AGU PUBLICATIONS

JGR

Journal of Geophysical Research: Biogeosciences

RESEARCH ARTICLE

10.1002/2017JG003952

Key Points:
- Terrestrial catchments are an important source of OC to the coastal ocean
- Coastal marine sediments are more effective long-term sinks of terrestrial OC than their adjacent terrestrial catchments
- Over time, the terrestrial OC subsidy to coastal sediments become increasingly important as a mechanism for local OC storage

Supporting Information:
- Supporting Information S1

Correspondence to:
C. Smeaton,
cs244@st-andrews.ac.uk

Citation:
Smeaton, C., & Austin, W. E. N. (2017). Sources, sinks, and subsidies: Terrestrial carbon storage in mid-latitude fjords. *Journal of Geophysical Research: Biogeosciences*, *122*. https://doi.org/10.1002/2017JG003952

Received 21 MAY 2017
Accepted 5 SEP 2017
Accepted article online 14 SEP 2017

Sources, Sinks, and Subsidies: Terrestrial Carbon Storage in Mid-latitude Fjords

Craig Smeaton[1] and William E. N. Austin[1,2]

[1]School of Geography and Geosciences, Irvine Building, University of St Andrews, Saint Andrews, UK, [2]Scottish Association for Marine Science, Scottish Marine Institute, Oban, UK

Abstract Fjords are recognized as globally important sites for the burial and long-term storage of carbon (C) within sediments. The proximity of fjords to the terrestrial environment in combination with their geomorphology and hydrography results in the fjordic sediments being subsidized with organic carbon (OC) from the terrestrial environment. It has been well documented that terrestrial OC (OC_{terr}) is an important component of coastal sediments, yet our understanding of the quantity of OC_{terr} stored in these sediments remains poorly constrained. Utilizing Bayesian isotopic sediment fingerprinting techniques to the surface sediments of Loch Sunart, we estimate that 42.0 ± 10.1% of the OC is terrestrial in origin. Through combining these outputs with sedimentary OC stock estimates, we have calculated that the surface sediments (0–15 cm) hold 0.1 megaton (Mt) OC_{terr} and estimate that the postglacial sediment held within the fjord contains 3.96 Mt OC_{terr}. When these totals are compared to the quantity of OC stored in the adjacent terrestrial environment, it is clear that the fjord's catchment stores a greater amount of OC_{terr} in the form of vegetation and soil. Though when normalized for area the results suggest that the marine sediments are a more effective long-term store of OC_{terr} than the adjacent terrestrial environment. This striking result highlights the importance of the terrestrial environment as a source of OC to the coastal ocean and that the OC_{terr} subsidy to the marine sediments is a significant mechanism for the long-term storage of OC in coastal marine sediments.

Plain Language Summary Fjords are known to provide an important climate regulation service through the burial and long-term storage of organic carbon (OC) within sediments. The proximity of fjords to the land combined with their geomorphology means that they are excellent carbon sinks for terrestrial OC transported to the coastal ocean by hydrologically processes. Though we know fjords trap OC lost from the land, we lack an understanding of how much terrestrial OC is stored in the sediment of fjords and how that compares to the adjacent catchment where large quantities of OC is stored in the vegetation and soils. This work uses sediment fingerprinting techniques to identify the source of the OC (terrestrial versus marine) in the sediment of Loch Sunart, a midlatitude fjord. Using previous sedimentary OC stock estimates for Loch Sunart combined with catchment data, we have calculated and compared the amount of terrestrial OC stored in both environments. When compared, the catchment holds a far greater amount of terrestrial OC. Though when we consider the difference in size of the catchment versus the fjord, the results show that the fjord is a more effective long-term store of terrestrial OC than its adjacent catchment.

1. Introduction

The burial and storage of organic carbon in the coastal ocean is a key component of the carbon cycle at both local and global scales (Bauer et al., 2013). Globally, an estimated 276.6 megaton (Mt) of C is buried in the coastal zone each year, with approximately 126.2 Mt buried in depositional areas, that is, estuaries and the adjacent continental shelf (Duarte et al., 2005). An improved understanding of the factors regulating these C fluxes and their sources and an ability to account for the carbon stored in these coastal sediments remain important global challenges, with far-reaching implications for integrating the coastal ocean into global carbon budgets (Bauer et al., 2013).

A key component of our understanding of the coastal carbon cycle is the growing recognition of significant terrestrial OC subsidies into coastal marine sediments. The proximity of the coastal ocean to the terrestrial biosphere means that both environments have a closely interlinked C cycle. The terrestrial environment is an important source of both particulate (POC) and dissolved (DOC) organic carbon to the coastal ocean (Bianchi, 2011). Current global fluvial OC input to the coastal ocean is estimated to be

500 Mt C yr^{-1} with between 30 and 70% of this total being utilized biologically or stored in some form in the coastal ocean (Bauer et al., 2013). It has been suggested that the coastal ocean provides an important climate regulating service (Smith et al., 2015) by preventing much of this subsidized terrestrial OC from reaching the continental shelf/open ocean, where the potential for remineralization and loss to the atmosphere as CO_2 is enhanced. While there has recently been progress at the global scale in estimating the magnitude of the OC subsidy provided by the terrestrial environment to coastal sediments, the long-term storage of this OC at the local and regional scales remains poorly constrained. Without a good understanding of these local and regional processes we are unlikely to improve on the current global estimates and, in turn, provide the necessary constraint to quantify the magnitude of terrestrial OC subsidy to the coastal ocean.

Fjords are natural sediment traps at the land-ocean interface and effectively serve to highlight the global importance of the terrestrial OC subsidy to the costal ocean. The burial and long-term storage of OC in fjords is known to be significant (Smeaton et al., 2016; Smith et al., 2015), but, as with the majority of coastal environments, the long-term terrestrial OC subsidy into these systems is poorly constrained. Fjords are known to be global hot spots for OC burial, capturing approximately 11% of the total OC buried in marine sediments annually (Smith et al., 2015). The largely landlocked nature of fjords and their natural capacity to trap sediment suggests a significant proportion of the buried OC should be of terrestrial origin (OC_{terr}). It has been estimated that 21 ± 16 Mt of OC is buried in fjords annually with approximately 55% of this carbon originating from terrestrial sources (Cui, Bianchi, Hutchings et al., 2016). Studies to determine the sources of coastal OC have largely been focused in the high-latitude fjords of New Zealand (Cui, Bianchi, Hutchings et al., 2016; Cui, Bianchi, Savage et al., 2016; Hinojosa et al., 2014; Smith et al., 2015), Chile (Sepúlveda et al., 2011; Silva et al., 2011), and Svalbard (Faust et al., 2014; Koziorowska et al., 2016), with only limited data available for midlatitude fjords (Loh et al., 2008). While these studies show the presence of significant amounts of OC_{terr} in fjordic sediments, the net long-term terrestrial subsidy to coastal sedimentary OC stores over extended (decadal to millennial) time periods remains poorly constrained.

Here we determine the origin(s) of the OC held within the marine sediment of a midlatitude fjord. In conjunction with total marine sedimentary OC stock estimations (Smeaton et al., 2016), we make direct comparisons between estimates of postglacial OC_{terr} stored in the fjord sediment and the OC_{terr} held within the soil and living biomass of the adjacent catchment. As in previous studies, we demonstrate a significant midlatitude OC_{terr} contribution into the sediment stores of the adjacent coastal ocean. However, we also highlight that while significant terrestrial "losses" are generally well known, they remain largely unaccounted for as a net OC subsidy to long-term coastal marine sediment stores. Here we set out a framework for the quantification of C in adjacent terrestrial and coastal sedimentary environments which helps to highlight the very significant OC_{terr} subsidy received by coastal marine sediments.

2. Study Site

Loch Sunart (56.708°N, 5.749°W), a fjord on the west coast of Scotland (Figure 1), is one of the longest fjords in Scotland (30.7 km) with a maximum depth of 145 m. The fjord consists of three basins separated by sills at depths of 6 m and 31 m (Gillibrand et al., 2005). In addition, Loch Teacuis a small branch fjord (5.8 km long) connects into the middle and outer basin of Loch Sunart. It has been calculated that Loch Sunart holds 1928.3 ± 7.3 Mt of sediment of which an estimated 9.4 ± 0.2 Mt of postglacial OC is stored (Smeaton et al., 2016).

The combined catchments of Loch Sunart and Loch Teacuis cover an area of 299 km^2. The catchment is covered by carbon-rich soils (Figure 2) with 63% of the soil consisting of peaty gleys (Soil Survey of Scotland, 1970–1987). The mean depth of the soil is approximately 50 cm with an estimated maximum depth of 100 cm (Bibby et al., 1982). The vegetation and land cover is largely dominated by acid grassland, coniferous forest, and broad leave woodland (Morton et al., 2011) (Figure 2). The catchment geology consists of metamorphic and igneous rocks with minimal input potential from petrogenic/fossil carbon into the local system. The characteristics of Loch Sunart's catchment are similar to most midlatitude fjords of mainland Scotland (Edwards & Sharples, 1986) and are comparable to systems in New Zealand, Norway, and Canada (Howe et al., 2010; Syvitski & Shaw, 1995).

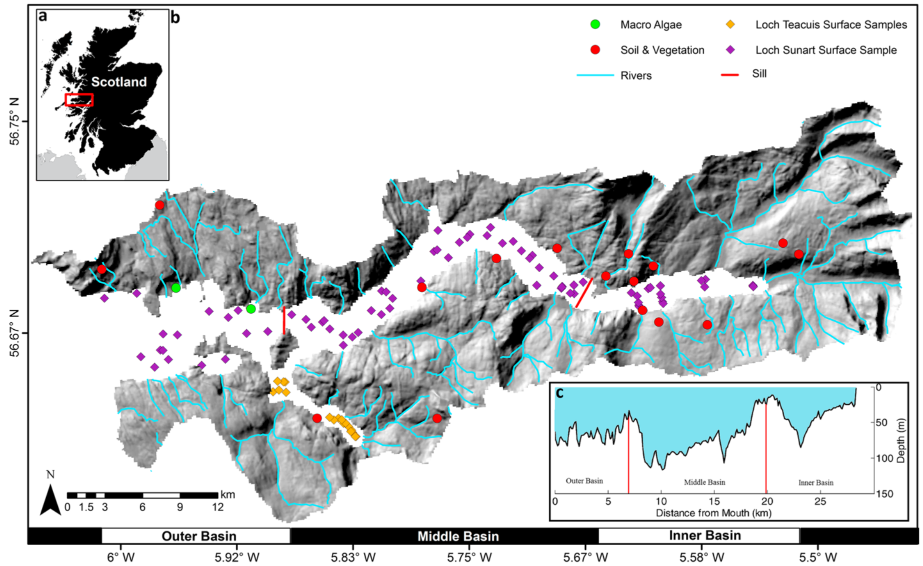

Figure 1. (a) Overview map illustrating the location of Loch Sunart in context to Scotland. (b) Map illustrating the topography and drainage network of the catchment. Sampling sites are highlighted: Loch Sunart (purple diamonds), Loch Teacuis (orange diamonds), soil and vegetation (red circles), and macroalgae (green circles). (c) Bathymetric profile of Loch Sunart developed utilizing data from Bates et al. (2004) and Baltzer et al. (2010).

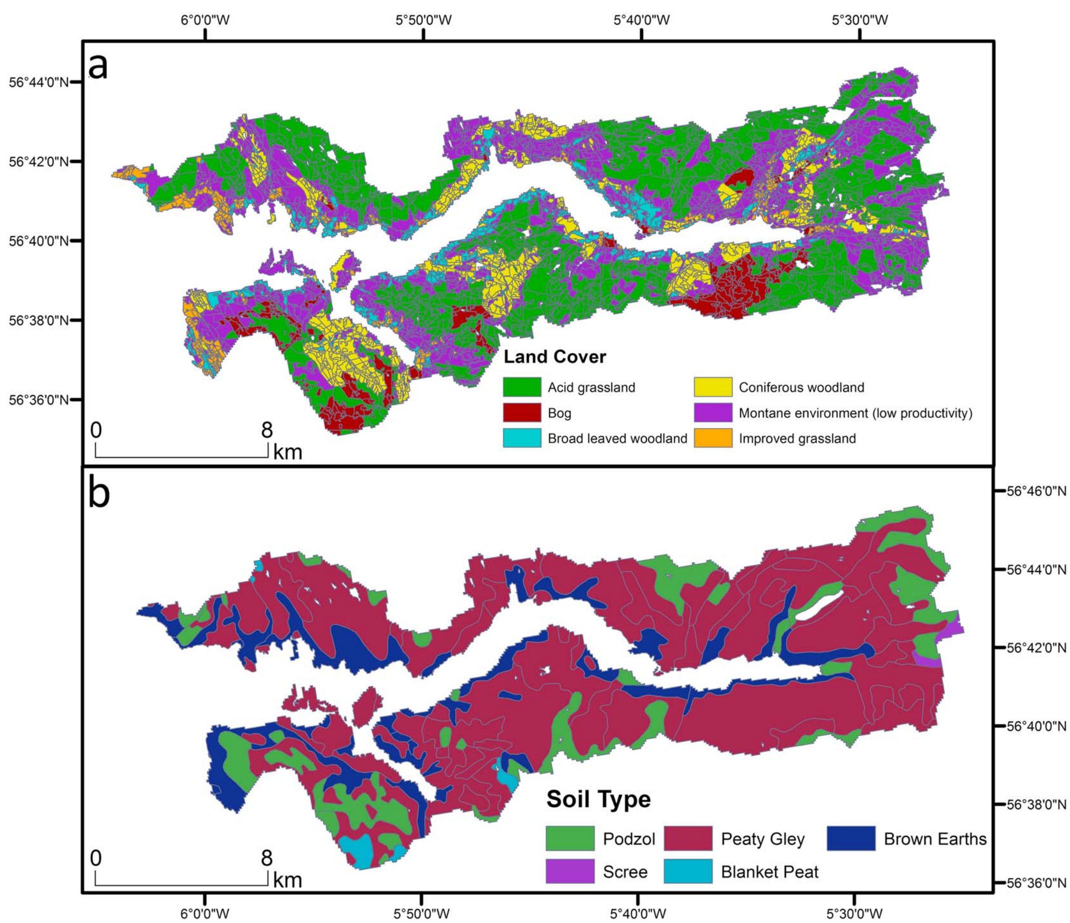

Figure 2. Maps illustrating the characteristics of the combined catchment of Loch Sunart and Teacuis. (a) Land cover (Morton et al., 2011). (b) Soil type (Soil Survey of Scotland, 1970–1987).

3. Methods

3.1. Overview

Bulk elemental data and stable isotope values have extensively been used to identify the source of OC in coastal systems (Cui, Bianchi, Savage et al., 2016; Gordon & Goni, 2003; Hinojosa et al., 2014; Sepúlveda et al., 2011; Silva et al., 2011; Smith et al., 2010; Thornton & McManus, 1994). End-member mixing models developed by Thornton and McManus (1994) and further developed by Gordon and Goni (2003) provide a quantitative framework to assess the relative inputs of OC_{terr} and OC_{mar} into marine sediments. These techniques have been furthered by the introduction of Markov chain Monte Carlo (MCMC) methodologies (Li et al., 2014) and standalone Bayesian isotope mixing models (Arendt et al., 2015; Morris et al., 2015). These mixing models use elemental ratios and stable isotope values to separate the terrestrial and marine components; OC_{terr} is isotopically lighter and has a high C:N ratio in comparison to OC_{mar} (Bauer et al., 2013; Meyers, 1994; Peter et al., 1978; Redfield et al., 1963). However, caution must be applied to the interpretation of downcore data because syndepositional and postdepositional degradation of different components of OC can alter the isotopic and elemental values of the overall sediment. The gradual downcore loss of labile OC below the sediment surface means that the resultant net OC_{terr} may increase in relative abundance as the more labile OC_{mar} is degraded. For the purposes of this study, we focus only on surface samples which are considered to best reflect the primary signal at the seafloor.

3.2. Sampling

Surface sediment samples (n = 98) were collected from Loch Sunart and Teacuis using a Van Veen grab sampler from the head to the mouth of each fjord (Figure 1). In total 83 samples were collected from Loch Sunart and 15 from Loch Teacuis (supporting information). Soil and vegetation samples were collected from throughout the catchment (Figure 1) to help characterize the range of terrestrial source values used in later modeling. In total, 12 soil samples were collect covering the full range of soil types in the catchment (Figure 2). Additionally, eight samples of living and dead vegetation were collected representing the three dominant groups of vegetation (grasses, coniferous, and broadleaved woodland) in the catchment. In order to represent the marine source values for macroalgae, samples were collected from Glenborrodale Bay (56.676, −5.906) and Camas Fearna (56.684, −5.960) within Loch Sunart (Figure 1). Further samples of phytoplankton, zooplankton, and benthic microalgae were provided by Marine Scotland Science from their sampling/monitoring station in Loch Ewe (57.849, −5.649) in North West Scotland. Detailed description of all samples collected can be found in Table 1.

3.3. Bulk and Stable Isotope Analysis

The surface, catchment, and marine samples were analyzed to determine bulk elemental and stable isotope values. Each sample was freeze dried and homogenized; approximately 12 mg of processed sediment was weighed out into tin capsules and a further 12 mg was weighed into silver capsules. The samples encapsulated in silver underwent acid fumigation step (Harris et al., 2001) to remove carbonate. After drying for 24 h at 40°C, OC and $\delta^{13}C_{org}$ were measured using an elemental analyzer coupled to an isotope ratio mass spectrometer at the Natural Environment Research Council (NERC) Life Science Mass Spectrometer Facility (Lancaster, UK). The samples in the tin capsules were analyzed for total carbon (TC), total nitrogen (TN), and $\delta^{15}N$ at the same facility. The standard deviation of $\delta^{13}C_{org}$ and $\delta^{15}N$ triplicate measurements (n = 15) were 0.07 ‰ and 0.17 ‰, respectively. $\delta^{13}C_{org}$ and $\delta^{15}N$ values are reported in standard delta notation relative to Vienna Peedee belemnite (VPDB) and air, respectively. By analyzing $\delta^{15}N$ separately, we negate the potential risk of the acid fumigation step altering the $\delta^{15}N$. The quantity of inorganic carbon (IC) in each sample was calculated by subtracting the OC from TC. C:N ratios are reported as molar ratios where C:N = (OC/12)/(TN/14).

3.4. Terrestrial and Marine Source Characterization

For the purposes of the binary mixing model mean values are required for the local terrestrial and marine environments to act as source inputs to the model. To represent the terrestrial environment two source values were calculated; one representing the local soil and vegetation and the second using values derived for land cover over the entirety of Scotland (Thornton et al., 2015). These terrestrial source values are represented by the weighted mean; the weighting for these calculations were produced through examination of soil and land use maps (Morton et al., 2011; Soil Survey of Scotland, 1970–1987) (Figure 2). The weighting for

Table 1
Detailed Description of Catchment and Marine Samples Collected to Represent the Local Terrestrial and Marine Environments With Their Associated Bulk Elemental and Stable Isotope Values (Relative to VPDB)

Sample ID	Latitude and longitude	Description	$\delta^{13}C_{org}$ (‰)	$\delta^{15}N$ (‰)	OC (%)	N (%)	C:N
Soil 1	56.700, −5.687	Gley (peaty gleys)	−28.38	4.36	13.18	0.85	18.09
Soil 2	56.702, −5.525	Gley (peaty gleys)	−27.71	2.94	38.68	1.82	24.79
Soil 3	56.693, −5.618	Brown soil (brown earth)	−28.01	4.81	13.83	1.19	13.56
Soil 4	56.717, −5.972	Blanket peat (dystrophic)	−27.59	2.89	44.22	2.83	18.23
Soil 5	56.633, −5.773	Blanket peat (dystrophic)	−27.87	1.88	28.51	1.90	17.51
Soil 6	56.669, −5.579	Gley (peaty gleys)	−28.26	2.31	31.03	2.94	12.31
Soil 7	56.695, −5.730	Gley (peaty gleys)	−28.07	3.36	17.35	1.59	12.73
Soil 8	56.684, −5.783	Brown soil (brown earth)	−27.87	1.39	40.64	2.50	18.97
Soil 9	56.691, −6.013	Podzol (humus-iron podzols)	−26.7	7.07	5.05	0.40	14.73
Soil 10	56.675, −5.625	Brown soil (brown earth)	−27.57	5.22	17.26	1.44	13.98
Soil 11	56.675, −5.625	Podzol (Humus-Iron Podzols)	−27.74	1.89	3.37	0.08	49.15
Soil 12	56.633, −5.859	Brown soil (brown earth)	−27.81	3.78	16.43	1.29	14.86
Veg 1	56.687, −5.632	Coniferous forest (mulch)	−29.73	0.31	47.96	1.80	31.09
Veg 2	56.687, −5.632	Coniferous forest (needles)	−30.29	1.65	52.34	0.91	67.10
Veg 3	56.671, −5.614	Coniferous forest (needles)	−29.59	0.48	51.69	1.20	50.25
Veg 4	56.671, −5.614	Coniferous forest (twiggy)	−29.32	1.66	51.41	1.28	46.86
Veg 5	56.689, −5.652	Deciduous forest (mulch)	−28.39	−0.20	49.9	2.20	26.46
Veg 6	56.689, −5.652	Deciduous forest (leaf matter)	−28.47	−0.20	49.29	1.95	29.49
Veg 7	56.697, −5.635	Acid grass	−27.40	4.50	37.12	3.50	12.37
Veg 8	56.697, −5.513	Acid grass	−27.86	4.30	42.47	4.20	11.80
MacA 1	56.684, −5.960	*Fucus vesiculosus*	−19.66	5.62	39.04	3.06	14.88
MacA 2	56.684, −5.960	*Fucus spiralis*	−19.11	6.18	39.14	2.87	15.91
MacA 3	56.684, −5.960	*Fucus vesiculosus*	−18.46	5.86	38.73	3.02	14.96
MacA 4	56.684, −5.960	*Ulva compressa*	−18.37	5.83	26.31	2.07	14.83
MacA 5	56.676, −5.906	*Fucus vesiculosus*	−17.21	5.98	38.62	2.81	16.03
MacA 6	56.676, −5.906	*Silvetia compressa*	−18.72	5.95	40.32	2.87	16.39
MacA 7	56.676, −5.906	*S. latissima*	−16.81	6.72	26.63	2.45	12.68
MacA 8	56.676, −5.906	*S. latissima*	−16.75	6.74	27.9	2.21	14.73
Phyto 1	57.849, −5.649	Bulk phytoplankton	−21.45	5.40	14.98	2.41	7.25
Phyto 2	57.849, −5.649	Bulk phytoplankton	−21.39	5.31	13.75	3.15	5.09
Zoop 1	57.849, −5.649	Copepods	−21.91	5.82	48.00	10.41	5.38
Zoop 2	57.849, −5.649	Euphausiids	−20.22	7.21	41.20	10.21	4.71
BMicA	57.849, −5.649	Bulk benthic microalgae	−17.4	−1.00	38.01	7.10	6.25

all calculations are detailed in the supporting information. Similarly, a representative marine source value was determined by using the phytoplankton, zooplankton, macroalgae, and benthic microalgae bulk elemental ratio and stable isotope values. In the absence of specific data to constrain these marine source contributions, we assumed that each marine component contributed equally to the marine source value.

The Bayesian mixing model can deal with additional source values, and we have therefore split the terrestrial source mean value into its constituent components (i.e., soil and vascular plants). The soil source value was calculated to represent the distribution of soil types within the catchment; the weighting to calculate this mean source value was determined from values quoted in the Soil Survey of Scotland (1970–1987). In addition, the weightings to calculate the vascular plant source value was produced through examination of the 2007 land cover map (Morton et al., 2011). The marine source value for the binary mixing model was also used with the Bayesian model approach.

3.5. End-Member Mixing Modeling

Initial analysis was carried out using a simple binary (or two end-members) mixing model designed to discriminate between OC_{terr} and OC_{mar} based upon the work of Thornton and McManus (1994). $\delta^{13}C_{org}$ values and C:N ratios were used separately as tracers and applied to the mixing model (equations (1)–(3)) to calculate the fraction of OC_{terr}.

$$\text{Fraction OC}_{terr} = \left(\delta^{13}C_{mar} - \delta^{13}C_{terr}\right)/\left(\delta^{13}C_{mar} - \delta^{13}C_{sample}\right) \quad (1)$$

$$\text{Fraction } OC_{terr} = \left(C : N_{mar} - C : N_{sample}\right) / \left(C : N_{mar} - C : N_{terr}\right) \quad (2)$$

$$\text{Fraction } OC_{terr} + \text{Fraction } OC_{mar} = 1 \quad (3)$$

These equations were augmented by the application of Markov chain Monte Carlo (MCMC) simulations to source variability in endmembers (Andersson, 2011; Li et al., 2014). The MCMC simulations were performed in the OpenBUGS software package (Lunn et al., 2009). Simply, 1,000,000 out of 100,000,000 random samples from a normal distribution of each end-member were taken to simultaneously populate the mixing models (equations (1)–(3)). Through this process a significant number of solutions are generated which follow a normal distribution. The mean, range (minimum and maximum), and standard deviation of source contributions were calculated for each sample from the MCMC solutions available.

The binary mixing model approach was expanded upon using the open source Bayesian isotope mixing model FRUITS (Fernandes et al., 2014). Bayesian models overcome some of the problems of the simpler models discussed above. These models integrate MCMC simulations throughout the process and are capable of using multiple tracers (isotopic and nonisotopic); in this study $\delta^{13}C_{org}$, $\delta^{15}N$, and C:N ratios were jointly utilized as tracers. This approach enabled the estimation of the proportional contribution of terrestrial and marine-derived OC and provided a direct comparison with the simpler binary models. We assumed that no isotope discrimination occurred between the source and incorporation into the surface sediment. We also assumed that the source values selected are representative of the entire region covered by the study. Work by Gordon and Goni (2003) showed that it was possible to split the terrestrial OC into inputs from soil (OC_{soil}) and vascular plants (OC_{vp}). However, the similarity of isotopic values of the terrestrial material within the study region make it difficult to separate the OC_{soil} and OC_{VP} components; for this reason a Bayesian approach was preferred over the standard three-end-member mixing model (Gordon & Goni, 2003). To assure the quality of model outputs the best practices outlined by Phillips et al. (2014) were followed. The outputs from the mixing models were used to spatially map the distribution of OC from different sources. The mapping was undertaken using a Kriging (with linear interpolation) gridding technique (Cressie, 1990).

3.6. Terrestrial and Marine OC Stocks and Burial

To determine the OC held in the soil and living biomass of the catchment, we utilized the outputs from the Countryside Survey 2007 (Henrys et al., 2012, 2016). The data from this survey allow for catchment specific estimates of soil (top 15 cm) and above ground living carbon (Figure 3). Using Bibby et al. (1982) soil depth estimations (mean = 50 cm) for the region, we estimated the total OC stock held within the catchment.

The combination of the mixing model outputs allow the OC_{terr} stock estimations for the top 15 cm (Figure 3) and postglacial sediment as a whole (Smeaton et al., 2016) to be derived. This calculation assumes that the OC_{terr} in the surface sediments is representative of the entire postglacial sediment sequence. In reality, the OC_{terr} input, burial, and preservation will have changed through the postglacial period, but, by assuming the modern sediments are characteristic of the postglacial sediment, we have been able to make first-order estimations of OC_{terr} content in the sediment.

The calculated OC stocks allow a like for like comparison of both the surface marine sediments and catchment soils (0–15 cm), as well as the OC_{terr} store (catchment versus fjord) as a whole. Additionally, through a normalization process according to the area of the postglacial store, a calculation of how effectively each environment stores OC_{terr} (terrestrial OC_{eff}) can be made.

3.7. A Framework to Conceptualize OC_{terr} Subsidies

We have used the methodology described above to estimate the quantity of OC_{terr} held in the surface sediments of the loch, as well as the living biomass and the topsoil of the catchment. To understand the OC_{terr} subsidies between terrestrial and coastal marine environments, there is also a need to better understand the transfer of OC from the catchment and the fate of that OC within the fjord. While a complete understanding of the fate of OC within the fjord environment is beyond the scope of this study, the burial of OC_{terr} can be calculated by combining the estimated proportion of OC_{terr} with the previous estimated OC burial rates for Loch Sunart (Smeaton et al., 2016), these rates represent how much OC is buried annually and take into accounted the total OC lost during the burial process. To estimate how much OC is lost from the

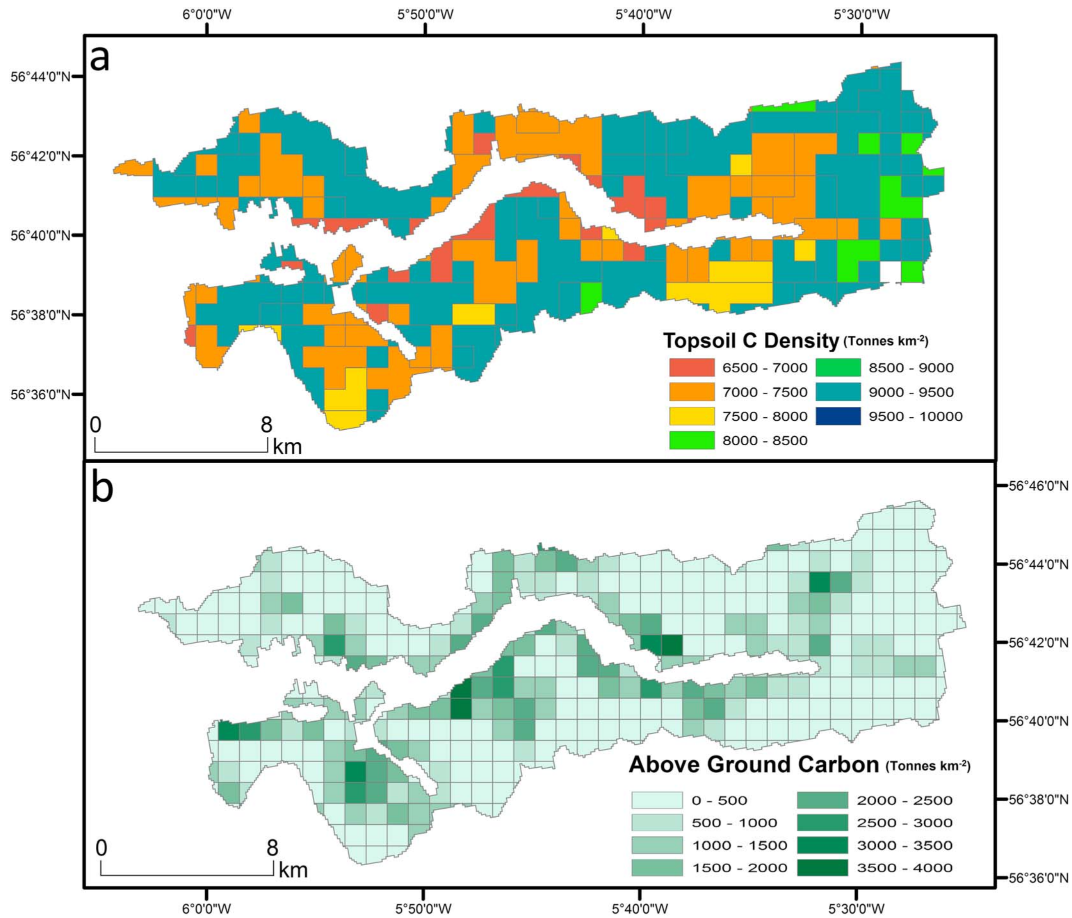

Figure 3. Maps illustrating (a) the topsoil carbon density (0–15 cm) and (b) aboveground carbon (living biomass). Maps produced from data collected for the Countryside Survey 2007 (Henrys et al., 2012, 2016).

catchment, we have used land cover data (Morton et al., 2011) and regional soil erosion rates for each land cover type from the wider literature (Borrelli et al., 2016; Duck & McManus, 1990; Grieve et al., 1994; Knox et al., 2015; Ledger et al., 1974; McManus & Duck, 1985; Panagos et al., 2015). To validate this approach two checks were employed: first, we determined the loss of OC through the fluvial system (rivers and runoff) using the methodology set-out by Loh et al. (2008) combined with flow (Payne et al., 1989), catchment (Edwards & Sharples, 1986), and soil C data from this study to estimate mean river discharge and OC loading (supporting information). Second the Revised Universal Soil Loss Equation (RUSLE) model (Panagos et al., 2015) was utilized to determine the potential soil erosion by water for the catchment. Through comparison of the fjord and catchment data, we can begin to understand the linkages between the two environments in terms of OC supply and delivery. Despite the qualitative nature of this framework, it allows an opportunity to conceptualize the processes that govern the subsidy of OC_{terr} from the terrestrial to the adjacent coastal environment.

4. Results

4.1. Bulk and Stable Isotope Analysis

Bulk elemental data show a clear distinction between the quantities of OC and IC held in the sediment of each subbasin of the fjord. The quantity of OC in the surface sediment of Loch Sunart ranges from 8.86% to 0.35% declining in a seaward direction away from the head of the fjord. The sediment in the inner basin contains the most OC with a mean value of 3.07 ± 2.3%, declining to 2.11 ± 1.81% and 1.51 ± 0.98 for the middle and outer basins, respectively. Loch Teacuis's sediment has a mean value of 4.74 ± 1.72%. The quantity of IC in each basin in negatively correlated with the OC with sedimentary IC increasing in a

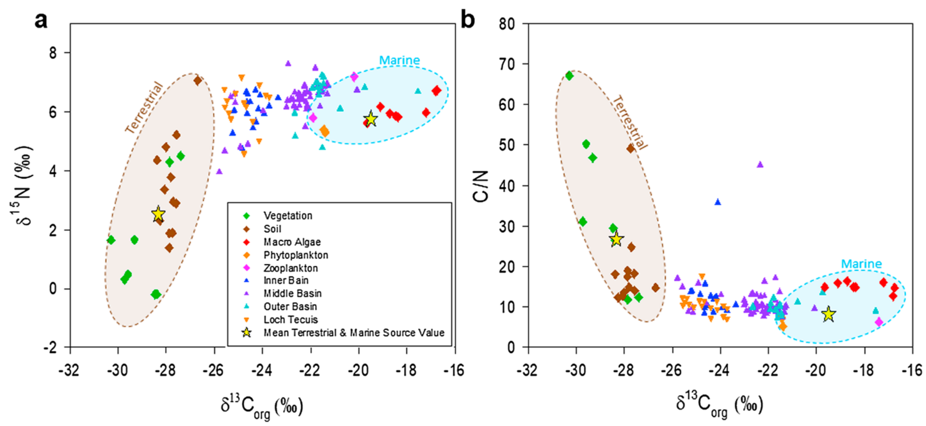

Figure 4. Cross plots (a) $\delta^{13}C_{org}$ versus $\delta^{15}N$ and (b) $\delta^{13}C_{org}$ versus C:N in surface sediment of Loch Sunart and Tecuis with the shaded envelopes illustrating the range of the terrestrial and marine source values. Additionally, OC data from the grab samples show a relationship between the composition of the OC and the salinity gradient within the fjord (supporting information).

seaward direction. The average C:N ratios were calculated for Loch Sunart (11.36 ± 5.64) and Loch Teacuis (11.22 ± 1.98).

Stable isotope ratios from the OC in surface sediments show a transition from isotopically depleted $\delta^{13}C_{org}$ signatures in the inner basin to more enriched values in the middle and outer basins (Figure 4). Samples from Loch Teacuis have depleted $\delta^{13}C_{org}$ values mirroring that of the inner basin of Loch Sunart. Sediment OC $\delta^{15}N$ ratios remain consistent throughout the loch but show greater variability in the inner basin of Loch Sunart. Similarly, the $\delta^{15}N$ values of Loch Teacuis closely match that of the inner basin of Loch Sunart. The average $\delta^{13}C_{org}$value for the fjord was −22.37 ± 1.13‰ for $\delta^{13}C_{org}$ and 6.41 ± 0.65‰ for $\delta^{15}N$. When compared to values from fjords around the world, the $\delta^{13}C_{org}$ values appear typical, but $\delta^{15}N$ values are significantly more enriched than those reported from other fjord sediments (Table 2), likley due to a greater than average OC input from marine sources.

Table 2
Surface Sediment $\delta^{13}C$ and $\delta^{15}N$ Isotopic Values (Relative to VPDB) for Loch Sunart (Broken Down by Basin) and Loch Teacuis

Fjord	Location	$\delta^{13}C_{org}$ (‰)	$\delta^{15}N$ (‰)	Reference
Loch Sunart	United Kingdom (Scotland)	−22.37 ± 0.13	6.41 ± 0.65	This study
Inner Basin		−24.42 ± 0.49	6.07 ± 0.56	
Middle Basin		−22.61 ± 1.23	6.39 ± 0.65	
Outer Basin		−21.37 ± 1.80	6.56 ± 0.68	
Loch Teacuis		−24.79 ± 0.62	6.19 ± 0.69	
Loch Creran	United Kingdom (Scotland)	−24.7 to −21.3		Loh et al. (2008)
Loch Etive		−25.8 to −25.7		
Nordasvannet Fjord	Norway (Mainland)	−23.9		Muller (2001)
Trondheimsfjord		−23.0 ± 1.1		Faust et al. (2014)
Hornsund	Norway (Svalbard)	−24.45 ± 0.31	4.00 ± 0.48	Koziorowska et al. (2016)
Adventfjord		−25.16 ± 0.40	2.83 ± 0.97	
Kongsfjord		−21.6 to −23.9		Kuliński et al. (2014)
Saguenay Fjord	Canada	−26.7 to −25.9		St-Onge and Hillaire-marcel (2001)
Clayoquot Sound		−27.04 to −22.58		Nuwer and Keil (2005)
Fiordland	New Zealand	−28.7 to −24.7		Smith et al. (2010)
		−25.10 ± 2.59	4.55 ± 2.59	Hinojosa et al. (2014)
		−27.82 to −23.50		Cui, Bianchi, Hutchings, et al. (2016)
		−28.1 to −23.3		Cui, Bianchi, Savage, et al. (2016)
Norther Patagonia	Chile	−28.2 to −19.1	1.3 to 9.0	Sepúlveda et al. (2011)
		−26.7 to −20.1		Silva et al. (2011)
Southern Patagonia		−22.1 to −19.7		Silva and Prego (2002)

Note. Listed for comparison are published equivalent isotopic values from the surface sediment of fjords around the world.

Table 3
Source Type and Their End-Member Values as Used With Mixing Models

Source	$\delta^{13}C_{org}$ (‰)	$\delta^{15}N$ (‰)	OC (%)	N (%)	C:N
Soil	−27.90 ± 0.43	3.52 ± 1.78	20.51 ± 8.52	1.46 ± 0.65	20.17
Vegetation	−28.61 ± 0.35	2.21 ± 2.44	45.97 ± 0.39	2.51 ± 0.01	29.47
Terrestrial (soil + veg)	−28.33 ± 0.70	2.52 ± 1.77	35.11 ± 9.56	1.84 ± 1.02	26.75
Terrestrial (land cover)	−28.08 ± 0.1	2.99 ± 0.2	18.54 ± 1.6	0.99 ± 0.07	25.79
Marine	−19.50 ± 0.58	4.24 ± 0.37	32.89 ± 3.01	5.36 ± 0.30	8.14

The results of the bulk elemental and stable isotope analyses of the catchment and marine samples collected as source values for the mixing models are outlined in Table 1. As expected, the samples fall into two groups representing the terrestrial (soil and vegetation) and marine (phytoplankton, zooplankton, benthic micro algae, and macroalgae) environments (Figure 4). The marine sediment samples themselves fall between these two groups, suggesting a mixture comprised from these sources.

4.2. End-Member Mixing Model

4.2.1. OC Source Characterization

Each source was characterized and assigned a $\delta^{13}C_{org}$, $\delta^{15}N$, and C:N value (Table 3) which was utilized in both the binary and Bayesian mixing models. The composite marine source value corresponds well to other studies (Cloern et al., 2007; Hinojosa et al., 2014; Sepúlveda et al., 2011; Thornton & McManus, 1994). The two methods of calculating the terrestrial source value have resulted in similar $\delta^{13}C_{org}$ and $\delta^{15}N$ values but the %OC and %N do vary significantly resulting in different C:N ratios. Despite differences between the methods, both results yield values which fall within the ranges found in the literature (Faganeli et al., 1988; Hinojosa et al., 2014; Thornton & McManus, 1994; Sepúlveda et al., 2011; Silva et al., 2011). The values assigned to the soil match those found in the literature for Scottish soils in this region (Ficken et al., 1998; Schmidt & Gleixner, 2005). The values assigned to the vegetation source are consistent with grasses, coniferous, and broadleaved trees (Cloern et al., 2007; Sepúlveda et al., 2011; Silva et al., 2011; Smith et al., 2010).

4.2.2. End-Member Mixing Modeling

The binary and Bayesian mixing models were run to determine the fraction of OC_{terr} within each of the surface samples. Each model was run separately with the terrestrial sources values derived from land cover and the composite soil/vegetation data (Table 3). The results produced using the different source values were combined to calculate the percentage of OC_{terr} within each sample. The average quantity of OC_{terr} was calculated for Loch Teacuis and each basin of Loch Sunart and the loch as a whole (Table 4).

The nature of the Bayesian model (i.e., combining multiple tracers and MCMC integration) means that we have high confidence in the results, further supported by the binary mixing model results (Table 4). For the purposes of this study, we have therefore used the Bayesian model results to assess the subsidy of OC_{terr} into the marine sediments of Loch Sunart. The greatest amount of OC_{terr} is found in the inner basin decreasing in the middle and outer basins. An estimated 42% of the OC in the surface sediment of Loch Sunart is terrestrial in nature, while 65% of Loch Teacuis's sediment originates from terrestrial sources. These subsidies are comparable to those report by Cui, Bianchi, Hutchings et al. (2016) who estimated that 55% of OC entering fjords globally is terrestrial in origin. However, when spatially mapped, there is a clear

Table 4
Percentage of Terrestrially Derived OC Held Within the Sediment of Loch Sunart and Loch Teacuis Broken Down by Basin and Mixing Model

Tracer	Inner basin	Middle basin	Outer basin	Loch Sunart	Loch Teacuis
			Two-end-member model		
$\delta^{13}C$	58.6 ± 11.0	37.2 ± 12.2	23.8 ± 12.7	34.8 ± 12.3	63.1 ± 10.8
C:N	52.6 ± 24.7	45.1 ± 23.0	36.2 ± 20.7	42.7 ± 22.4	49.4 ± 23.9
			Bayesian end-member model		
$\delta^{13}C$, $\delta^{15}N$, C:N	61.4 ± 7.6	41.8 ± 9.8	36.1 ± 11.4	42.0 ± 10.1	64.8 ± 5.2

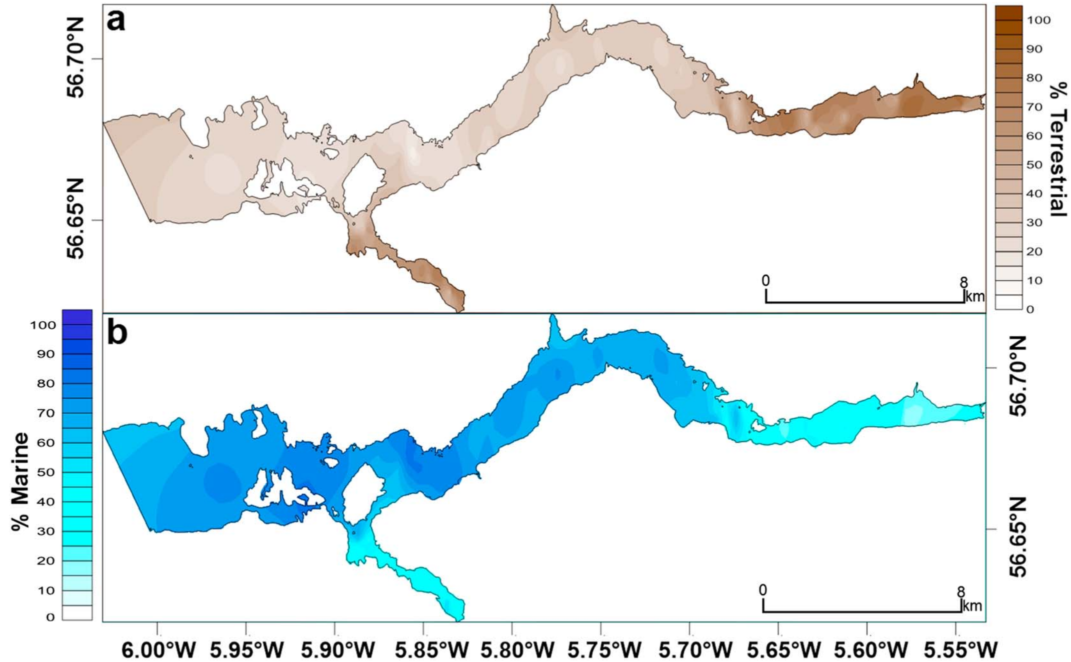

Figure 5. Modeled spatial distribution of (a) % terrestrial derived OC and (b) % marine derived OC within the surface sediment of Loch Sunart and Loch Teacuis.

transition from the terrestrially dominated inner basin through to the largely marine influenced outer basin of the fjord (Figure 5). Our mapping also illustrates the similarity of OC_{terr} in the highly constricted Loch Teacuis and the inner basin of Loch Sunart. High quantities of OC_{terr} are found locally in the middle basin of Loch Sunart; these hot spots occur at the mouth of larger streams. More generally, there is likely to be diffuse OC_{terr} input down the length of fjord from nonpoint sources (i.e., hillside runoff) and from advective processes within the water column.

4.3. Terrestrial and Marine OC Stocks

By combing the postglacial stock and burial rate estimations for Loch Sunart (Smeaton et al., 2016) with the %OC_{terr} calculated from this study, we estimated that the postglacial sediments hold 3.96 ± 0.1 Mt OC_{terr} with a further estimated 737 t OC_{terr} being buried annually. We calculated the quantity of OC held in the living biomass of the catchment to be 0.28 Mt and estimated the soil OC content at 7.97 Mt OC; the catchment has a whole holds 8.25 Mt OC. Area normalized totals were calculated (Table 5) which show not unexpectedly that Loch Tecuis, as the most landlocked location is the most effective at storing OC_{terr} followed by the inner basin of Loch Sunart (Figure 6).

When the depth integrated estimates of OC_{terr} stocks are area normalized, our results show that despite the significantly greater quantity of OC_{terr} held within the catchment the fjord sediments area a more effective store of OC_{terr}.

4.4. First-Order OC_{terr} Subsidy Estimates

This work has calculated that Loch Sunart's surface sediments (top 15 cm) hold 0.1 Mt OC_{terr} and that an estimated further 737 t OC_{terr} is added annually into this store. We estimate that between 7,500 and 15,000 t OC is lost annually from the catchment's soil (supporting information); this estimate is supported by both the fluvial export calculations and the RUSLE model outputs (supporting information). When used in conjunction with the fjord burial rates, we calculate that between 5 and 10% of the OC lost from the catchment is buried in the sediment. The additional ~90% of missing OC_{terr} could be removed from the system through multiple pathways (Figure 7). The primary processes most likely to account for this missing OC_{terr} are (i) some of the OC_{terr} export may be in a dissolved rather than particulate form (Bauer et al., 2013; Bianchi, 2011); (ii) some of the eroded soils may not reach the fjord and may be deposited within the catchment (e.g., floodplains)

Table 5
Calculated OC and OC_{terr} Carbon Stocks for the Sediment and Adjacent Catchment of Loch Sunart

	Area (km^2)	Total OC (Mt)	Terrestrial OC (Mt)	Terrestrial OC_{eff} (Mt OC_{terr} km^{-2})
Catchment: topsoil (top 15 cm)	299		2.39	0.008
Catchment: topsoil (50 cm)[a]	299		7.97	0.027
Catchment: living biomass	299		0.28	0.001
Catchment (soil + living biomass)	299		8.25	0.028
Loch Sunart (top 15 cm)	47.3	0.23	0.10	0.002
Inner Basin (top 15 cm)	5.5	0.03	0.02	0.004
Middle Basin (top 15 cm)	24.7	0.15	0.06	0.003
Outer Basin (top 15 cm)	17.1	0.05	0.02	0.001
Loch Teacuis (top 15 cm)	2.2	0.02	0.01	0.005
Loch Sunart (postglacial sediment)[b]	47.3	9.4	3.96	0.084

Note. Additionally, the area normalized totals are listed indicating how effectively each environment stores OC.
[a]Calculated by applying surface soil OC values (Henrys et al., 2012; 2016) to soil depth data (Bibby et al., 1982).
[b]Calculated by applying surface OC values to the postglacial sediment as quantified in Smeaton et al. (2016).

(Wang et al., 2014); (iii) some of the OC_{terr} may be degraded and lost to the atmosphere during transport in the fluvial system (Dinsmore et al., 2013; Leith et al., 2015), within the water column of the fjord (Burt et al., 2013) and within the sediment itself (Arndt et al., 2013; Glud et al., 2016); and (iv) some of the OC_{terr} may be exported further offshore to the continental shelf and beyond (Bischoff et al., 2016; Haas et al., 2002; Painter et al., 2016) bypassing the coastal sediment. It is likely that other fjords may be more efficient traps of OC_{terr} than these results suggest; we know that Loch Sunart does not suffer from periods of water column hypoxia (Gillibrand et al., 2006) and the absence of this OC preservation mechanism likely results in lower OC_{terr} preservation in comparison to sites with hypoxic conditions (Middelburg & Levin, 2009; Woulds et al., 2007). Therefore, while certain process may both under and overestimate certain aspects of annual OC_{terr} gains and losses from the catchment to the fjord sediments, they do serve to provide an overview of OC_{terr} subsides to the coastal ocean.

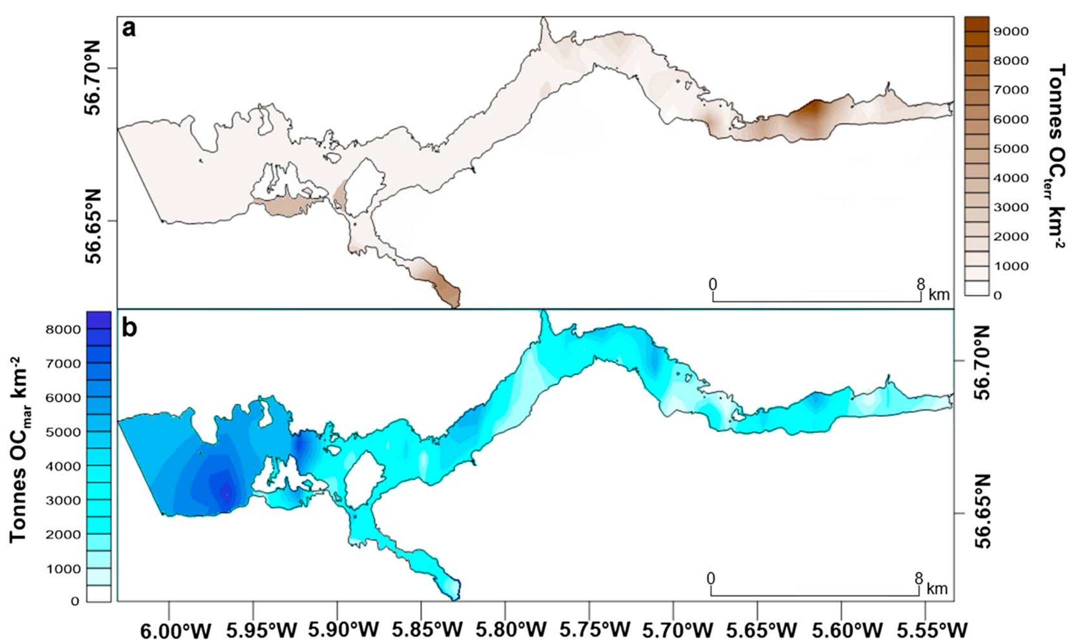

Figure 6. Area normalized (a) OC_{terr} and (b) OC_{mar} stock estimations (t OC km^{-2}) for the surface sediments (0–15 cm) of Loch Sunart and Tecuis.

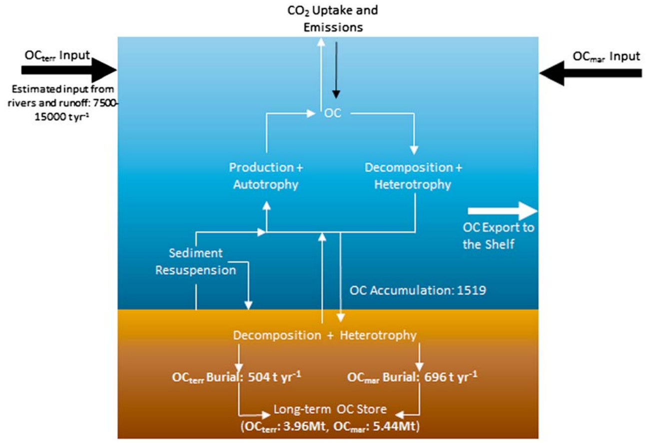

Figure 7. Conceptual diagram highlighting the pathways (white arrows) through which the OC input from external sources (black arrows) are processed within the water column and sediment of a fjord. Additionally, the mean values for soil erosion (t yr^{-1}), OC accumulation (t yr^{-1}), and burial (t yr^{-1}) calculated for Loch Sunart and its catchment are shown.

The terrestrial subsidy of C into long-term marine sediment storage is far from being an efficient or straightforward burial process (Figure 7). The fertilization of the coastal ocean through the input of OC_{terr} can result in increased primary production (Bianchi, 2011; Bauer et al., 2013) and, when combined with the high sedimentation rates found within fjords, creates a secondary, indirect pathway for OC_{terr} storage in these coastal sediments. The mechanisms that regulate this system are well characterized (Burd et al., 2016), yet rates of exchange between these C pools are still poorly understood. For example, work focusing on a shallow coastal system estimated that >25% of the OC_{mar} reaching the sediment could be attributed to OC_{terr} fertilization of the marine system (Watanabe & Kuwae, 2015). While the typically deeper water depths found in fjords may reduce this estimate, it is clear that this process potentially provides an indirect secondary pathway for OC_{terr} to reach coastal marine sediment stores.

5. Discussion

The results of this study serve to highlight the important role that the terrestrial environment plays in contributing C to coastal sedimentary systems. We have estimated the quantity OC_{terr} stored in coastal marine sediment and compared it to that of the adjacent catchment. The results indicate that on a like for like basis (i.e., 0–15 cm) the catchment soil stores a greater amount of OC_{terr} than the marine sediment; similarly, the living biomass within the catchment holds more OC_{terr}. This is, of course, expected because both the soil and living biomass are wholly terrestrial in nature, while the marine sediments have OC inputs derived from both the terrestrial and marine environment. While the living biomass of the catchment itself holds more OC_{terr} when normalized for area, the marine sediment is a more effective store of OC_{terr} (Table 5). Taking into consideration the full postglacial depth extent of these stores, the soils remain the largest store of C, but our results suggest that the coastal marine sediments provide a more effective long-term store of OC_{terr} than the adjacent terrestrial environment. Any direct comparison of these stores must consider their longevity as effective C stores. Storage potential varies significantly from short-term storage within the living biomass (days to decades) to the long-term storage of OC in soils and marine sediments (10^3 years). A further important and underreported consideration is the long-term stability of these stores. The terrestrial stores are vulnerable to short and long-term environmental change such as soil erosion (Cummins et al., 2011) and fire (Davies et al., 2013), both of which are increasing in regularity with growing climatic and anthropogenic pressure. In contrast, the often restricted nature of fjord hydrography combined with their deep coastal water setting can provide the OC stored in these sediments with a far greater level of protection than the adjacent catchments. This is not to imply that these marine stores will not be affected by recent anthropogenically driven changes but rather that in the short to medium term they will be buffered from the immediate impacts of these alterations to the wider environment. It is therefore important to recognize that the subsidy of OC_{terr} from an inherently vulnerable terrestrial system to a potentially stable long-term store within coastal sediments may provide a previously unrecognized climate-regulating service.

To understand the OC_{terr} subsidy to the coastal sedimentary environment, a good understanding of the processes that govern the transfer of C from one pool to another (i.e., terrestrial to marine) is required. Examination of our results from the integrated catchment and fjord system suggest that different processes govern the different environments. The terrestrial environment is dominated by cyclical processes (Kirkels et al., 2014), whereby the catchment itself constantly accumulates and erodes OC_{terr} stores while approaching a state of relative equilibrium. By contrast, the mechanisms that control and govern the postglacial sedimentary storage of OC_{terr} in fjords are largely cumulative (Smeaton et al., 2016; Smith et al., 2015), with fluvial and hillside processes subsidizing the sediment with OC_{terr} at the same time that primary production within the fjord and adjacent seas contributes an OC_{mar} source to the same sediments. If we consider the fjord and its catchment as a single, integrated system, the OC_{terr} subsidy to coastal marine sediments becomes increasingly important as the main mechanism by which the system as a whole stores

OC_{terr} over the long term (i.e., interglacial periods). We hypothesize that the catchment may be approaching a state of equilibrium and that the annual new production of OC_{terr} may not be adding significantly to the terrestrial store. Ultimately, these systems may successfully store more OC_{terr} through the subsidy of C from the catchment to the coastal sediments. Through this process of C subsidy, coastal marine sediments become effective and stable long-term repositories for OC_{terr} storage. This new understanding highlights the growing imperative to critically reevaluate terrestrial OC *losses* as net *gains* in the marine environment.

While the subsidy of coastal sediments with OC_{terr} may represent an effective mechanism for the long-term storage of OC, it remains unclear how these marine stores will respond to increased anthropogenic pressure on their adjacent catchments. Such pressures will certainly act to disturb catchment soils and vegetation and should therefore act to increase the soil, nutrient, and associated OC_{terr} flux to the adjacent coastal ocean. It remains something of an open question as to whether or not the anthropogenically driven OC_{terr} flux has enhanced C storage in coastal marine sediments, but it seems likely that this is happening and that it requires urgent quantification.

6. Conclusions

A comparison of Loch Sunart with other midlatitude fjords (Edwards & Sharples, 1986) and to fjords with similar glacial history (New Zealand, Norway, and Canada) (Syvitski & Shaw, 1995) suggests that our findings are likely to be prevalent throughout many of these middle- and high-latitude coastal sedimentary systems. This work suggests that fjordic sediments contain significant stores of OC_{terr} and could potentially provide a largely unrecognized climate regulation service through the subsidy of OC_{terr} from the catchment to the adjacent marine environment. Within these environments it appears that OC_{terr} transfers from land to sea in recent times have been effectively transferring OC_{terr} from an inherently unstable store within the catchment to a far more stable and long-lived OC_{terr} store in marine sediments. Ironically, this probably means that fjordic marine sediment systems are a more effective long-term store of OC_{terr} than their adjacent terrestrial catchments.

Acknowledgments

This work was supported by the Natural Environment Research Council (grant NE/L501852/1) with additional support from the NERC Life Science Mass Spectrometry Facility (CEH_L_098_11_2015) and the NERC Geophysical Equipment Facility (NGGFSC Minor Loan 1031). The authors would like to thank Alix Cage (Keele University), Simon Mclaughlin (RSPB), Peter Madden (Forestry Commission Scotland), Ben McKeown (Ardnamurchan Estate), and Marine Scotland Science for providing and assisting in the collection of sample material. Additionally, we would like to thank Helen Grant (NERC LSMSF) for laboratory support. Finally, we thank two anonymous reviewer and Miguel Goni whose insightful comments improved this paper. All original data are attached as supporting information to this manuscript. In addition, all data will be made publicly available at the NERC National Geoscience Data Centre (NGDC).

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