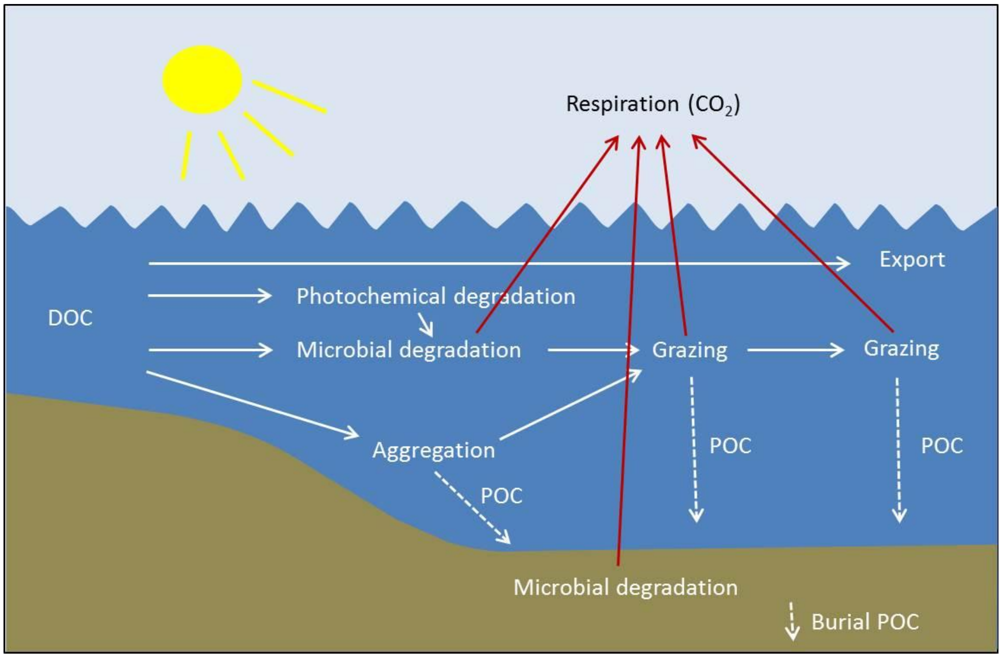

Figure 2. Schematic drawing of the potential fate of DOC in the estuary including a) aggregation; b) respiration by bacteria and higher organisms; c) photo-degradation and d) burial of organic carbon
Note: White arrows indicate transformation of DOC (dotted white arrows indicate sedimentation/burial) and red arrows indicate respiration (Borch et al. 2004).

These 4 processes are explained in some detail in this section.

3.1 Aggregation of dissolved organic matter in estuaries

Most organic carbon transported by rivers is in the form of dissolved organic carbon (DOC), which is subject to aggregation as it reaches the estuary. The term aggregation refers to the transformation of

DOC into the particulate phase, due to the physico-chemical changes occurring during estuarine mixing (i.e. where freshwater from the river meets the saltier water from the ocean). In the estuary, there are often strong vertical as well as horizontal gradients in both salinity and pH.

Adsorption and flocculation are the two general mechanisms by which DOC aggregates within the estuary. Once in particulate form, organic carbon can be removed from the water column through sedimentation or by filter feeding organisms. **Adsorption** is the attraction of dissolved organic carbon onto inorganic mineral surfaces such as clay particles and sand grains, covering these particles with an organic carbon film coating. **Flocculation** is the combination of large dissolved organic carbon molecules (e.g. humics) into larger particles. The rapid change in salinity within the estuary 'neutralizes' the surface charge of these particles, allowing them to aggregate into larger particles and eventually settle on the seabed (see Figure 2 above).

Biological aggregation involves the grazing of particulate organic carbon (POC) and associated microbes by higher trophic levels in the water column. Organic carbon is then further degraded and respired by fauna, before being repackaged into fecal material, which can rapidly sediment out of the water column (see Figure 2 above). The importance of physic-chemical aggregation in estuaries has been debated in the literature but results typically show that <10% is removed in this way (when salinity >5ppt). Little information is available on how much OC removal biological aggregation is responsible for in estuaries.

3.2 Photo-chemical degradation of dissolved organic matter in estuaries

Dissolved terrestrial organic matter typically absorbs light in the blue and ultraviolet (UV) part of the light spectrum (Markager et al., 2004), causing waters in many estuaries to have a yellow-brownish colour. The dissolved organic matter that absorbs light in blue and UV spectrum is often referred to as CDOM (Coloured Dissolved Organic Material). Light energy from the sun is capable of breaking the strong molecular bonds in the otherwise refractory terrestrial organic matter, splitting it into smaller organic compounds that can be further degraded within the estuary into CO_2 and nutrients through microbial respiration (see Figure 2 above).

The light absorption and subsequent photo-degradation of CDOM has consequences for the structural changes of the CDOM pool in the estuary, and also for light availability in the underlying water column. The CDOM concentration is typically high within the surface layer of the estuary due to density stratification between the incoming freshwater from the river and the underlying, more saline, seawater. Hence, the absorption of light can restrict the algal production to the upper few metres of the water column. In a recent study in Loch Etive, on the Scottish west coast, the CDOM was found to absorb almost 50% of the photosynthetically available radiation (PAR) and reduce the euphotic depth to between 7-10m during the spring bloom (McKee et al., 2002). Availability of nutrients below the euphotic depth indicated that light availability was the limiting factor for phytoplankton growth there.

3.3 Microbial degradation of dissolved organic matter in estuaries

Whereas aggregation and photo-degradation of organic carbon takes place at specific interfaces such as surface waters and in the turbidity maximum, microbial respiration of DOC takes place throughout the water column and in the sediments, as the water column can contain millions of bacterial cells per ml of seawater, and the sediments typically contain several orders of magnitude more (Markager et al., 2004).

Microbial respiration involves the conversion of DOC into bacterial biomass and CO_2 while consuming oxygen. The bacterial biomass can then be passed on to higher trophic levels through grazing by small unicellular organisms (e.g. by protozoa, flagellates and ciliates) which in turn are prey for larger organisms. Thus, the DOC is continuously transferred and respired throughout the food web and some of the more refractory carbon is ‘repackaged’ again as POC (e.g. zooplankton fecal material) which can sediment out of the water column (see Figure 2 above).

The released CO_2 is incorporated into the seawater carbonate system (see Equation 1 above), where it can be used again for algal production or eventually escape back to the atmosphere through equilibration (see Figure 2 above). Estuarine sediments are typically sites of very intense biogeochemical activity. High sedimentation rates of terrestrial- as well as marine organic carbon (e.g. dead phytoplankton) often promote high carbon turn-over rates, and associated high oxygen consumption rates in estuarine sediments.

The supply of organic matter typically sustains high abundance and biomass of sediment dwelling animals, which in turn facilitate the turn-over of carbon due to their mixing and irrigation activity in the sediments. High benthic oxygen flux rates, often >20 mmol m^{-2} d^{-1} and in some cases >100 mmol m^{-2} d^{-1}, have been recorded in UK estuaries (Overnell et al., 1996; Trimmer et al., 1998; Trimmer et al., 2000; Loh et al. 2002; Overnell, 2002). Oxygen fluxes typically increase towards the mouth of the estuary where the OM sedimentation rate, mostly of terrigenous origin, is the highest (see Table 1 below). These flux values are generally higher than oxygen flux values measured in e.g. North Sea shelf sediments (Bakker and Helder, 1993; Canfield et al., 1993; Stahl et al., 2004), indicating high OC degradation rates within estuaries.

Table 1. Range of sediment oxygen flux rates in UK estuaries

Location	**O_2 flux (mmol m^{-2} d^{-1})**	**Study**
Thames estuary	12.2 – 241.3	Trimmer et al 2000
Great Ouse estuary	57.5-76.7	Trimmer et al. 1998
Loch Etive	8.0-52.3	Overnell et al. 1996; 2002
Loch Goil	15.1-23.6	Overnell et al. 1996
Loch Fyne	10.7-20.4	Overnell et al. 1996
Loch Creran	6.6-18.7	Loh et al. 2002

3.4 Burial of dissolved organic matter in estuaries

The residual sedimentary organic carbon that is not respired by the benthos will eventually be preserved (i.e. buried) in deeper anoxic sediments and hence removed from the marine carbon cycle over potentially geological timescales.

Shelf and coastal sediments are one of the most important sinks for carbon on earth (Berner, 1982). Their carbon sequestration efficiency for OC is dependent on several factors including the OC sedimentation rate (Canfield, 1994), the OC oxygen exposure time in the sediment (Hartnett, Devol et al., 1996) as well as the quality of the incoming OC (Stahl, Tengberg et al., 2004). As oxygen is the most energetically favourable electron acceptor for OM oxidation, aerobic respiration is the most efficient microbial pathway for the degradation of OM (Glud, 2008). The longer OM is exposed to oxic conditions in the sediments the more it is degraded. However, higher sedimentation rates lead to higher benthic oxygen demand and shallower oxygen penetration in surface sediments and hence shorter oxygen exposure (Glud, 2008). Once OM has escaped the surficial oxic layer it is subject to a set of anaerobic respiratory pathways i.e. denitrification, metal oxide- and sulphate reduction respectively) of sequentially declining degradation efficiency (Glud, 2008).

The high sedimentation rates found in several Scottish sea lochs (Ansell, 1974; Overnell and Young, 1995; Loh et al., 2010) and high respiration rates (see Table 1 above) indicate that sea Lochs are hotspots for recycling and burial of OM. Rivers discharging into these sea lochs (Etive, Linnhe and Creran) have large catchment areas extending into the Scottish highlands with its vast areas of peatland bogs, which supply the lochs with significant amounts of highly degraded organic detritus (Ansell, 1974; Loh et al., 2008).

Loh et al. (2010) and Overnell & Young (1995) measured average OC burial rates of 0.4 and 0.25 g C m^{-2} d^{-1} in Loch Linnhe and Loch Creran, respectively. These are high OC burial rates, comparable to other coastal habitats with high carbon sequestration potential (e.g. seagrass meadows, saltmarshes) and significantly higher than typically found in North Sea shelf sediments, as shown in Table 2.

Table 2. Comparison of long term OC accumulation rates between different coastal/marine habitats

Habitat	**OC accumulation rate ($g C m^{-2} yr^{-1}$)**	**Source**
Tidal saltmarshes	210	Chumura et al. 2003
Seagrass meadows	45-190	Duarte et al. 2005
Sea loch sediments	91-146	Loh et al. 2010; Overnell et al 1995
Muddy shelf sediments	53	Stahl et al. 2004

Given the vast number of sea lochs on the west coast of Scotland, and the total area of their sub-tidal sediments (Edwards and Sharples, 1986), and assuming an overall burial rate between 0.25 and 0.40 g C m^{-2} d^{-1}, the carbon sequestration potential in Scottish sea loch sediments (10.1 -$17.8*10^{10}$ g C yr^{-1}) could be of an order of magnitude higher than, for example, the total average sequestration potential in Scottish saltmarshes ($14.1*10^{9}$ g C yr^{-1}; Stahl, 2012).

Saltmarshes have recently been highlighted as important 'blue carbon' sinks (Laffoley and Grimsditch, 2009). However, there are large uncertainties in the values presented, as they are based on a limited number of measurements from a few sea lochs and there may be large discrepancies between individual lochs. Although OC accumulation rates in muddy shelf sediments (e.g. Stahl et al., 2004) are lower than the rates found in, for example, sea loch sediments (Overnell and Young, 1995; Loh et al., 2010), muddy/sandy mud shelf sediments within Scottish territorial waters most likely constitute the largest store of 'blue carbon' due to their extensive areal coverage (20-25% of shelf, estimated from Pantin, 1991).

4.0 Organic carbon budget for a Scottish estuary: Loch Creran - a case study

Due to the natural complexity of terrestrial organic matter and its multiple fates within an estuary, it is challenging to quantify all the different sources and sinks for organic carbon in an estuary. Hence, very few such studies exist for Scottish estuaries. Loh et al. (2010), however, established a complete OC budget for Loch Creran on the Scottish west coast, by measuring the riverine discharge of OC, benthic- and water column respiration, sediment deposition and accumulation as well as export of OC from the loch. They found that Loch Creran was a net sink for OC, where ~81% of the total OC input was either sequestered in the sediment (~41%) or oxidized within the loch (mainly in the water column), and the remaining ~19% was exported out of the loch, as shown in Figure 3.

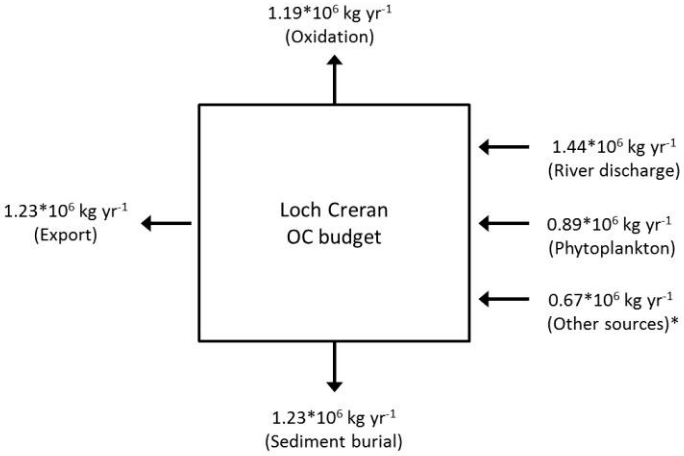

Figure 3. Organic carbon budget for Loch Creran.
*'Other sources' refers to OC input from anthropogenic sources such as fish farming in the loch (after Loh et al., 2010).

5.0 Climatic and land use effects on fluvial transport of organic matter

Strong relationships between export of terrestrially derived DOC and runoff have been found (Williams et al., 2004), often following seasonal patterns with increasing concentrations during spring and autumn periods. Therefore, it is likely that changes in the hydrological cycle due to climate change will have an effect on the export of terrestrial organic carbon through fluvial transport. Computer simulations as well as a recent study of several catchments along a climatic gradient in Europe have confirmed this (Williams et al., 2004; Mattsson et al., 2009).

Land use is another issue that might alter and increase the flux of terrestrial organic carbon through fluvial pathways to the ocean. Increased anthropogenic activity (e.g. agriculture) often leads to increased fluxes of DOC and nutrients from the terrestrial to aquatic environment (Mattsson et al., 2009). In Scotland land use associated with peatlands has been widely discussed in the literature (Wilson et al., 2011). One such activity is the draining of Scottish peatlands, which has been proven to have negative effects in the form of peatland erosion, biodiversity loss and increased carbon fluxes (Wilson et al., 2011 and references therein).

Both climate change and land use activity are thus likely to have an impact on the export of terrestrial DOM into Scottish estuaries, where it can undergo further degradation back to CO_2, get buried in the sediments or be exported to the open sea. Increased loading of DOM will lead to higher biological oxygen demand in the marine environment and an increased nutrient input, which could stimulate further marine primary production (i.e. eutrophication). On the other hand, increased concentrations of CDOM could also lead to stronger light attenuation in the water column (McKee et al., 2002), which could limit the PAR availability for primary producers.

However, the net effect of these two processes, i.e. stimulated production by increased nutrient input or decreased light availability by CDOM absorption, is difficult to foresee. Nonetheless, increased DOC input will lead to increased respiration and oxygen demand in the estuaries. Many Scottish sea lochs for example, have restricted water exchange with the sea due to their strong physico-chemical gradients and complex bottom topography. Thus, an increase in the biological oxygen demand might lead to hypoxia of bottom waters in the sea lochs, if the demand of oxygen exceeds the supply (Gillibrand, 2001). Hypoxia is well known to have negative effect on benthic communities (Pearson and Rosenberg, 1978), with risk of loss of biodiversity and ultimately ecosystem collapse if the hypoxia is severe and long lasting (Diaz and Rosenberg, 2008).

6.0 Conclusion

Significant amounts of organic matter are transported via fluvial pathways to Scottish estuaries such as sea lochs and firths. However, the literature and available evidence suggest that the majority of this input is currently retained within the ‘estuarine filter’, where it either undergoes further degradation to CO_2 or gets buried within deeper anoxic parts of estuarine sediments. Thus, it can be concluded that Scottish estuaries act as sinks for both terrestrially- as well as marine derived organic carbon (i.e. from estuarine production). The high organic carbon accumulation rates observed in sea loch sediments compare well with other high accumulating coastal habitats like saltmarshes and sea-grass meadows, and emphasise their regional importance as carbon stores. Finally, climate induced changes in precipitation patterns over Scotland will, in combination with continued land use such as peatland drainage, most likely lead to enhanced fluxes of terrestrial dissolved organic carbon into aquatic environments, where secondary effects like altered production and hypoxia are possible scenarios.

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