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Distribution of particulate and dissolved organic carbon in surface waters of northern scottish fjords

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ABSTRACT

Coastal waters can act concurrently as a source and sink of carbon (C), with coastal sediments trapping and storing significant quantities of C, while C is simultaneously released to the atmosphere through biogeochemical processes in the water column. The mid-to high-latitude fjords are recognised as hotspots for the burial and storage of OC in their sediments and for their potential to provide a long-term climate regulation service. Yet the distribution of particulate (POC) and dissolved organic carbon (DOC) in fjord water columns remains a significant knowledge gap, that is particularly acute in temperate vegetated systems such as those in the North Atlantic. Here we present POC and DOC data from surface waters across four Scottish fjords with differing meteorological, catchment and submarine geomorphological characteristics to both quantify and understand the factors governing the spatial distribution of POC and DOC in fjords. The measured POC and DOC concentrations in the surface waters of the four fjords are broadly analogous to other temperate vegetated fjord systems around the world. Within these systems, local factors such as submarine geomorphology is the primary factor driving POC and DOC dispersal in fjord surface waters. The data highlights that fjords are important pathways for the transport and storage of POC and DOC in the coastal ocean and that a greater focus is required on the water column OC to allow the role fjords play in near- (water column) and long-term (sediment) climate regulation to be quantified.

1. Introduction

Annually rivers deliver 250 Mt of dissolved organic carbon (DOC) and 150 Mt of particulate organic carbon (POC) to the global oceans (Bianchi et al., 2011; Ducklow and McCallister, 2004; Hedges et al., 1997; Hedges and Keil, 1995; Meybeck, 1982). Therefore understanding the distribution of both POC and DOC across the land-ocean interface is crucial to constrain the coastal and marine carbon (C) cycles (Bauer et al., 2013). Yet in many coastal environments, the transport and distribution of DOC and POC in the water column remains largely under researched. One such environment is fjords, though they only occupy an area 455,000 km² globally, they are recognised as important hotspots for the burial and storage of organic carbon (OC) within their sediments (Cui et al., 2016; Smeaton et al., 2017; Smith et al., 2015). Globally it is estimated that fjords bury 18 Mt of OC annually (Smith et al., 2015) with between 55 and 62% of that OC originating from terrestrial sources (Cui et al., 2016). Inshore and coastal waters are known to act as reactors of OC simultaneously being sinks (sediments) and sources (water column)

of C (Aufdenkampe et al., 2011; Cole et al., 2007). Yet the distribution of POC and DOC in the water column of fjords remains a significant knowledge gap.

It is estimated that 815 Mt of sediment enter fjords from rivers annually (Dürr et al., 2011), yet water column POC and DOC data is only reported for seven vegetated non-glaciated fjords globally (Mathew et al., 2021; Nima et al., 2016; van Breugel et al., 2005; Yamashita et al., 2015) with no records originating from mid-latitude systems such as those found in UK. Currently it is estimate that between 0.53 and 1.15 Mt OC yr⁻¹ is exported from UK rivers to the coastal ocean in the form of DOC (Williamson et al., 2021; Worrall et al., 2020). The rivers supplying the fjords are poorly represented in these estimates, yet you would expect a high flux of POC and DOC from land to sea due to the topography, (Bianchi et al., 2020; Howe et al., 2010), OC rich soils (Lilly and Donnelly, 2012) and high rainfall (Edwards and Sharples, 1986) that characterise the catchments of the mid-latitude fjords of Scotland.

Here we explore the concentration and distribution of POC and DOC across surficial waters of four mid-latitude fjords in NW Scotland. Each

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of the four fjords have differing, catchment, submarine geomorphology and meteorological characteristics allowing the mechanisms, which govern POC and DOC in the surficial waters to be explored, furthering our understanding of the role of fjords both in the coastal and global C cycle.

2. Study area

The mid-latitude fjords of Scotland occupy an area of 2608 km² (Smeaton et al., 2021) and drain ~18% of Scotland's land. This study focuses on four fjords in the NW of Scotland, from Loch Eriboll the most northerly mainland fjord down to Loch Torridon (Fig. 1). Fjords are defined by their over-deepened geomorphological characteristics (Bianchi et al., 2020; Howe et al., 2002) but even within the four systems in this study there are unique geomorphic differences that potentially govern the transport and fate of OC. Potentially, one of the main drivers of OC transport in the systems in how geomorphologically restricted they are by submarine sills. Shallow or numerous sills can reduce the inflow of water from the wider marine environment into the fjord. Though the mid-latitude fjords are silled they are significantly less restricted than the fjords of New Zealand and Chile (Bianchi et al., 2020). The mid-latitude fjords of Scotland are well ventilated with regular flushing events (Austin and Inall, 2002; Gillibrand et al., 2005; Gillibrand and Amundrud, 2007) resulting in well oxygenated bottom waters and the sustained input of marine derived OC which likely impacts the concentration of POC and DOC in the fjords.

The four fjords in this study can be broadly characterised as single sill, single basins systems (Loch Eriboll and Inchard) and multi sill, multi basin fjords (Loch a' Chàirn Bhàin and Torridon), these difference have been shown to strongly govern the fjords ability to trap POC (Bianchi et al., 2020; Smith et al., 2015).

Loch Eriboll (Fig. 1b) is 32.4 km² with a maximum depth of 68 m, the fjord has one sill at a depth of 25 m separating the fjord from the coastal

ocean. Loch Inchard (Fig. 1c) is the smallest of the fjords in this study occupying an area 4 km² with a maximum depth 61 m. Loch Inchard has one sill at 24 m depth separating the fjord from the coastal ocean.

Loch a' Chàirn Bhàin (Fig. 1d) is a branched fjord with parallel upper basins in the form of Loch Glendu and Glencoul. The fjord as a whole is $16.4~{\rm km}^2$ with a maximum depth of 108 m. The fjord has six sills separating the basins, the outer sill is at a depth of 33 m, the sills separating Loch Glendu and Loch Glencoul are at depths of 24 and 5 m respectively. Loch Torridon is the largest and deepest of the four fjords occupying an area of 73 ${\rm km}^2$ with a maximum depth of 145 m. Like Loch a' Chàirn Bhàin, the fjord is multi basin separated by three sills at depths of 40, 53 and 11m.

Beyond geomorphology the other likely drivers of POC and DOC is the soil type within the catchments and if a mechanism to mobilise this OC in the soil is present (land use, extreme climate). The catchments of Scotland's fjords have a wide variety of soils ranging between OC rich peats to OC poor fluvial deposits (Soil Survey of Scotland, 1981). Additionally, the pressure on the soils also differs significantly, with land uses ranging between baron montane environments to industrial forestry and pasture agriculture. The catchments of the four fjords have a similar soil architecture and land cover but the meteorological and hydrological conditions differ significantly (Table 1).

3. Methods

3.1. Sampling

Surface seawater samples (1000 ml) were collected on board the *MRV Alba na Mara* from approximately a depth of 1 m between the 23rd of July and 7th of August 2018. Samples were collected from 60 stations across the four fjords (Fig. 1). The water samples were collected in acidwashed 1000 ml opaque HDPE bottles. At each sampling station the water temperature (°C), pH and turbidity (NTU) was recorded.

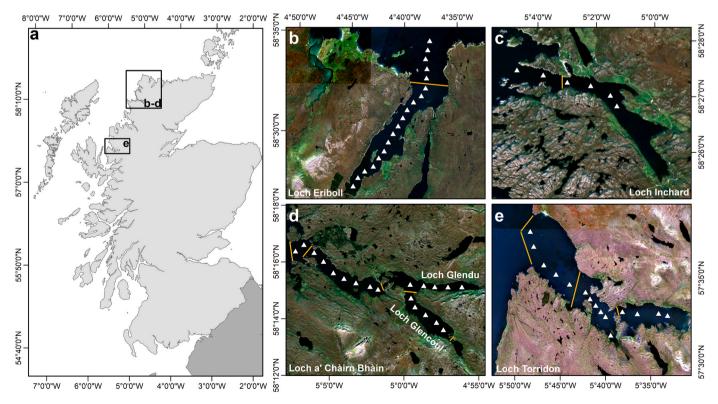


Fig. 1. Location of the fjords and associated sampling sites investigated within this study. (a) Overview of study sites within the context of Scotland. Sampling sites at (b) Loch Eriboll, (c) Loch Inchard, (d) Loch a' Chàirn Bhàin (e) Loch Torridon. Orange lines indicate the locations of the submarine sills (Edwards and Sharples, 1986). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Table 1
Catchment characteristics of each of the four fjords. Catchment and meteorological data derived from Edwards and Sharples (1986), soil and land use data derived from the Soil Survey of Scotland (1981).

Fjord	Catchment (km²)	Rainfall (mm yr ⁻¹)	Runoff (Million m ³ yr ⁻¹)	Dominant Soil (% coverage)	Dominant Land Cover (% coverage)
Eriboll	107	1500	134	Peaty Gley (49%)	Heather Grassland (64%)
Inchard	84	1750	126	Peaty Gley (69%)	Heather Grassland (72%)
a' Chàirn Bhàin	174	1750	260	Peat Gley (61%)	Heather Grassland (68%)
Torridon	241	2000	421	Peaty Gley (48%)	Heather Grassland (62%)

3.2. Sample treatment and analysis

On-board the vessel a known volume of sample (500-1000 ml) was filtered onto a pre-weighed and combusted (4hr and 450 $^{\circ}$ C) 0.45 μm glass fibre filters (Whatman GF/F) under gentle vacuum (<0.1 MPa). The filters were frozen at -20 °C and stored until analysis. On return to the laboratory, the filters were freeze dried and reweighed allowing the calculation of Total Suspended Material (TSM) (Nima et al., 2016). Once dry the filters underwent acid fumigation where the filters were exposed to concentrated Hydrochloric acid (HCl) fumes for 24hrs in a desiccator to remove carbonate (Harris et al., 2001). The OC content (%) of the material held within the filter was determined using an Elemntar Vario EL Elemental (EA) following the methodology of Nieuwenhuize et al. (1994) and Verardo et al. (1990). The EA was calibrated using Acetanilide and the precision of the analysis is estimated from repeat analysis (n=12) of standard reference material B2178 (Medium Organic content standard from Elemental Microanalysis, UK) with C data varying by 0.07% from known values.

A fraction of the filtrate (50 ml) was transferred to pre-combusted opaque glass bottles and the pH of the sample was adjusted to pH 2 using 2M HCl. The acidified samples were stored at 4 $^{\circ}$ C in the dark prior to analysis (<1 week). The DOC was quantified using a total organic carbon analyser (TOC) where the sample was acidified with 0.1 ml of 2M HCl and sparged with oxygen to assure the removal of inorganic carbon prior to thermal oxidation to quantify the organic fraction. Final DOC concentrations are reported as average values of triplicate measurements. The precision of the analysis was estimated to be $\pm 2~\mu mol$ C determined with low carbon water (MilliQ) and CRM seawater reference standards (Hansell, 2005).

4. Results and interpolation

4.1. Particulate organic carbon (POC)

The concertation of POC across the surface waters of the fjords ranges between 35.7 and 290.7 μ mol C L $^{-1}$ with the highest concentrations found in the upper basins at the head of each of the fjords. The POC concentrations found in the four fjords is comparable to the limited data from other vegetated non-glaciated fjords (Table 2).

The surface waters of the upper region of Loch Eriboll (Stations E1-E7) are characterised by POC concentrations averaging 227.5 \pm 38.0 $\mu mol~C~L^{-1}$ in comparison the lower stations in the fjord have an average POC concentration of 83.8 \pm 44.3 $\mu mol~C~L^{-1}$ (Fig. 2). The significant differences between these two zones is likely driven by terrestrial OC input at the head of the fjord which is subsequently trapped around the island of Eilean Choraidh situated in the upper portion of Loch Eriboll

Table 2 Comparison of POC and DOC concentrations (μ mol C L $^{-1}$) across other vegetated and glaciated fjords as well as data from the wider coastal ocean. *Errors expressed as standard error opposed to standard deviation in the other data.

	DOC (μ mol C L ⁻¹)	POC (μ mol C L $^{-1}$)	Reference
Loch Eriboll	72.9 ± 5.6	132.4 ± 83.9	This Study
Loch Inchard	67.4 ± 4.0	63.1 \pm 23.6	
Loch a' Chàirn Bhàin	$\textbf{61.4} \pm \textbf{19.0}$	$145.2 \pm \\64.2$	
Loch Torridon	24.9 ± 3.9	$108.8 \pm \\35.0$	
Vegetated Fjord	s		
New Zealand Norway	89–157 48–108	35–260	Yamashita et al. (2015) (Mathew et al., 2021; Nima et al., 2016; van Breugel et al., 2005)
Glaciated Fjords	<u> </u>		
Chile	20–200	62.3 ± 8.9	(González et al., 2019; Marshall et al., 2021)
Greenland	100 ± 22		(Lund Paulsen et al., 2019)
Norway	114–136	24-62	(Gašparović et al., 2007; 2005)
Svalbard	20–220	11–30	(Brogi et al., 2019; Holding et al., 2017; Osterholz et al., 2014; Zhu et al., 2016)
Other Coastal W	aters (Global)*		
Coastal Waters	285.7 ± 5.7		Barrón and Duarte (2015)
Open Coastal Zone	126.4 ± 2.6		
Vegetated	246.0 \pm		
Coastal Waters	17.6		
Estuaries	329.8 ± 7.0		

(Fig. 1). Additional the island itself could be a source of terrestrial OC for the surrounding surface waters. Stations E11- E13 though distal are adjacent to a fin-fish (Salmon) aquaculture site potentially leading to the increased POC found at these sites.

Loch Inchard is an outlier as the POC varies little within the fjord, with the surface waters averaging a POC value of $63.1\pm23.6~\mu mol~C~L^{-1}.$ Loch Inchard is the smallest of the fjords studied and due to its size samples were not collected from its upper reaches, likely resulting in the lack of variability and initial spike in POC values observed at the head of the other fjords. When we consider this data to be a snap-shot of the lower section of the fjord the data has similar features (i.e. low concertation and low variability) to that observed in lower portions of the other systems (Fig. 2).

Loch Glendu and Glencoul act as parallel upper basins for Loch a' Chàirn Bhàin, yet they have significantly different POC concentrations. The surface waters of Loch Glendu have an average POC concentration of 141.5 \pm 27.7 μmol C L^{-1} compared to 220 \pm $6\bar{4}$ μmol C L^{-1} found in Loch Glencoul. The catchments of these two fjords have the same soils and land usage and are subject to the same meteorological conditions (Table 1) yet the POC concentration differ. The only major difference between the two fjords is the depth of the sills, Loch Glendu is separated from the main basin of the fjord by a sill at a depth of 24 m versus a 5 m sill separating Loch Glencoul. This extremely shallow sill separating Loch Glencoul from the other basins results in the fjord being extremely restricted and in turn the surface waters likely retain a significant portion of the terrestrial OC that enters the system. Loch a' Chàirn Bhàin exhibits similar POC concentrations (113.7 \pm 32.6 μ mol C L⁻¹) to the lower portions of the other fjords. Station CB7 has a significantly higher concentration of POC than surrounding stations (Fig. 2), this sampling site sits above the outer sill of the fjord. The exchange of coastal and fjord water across submarine sills can result in changes in water temperature, salinity, density and tidal circulation (Cuthbertson et al., 2021;

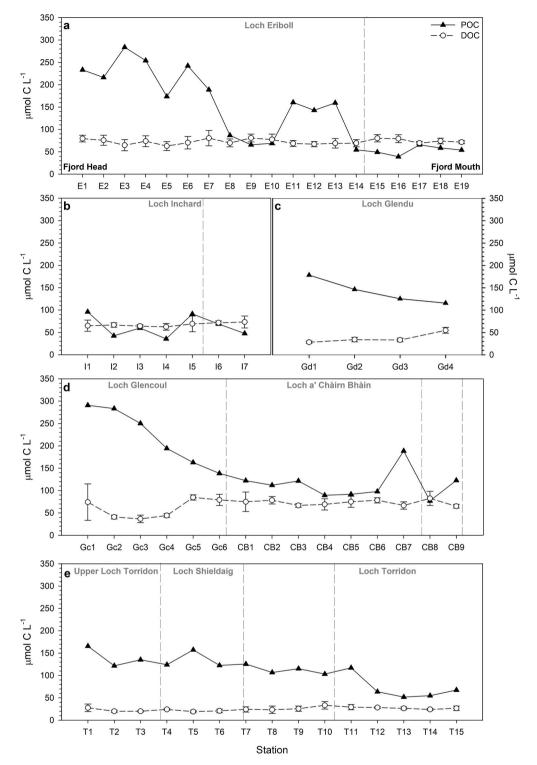


Fig. 2. POC and DOC concentrations across the sampling transects from the head to mouth of the fjords (a) Loch Eriboll (b) Loch Inchard (c) Loch Glendu (upper basin of Loch a' Chàirn Bhàin) (c) Loch a' Chàirn Bhàin (d) Loch Torridon. Dashed lines indicate the location of the submarine sills (Fig. 1).

Gillibrand and Amundrud, 2007; Rüggeberg et al., 2011) these changes potentially facilitate the retention of both marine and terrestrial derived POC in the surface waters around the sill.

POC concentrations of the surface waters of Loch Torridon have a similar pattern to that in the other systems (i.e. higher POC concentrations at the head of the fjord) yet the decrease in POC to the mouth of the fjord is muted in comparison to that observed in Loch Eriboll and a' Chàirn Bhàin (Fig. 2). POC concentrations at stations T12-T15 (59.5 \pm 7.4 μmol C L^{-1}) are approximately half that observed in the rest of the

fjord (126.8 \pm 19.4 μ mol C L $^{-1}$). Theses samples are from an area of the fjord completely open to the wider coastal ocean, likely resulting in the dilution of the POC, resulting in the lower concentration observed.

The pattern of high POC at the head of the fjord which decreases as you move to the mouth is prevalent in all the fjords and is likely primarily driven by terrestrial OC input (Bauer et al., 2013; Bianchi et al., 2011). As this terrestrial material is transported towards the coastal ocean a combination of factors, drive the decrease in POC with local factors (e.g. sill height) potentially determining the rate of the decrease

(Fig. 2). The most likely drivers for the decrease in POC is the organic matter has (i) fallen out of the water column and been incorporated into the sediment, (ii) been consumed by macro and microorganisms, (iii) been diluted as the fjords becomes more exposed to the wider coastal ocean.

From the surficial sediments of mid-latitude fjords we know that a significant proportion of OC trapped and incorporated into the sedimentary store originates from marine sources (Smeaton and Austin, 2017; Smeaton et al., 2021) therefore to attribute all the changes in POC concentration in the surface waters to terrestrial OC input alone would be a mistake. Marine OC tends to accumulate in the sediments found in the outer regions of fjords (Cui et al., 2016; Smeaton and Austin, 2017), the surface water POC in these areas exhibit relatively low concentrations and variability in comparison to the upper sections (Fig. 2) potentially due to the quantity of OC provided by each source (terrestrial vs marine).

4.2. Dissolved organic carbon (DOC)

The concentration of DOC across the fjords ranges between 19.5 and $84.7~\mu mol~C~L^{-1}$ which is comparable to values observed in Norwegian fjords (Mathew et al., 2021; Nima et al., 2016; van Breugel et al., 2005). In contrast the DOC concentrations observed in the four mid-latitude fjords are low when compared to the systems of New Zealand (Table 2). The disparity between the data is twofold (i) the current available DOC data does not represent the true range of DOC values observed in either the Scottish systems or fjords globally. (ii) The difference in environments makes comparison difficult, New Zealand fjords are highly restricted and mostly hypoxic (Smith et al., 2015) leading to high DOC concertation in comparison to the more open well ventilated mid-latitude fjords of Scotland (Austin and Inall, 2002; Gillibrand et al., 1995).

Unlike the POC where there are clear trends from the head to the mouth of the fjords, the DOC exhibits little variability down the fjords (Fig. 2). The DOC concertation of Loch Eriboll (72.9 \pm 5.6 μ mol C L⁻¹), Inchard (67.4 \pm 4.0 μ mol C L $^{-1}$) and Torridon (24.9 \pm 3.9 μ mol C L $^{-1}$) and a' Chàirn Bhàin (72.9 \pm 6.2 μ mol C L $^{-1}$) show little variation yet the restricted Loch Glendu and Glencoul do differ (Fig. 2). Both these fjords have low DOC concentrations at the head of the fjords, which slowly rise as you move towards the sill. The concertation of POC in these basins is believed to be strongly linked to terrestrial input yet the low concertation of DOC indicates low terrestrial DOC input. This suggests that the terrestrial environment may not play the same role with DOC as it does with POC and marine OC input is potentially driving the DOC load within the surface waters. This trend is most abrupt in Loch Glencoul which is the most restricted of all the fjords, the shallow sill is potentially preventing the exchange of water reducing the quantity of marine derived DOC entering the highly restricted basin (Fig. 2).

The concertation and distribution of POC and DOC in the fjords seems decoupled as the mechanisms that drive the changes in POC in the fjords generally fail to explain the trends observed in the DOC. A potential reason for the low concentrations and low variability in the DOC is the weather conditions prior and during sampling. In Scotland, July 2018 was the fourth hottest on record and only experienced 66% of rainfall compared to the average between 1981 and 2010. The hot dry conditions that led up to the sampling will have likely reduced the export of DOC to the fjords potentially accounting for the low concentrations observed. The weather conditions likely also impacted the POC but with the current data it difficult quantify such impacts.

5. Conclusion

The results from this study illustrate distinct differences in the concentration and distribution of POC and DOC within the four fjords. The POC and DOC concentrations in the fjords are likely heavily influenced by local factors with submarine geomorphology driving the dispersal of

the POC and DOC throughout the fjords.

Though limited the data highlights that fjords are important pathways for the transport and storage of POC and DOC in the coastal ocean. Yet these systems remain missing from most national and global compilations of coastal DOC and POC due to the current dearth of data. To overcome this hurdle and shed greater light on the mechanism that drive POC and DOC fluxes in fjords, we initially recommended:

- Repeat seasonally sampling of fjords to better understand the impact of weather and oceanography on the concertation and distribution of POC and DOC.
- (ii) Expand the analytical approaches to understand the source of the POC and DOC (i.e. C/N and stable isotopes)
- (iii) Better constrain the fluvial POC and DOC export to the fjords through increased monitoring.

CRediT authorship contribution statement

Craig Smeaton: Writing – review & editing, Writing – original draft, Visualization, Resources, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **William E.N. Austin:** Writing – review & editing, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

All data used in this research is included in the manuscript and supplementary material.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ecss.2022.107952.

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