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A Sedimentary Carbon Inventory for a Scottish Sea Loch (Fjord): An Integrated Geochemical and Geophysical Approach.

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Abstract

Quantifying sedimentary carbon stocks in the coastal ocean is key to improving our understanding of long-term storage of carbon in the coastal ocean and to further constraining the global carbon cycle. Here we present a methodological approach which combines seismic geophysics and geochemical measurements to quantitatively estimate the total stock of carbon held within marine sediment. Through the application of this methodology to Loch Sunart a sea loch (fjord) on the west coast of Scotland we have created the first sedimentary carbon inventory for a fjordic system. The sediment of Loch Sunart holds 26.88 ± 0.52 Mt of carbon split between 11.05 ± 0.23 Mt and 15.02 ± 0.35 Mt of organic and inorganic carbon respectively. This quantative estimate of carbon stored in Loch Sunart in significantly higher than previous estimates. Through comparison to Scottish peatland carbon stocks we have determined that Loch Sunart on a per are basis is a significantly more effective store of carbon. This initial work supports the concept that fjords are important environments for the burial and long-term storage of carbon and therefore should be considered as unique environments while considering coastal carbon stocks.

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1 Introduction

The rising prominence of Blue Carbon, i.e. carbon (C) which is stored in coastal ecosystems, notably, mangroves, tidal marshes, seagrass meadows and sediments has forced a reassessment of our knowledge of C in the coastal ocean (Nellemann et al., 2009). In recent years there have been a number of reviews (Bauer et al., 2013 & Cai et al., 2011) highlighting knowledge gaps and the limited understanding of both the C sources and sinks in the coastal ocean (Bauer et al. 2013). Quantifying the stores of C in the coastal ocean is the first step to a better understanding of coastal carbon dynamics. Global C burial in the coastal zone is estimated in the region of 237.6 Tg yr^{-1} with approximately 126.2 Tg yr^{-1} of C being buried in depositional areas i.e. estuaries and the shelf (Duarte et al., 2005). The lack of regional and national coastal sedimentary C inventories means these global estimates cannot be confirmed or further constrained.

One of the rare examples of a national marine C inventory was carried out by Burrows et al. (2014) producing initial estimates of Blue Carbon in Scottish territorial waters; they calculated that these waters stored 1,757 Mt C, with coastal and offshore sediments acting as the main repositories. Burrows et al. (2014) suggested that the majority of this organic carbon (OC) was held in sea loch (fjord) sediments.

It has been long known that fjords are important stores of C (Syvitski et al., 1987) and that C burial in sediments is the most significant mechanism of long-term (>1000years) OC sequestration in the coastal ocean setting (Hedges et al., 1995).. The work by Smith et al. (2015) has also shown that globally fjordic systems act as a CO_2 "buffer" by efficiently capturing and burying labile terrestrially derived OC and preventing it from entering the adjacent ocean system where it is prone to recycling. These authors have calculated that 11% of annual global marine carbon sequestration occurs within fjords.

Despite these findings, much of the global research to assess and quantify C stocks is disproportionately skewed towards the terrestrial environment (e.g. Yu et al., 2010). This trend is also found at the regional scale where there have been multiple studies quantifying the carbon held within Scottish soils (Aitkenhead et al., 2016, Bradley et al., 2005, & Chapman et al., 2013) and peats (Aitkenhead et al., 2016, Howard et al., 1995, Cannell et al., 1999 & Chapman et al. 2009).

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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In addition to the challenges of access and cost to sample these environments when compared to the adjacent terrestrial environment, it might also be argued that the sparsity of marine sedimentary C inventories is due to the lack of a robust methodology to quantify these C stores. Syvitski et al. (1987) commented that "the development of a methodological approach to quantify the C in the sediment of a fjord must be a priority", yet in the subsequent years there has been relatively little progress towards this goal.

The absence of a robust methodology to quantify the C held in marine sediments is illustrated by Burrows et al. (2014), who estimated that there is 0.34 Mt OC stored in the sediments of Scottish sea lochs (fjords). However, these calculations only take into account an estimate of OC in the top 10 cm of sediment, despite the fact that sediment depths of >25 m are common in Scottish sea lochs (Baltzer et al., 2010, Howe et al. 2002). Therefore, it is likely that current best estimates of the quantity of OC has been significantly underestimated and that the presence of significant quantities of inorganic carbon (IC) held within sea lochs sediments has been overlooked.

This study combines geochemical, geophysical and geochronological techniques to produce a methodology capable of delivering quantitative first-order calculations of the mass of C stored within the sediment of a sea loch and, potentially, of achieving the goal set out by Syvitski et al. (1987). This work provides the first carbon inventory for a fjord and further develops the concept of these fjords as being globally important sites for the burial of C as set out by Smith et al. (2015).

2 Material & Methods

2.1 Study Area

Loch Sunart is a sea loch on the West coast of Scotland (Fig.1). The loch is 30.7 km long and covers an area of 47.3 km^2 with a maximum depth of 145 m. It consists of three basins separated by shallower, rock sills. The inner basin is separated from the middle basin by a sill at approximately 6 m depth, while the middle and outer basins are separated by a sill at approximately 31 m depth (Edwards & Sharples, 1986; Gillibrand et al., 2005). The silled nature of the bathymetry allows the loch to act as a natural sediment trap for both terrestrial and marine derived materials (e.g. Nørgaard-Pedersen et al., 2006).

Loch Sunart's catchment covers 299 km^2; the main tributaries of the loch are the Rivers Carnoch and Strontain; the latter has a mean daily discharge of 1409 m^3 (2009-2013). The

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Manuscript under review for journal Biogeosciences
Published: 17 June 2016

mean annual precipitation in Loch Sunart's catchment is 2632 ± 262 mm (Capell et al., 2013). The combination of small catchment size and high precipitation means that the flow network is sensitive to precipitation changes which can result in a flashy flow regime (Gillibrand et al., 2005).

The catchment is largely dominated by high relief and poorly developed soils. The bedrock consists primarily of igneous and metamorphic rocks, overlain by gley and podzol soils with limited peat in the upper catchment (Soil Survey of Scotland., 1981). Exposed rock is common on the steep slopes; much of the catchment's vegetation can be found by streams or on the loch shore and is dominated by both commercial forestry and natural woodlands; there is only very limited agriculture within the catchment. The combination of steep, exposed slopes, poorly developed soil, a reactive river network and poorly developed vegetation typically results in high surface runoff and sediment transport (Hilton et al., 2011).

The characteristics of Loch Sunart and its catchment are representative of sea lochs across mainland Scotland (Edwards & Sharples., 1986), with the possible exception of Loch Etive which has a hypoxic upper basin (Friedrich et al., 2014). The sea lochs of the Scottish Islands (Shetland, Orkney & the Western Isles) differ from their mainland counterparts in that they are generally shallower and have catchments characterised by lower relief and are largely dominated by peat or peaty soil (Soil Survey of Scotland., 1981). Syvitski and Shaw's (1995) table of generalised fjord characteristics allows us to compare the sea lochs of mainland Scotland to other fjordic systems globally. The fjords of the Norwegian mainland, Canada and the Fiordland, New Zealand (Hinojosa et al., 2014) are characterised by similar climate, geomorphology, river discharge, basin water temperature and sedimentation rate as the sea lochs of Scotland. The fjords of mainland Scotland differ significantly from those in Greenland, Alaska, Svalbard and the Canadian Arctic, many of which still have active glaciers, resulting in very different sediment input regime.

2.2 Seismic Data Acquisition and Processing

2.2.1 Data Acquisition

A seismic geophysical survey of Loch Sunart took place in 2002 aboard the RV *Envoy* (Fig.2). A Seistec Boomer System was used to create seismic profile data throughout the loch. The data were recorded using an Elics-Delph data acquisition system coupled to the Differential Global Positioning System (DGPS). The Boomer system operated on a frequency

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of 1 to 10 kHz and had a pulse duration of 75 to 250 ms at a power of 150 J. The system has a depth resolution of 25 cm and can penetrate 100 m in soft sediment (Simpkin & Davies 1983). A total of 34 transects of the loch were acquired (Fig.2). The survey achieved an average penetration of 50 m; gas blanking prevented the signal from penetrating the sediment in some areas (Baltzer et al., 2010).

2.2.2 Defining Sedimentary Horizons

Each seismic profile was combined with the DGPS data and processed with the Petrel (Schlumberger) software package. Subsequent analysis was undertaken using the open source SeiSee (DMNG) software package. Initial interpolation, following Baltzer et al.'s (2010) methodology, defined the different seismic horizons (H) and the layers between the horizons which are defined as seismic units (U) numbered 1 to 3 from the basement horizon upwards (Fig 3). The compilation of the horizons and units allows the construction of an equivalent seismic stratigraphy for each sediment core and the loch as a whole.

Using SeiSee, points were picked along each of the four horizons creating polylines. Each polyline was split into points at 0.25 m intervals and each point was assigned an x,y,z coordinate that represents its geographic location and depth (relative to mean sea level).

2.3 Sediment Sampling

Eight sediment cores (Table.1) where collected from Loch Sunart (Fig.1) in 2001 using a gravity corer (GC) as part of the HOLSMEER project. This was supplemented with further sampling on a follow-up cruise on-board the *RV Calanus* in August 2013 where a short GC was collected to fill a gap between the original coring sites. These cores capture the post-glacial history of sediment accumulation within the loch, as confirmed by ^{14}C basal dates. Additionally, we accessed the lower sections of core MD04 2833 which was recovered using the CALYPSO giant piston corer from the *RV Marion Dufresne* in July 2004 as part of the IMAGES project. Sampling of Section VIII (1050-1200 cm) of MD04 2833 was undertaken to obtain sediment of inferred glacial origin for geochemical analysis (Baltzer et al., 2010).

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

2.4 Sediment Analysis

2.4.1 Physical Characteristics

Detailed sediment logging was undertaken for each of the cores (Supplementary Material). The gravity cores were sub-sampled at 10 cm intervals and high resolution sampling at 1 cm intervals was undertaken on the short core. Section VIII of glacial sediment core MD04-2833 was sub-sampled at 12 cm intervals. Each sub-sample was split for physical property and geochemical analyses. The wet (WBD) and dry bulk density (DBD) of the sediment was calculated following Dadey et al. (1992).

2.4.2 Bulk Elemental Analysis

To quantify the total carbon (TC) content, each sub-sample was freeze-dried and milled to a fine powder. A 20 ± 2 mg aliquot was placed in a tin capsule and measured on a COSTECH Elemental Analyser (EA) calibrated with acetanilide (Verardo et al,1990, Nieuwenhuize et al., 1994). Precision of the analysis is estimated from repeat analysis of standard reference material B2178 (Medium Organic content standard, Elemental Micro analysis, UK) C = 0.07% N = 0.02% (n = 8).

To quantify OC, the process was repeated with the addition of H_2SO_3 to remove the inorganic carbon (IC). After acidification vessels were placed in a vacuum desiccator to remove any remaining CO_2 and the sample was then freeze-dried to remove the H_2SO_3. IC was calculated from the difference between TC and OC measurements. The mean standard deviation of TC and OC triplicate measurements (n=10) were 0.04 %, 0.17 % respectively.

2.4.3 Sediment Geochronology

Basal radiocarbon dates for five of the gravity cores were obtained by accelerator mass spectrometer (AMS) radiocarbon dating of marine carbonate material (mollusc). This was carried out at the University of Aarhus, Denmark (AAR), Centre of Accelerator Mass Spectrometry, USA (CAMS) and the NERC Radiocarbon Laboratory, Scotland (SUERC). The radiocarbon dating was used to validate the Holocene chronology of the seismic stratigraphy. A single MD04-2833 sample was processed at Laval University, Canada (UL) to confirm that the sediment was early post-glacial in age. Dates were calibrated using OxCal 4.2.4 age modelling software (Bronk Ramsey., 2009 & Bronk Ramsey & Lee., 2013)

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Published: 17 June 2016

applying the Marine13 curve (Reimer et al., 2013) and the regional marine radiocarbon reservoir age correction: ΔR value of -26 ± 14 yr (Cage et al. 2006).

Sediment accumulation rates (SAR) were calculated as an approximation for the whole core using basal ages and a linear interpolation to the core top, assuming a contemporary surface. We recognise that the calculations will be crude and do not take into consideration factors such as compaction and possible changes in sedimentation rate, but these calculations provide initial insight into the variability of SARs within the loch and allow first-order C accumulation rates to be estimated.

2.5 Sediment Quantification & Characterisation

2.5.1 Digital Terrain Models (DTM)

The points collected from each seismic horizon were connected to form a DTM of that horizon. This was achieved using spatial modelling techniques in ArcGIS. The compiled *x,y,z* data were statistically tested to determine the gridding technique best suited to the interpolation of the data. Eleven gridding techniques where subjected to cross validation (Chiles & Delfiner 1999)(Supplementary Material).. The residual Z mean value and standard deviation were examined; the technique with the lowest residual Z mean and standard deviation for each horizon (and the data set as a whole) was chosen as the gridding technique best suited to the interpolation of the data. Kriging (with linear interpolation) (Cressie, 1990) with a 100 by 1000 node structure performed best and was chosen to create computationally efficient DTMs for each seismic horizon.

2.5.2 Volumetric Calculations

The horizon DTM grids were used to calculate the volume of sediment in each seismic unit and, by extension, within the loch as a whole. By subtracting one DTM grid from another (e.g. Surface DTM – Bedrock DTM) the volume between the grids was calculated. Three different numerical integration algorithms were used for this calculation (Eq.1,2,3). The net volume is reported as the mean of these three calculations. In the following formulae Δx represents the grid column spacing, Δy represents the grid row spacing and $G_{i,j}$ represents the grid node value in row *i* and column *j* (Press et al., 1988).

Trapezoidal Rule

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

The pattern of coefficients is {1,2,2,2,…,2,2,1}: (1)

$$A_i = \frac{\Delta x}{2}[G_{i,1} + 2G_{i,2} + 2G_{i,3} \cdots + 2G_{i,nCol-1} + G_{i,nCol}]$$

$$Volume \approx \frac{\Delta y}{2}[A_1 + 2A_2 + 2A_3 + \cdots 2A_{nCol-1} + A_{nCol}]$$

Extended Simpson's Rule

The pattern of coefficients is {1,4,2,4,2,4,2,…,4,2,1}: (2)

$$A_i = \frac{\Delta x}{3}[G_{i,1} + 4G_{i,2} + 2G_{i,3} + 4G_{i,4} + \cdots + 2G_{i,nCol-1} + G_{i,nCol}]$$

$$Volume \approx \frac{\Delta y}{3}[A_1 + 4A_2 + 2A_3 + 4A_3 + \cdots + 2A_{nCol-1} + A_{nCol}]$$

Extended Simpson's 3/8 Rule

The pattern of coefficients is {1,3,3,2,3,3,2,…,3,3,2,1}: (3)

$$A_i = \frac{3\Delta x}{8}[G_{i,1} + 3G_{i,2} + 3G_{i,3} + 2G_{1,4} + \cdots + 2G_{i,NCol-1} + G_{i,nCol}]$$

$$Volume \approx \frac{3\Delta y}{8}[A_1 + 3A_2 + 3A_3 + 2A_3 + \cdots + 2A_{nCol-1} + A_{nCol}]$$

2.5.3 Sediment Mass Quantification

The mean dry bulk density (DBD) for each seismic unit was calculated and assigned to the equivalent seismic units within each core. The spatial distribution of the DBD for each seismic unit was modelled, again using Kriging (with linear interpolation). The resulting contour plot was integrated with the volumetric model for each seismic unit to calculate the dry mass of the sediment held within that seismic unit. The integration process calculates the volume of sediment held within each of the DBD contours and multiplies that volume with the associated DBD value to calculate the mass of sediment.

2.5.4 Sedimentary Carbon Quantification

The same methodology used to integrate the volume and density data was used to combine bulk elemental data with the sediment dry mass calculations. Mean values for TC, OC and IC in each seismic unit were assigned to the seismic units from the available core data. Kriging (with linear interpolation) was again used to create contour maps representing the quantity of TC, OC and IC in each seismic unit and the mass of sediment held between the contours was multiplied by the percentage of the element quantifying the mass of TC, OC and IC held

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

within the loch's sediment. Finally, we calculated how effectively the loch stores C (C_{eff}) as a depth-integrated average value per km^2 for both the post-glacial and glacial derived sediments. This measure allows the loch's C stores to be directly compared with other C stores (peatlands, soil, etc.).

3 Results

3.1 Seismic Interpretation

3.1.1 Seismic Horizons and Units

Four horizons were identified throughout the loch (Fig.3): these represent the basement (H1) and the sediment water interface (H4) with two intermediate horizons (H2 & H3). Core stratigraphy (Baltzer et al. 2010) indicates that H2 divides the post-glacial and glacial sediment; while H3 splits the post-glacial sediment into two units. The seismic data displays a fifth horizon between H1 and H2 which is only present in the inner basin and partially in the middle basin. We interpret this as glacial sediment from the Younger Dryas, as confirmed by radiocarbon dating (Baltzer et al., 2010, Mokeddem et al. 2010); for the purposes of this paper, the horizon was amalgamated with H2.

A seismic stratigraphy was developed based on these horizons (Fig.3). U1 is interpreted as glacial sediment based on the observation of the short, discontinuous seismic reflections which are synonymous with poorly sorted material; the unit varies in thickness but never drops below a minimum thickness of 10 m. U2 is found throughout the loch with an average thickness of 5 – 10 m; the unit drapes over U1. U3 is the uppermost unit and has a homogenous thickness of around 1m; it is characterised by laminated acoustic reflections. Both U2 and U3 are interpreted as post-glacial infill of the loch; though clear in the seismic geophysics the boundary between U2 and U3 is poorly defined in the sediment lithology (Supplementary Material). Similar patterns in seismic stratigraphy have been observed throughout the west coast of Scotland (Binns et al., 1974a, b, Boulton et al., 1981 and Howe et al., 2002).

We compared our interpretation of the seismic data to the seismic interpretation of Baltzer et al., (2010); this exercise was designed to test the replicability of our interpretation and allow potential uncertainties in the seismic interpolation to be built into our future applications. The

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

comparison identified small differences in the depth of H1 (-0.17 m), H2 (+0.34) & H3 (-0.22 m). These differences were integrated into the volumetric calculations as an error term.

3.2 Sediment Geochronology

Calibrated radiocarbon dates for the gravity cores (Table.2) indicate that these cores are comprised of sediment accumulated during the post-glacial period (Holocene). The age of the deeper basal sediment of MD04-2833(Section VIII) was confirmed through dating of a mollusc (*Pecten maximus*); the calibrated age was 17041 ± 312 cal BP which, combined with the characteristic glacial core lithology of poorly sorted sedimentary material, indicates that this basal sediment of MD04-2833 was deposited by the retreat of the British ice sheet (BIS) at the end of the last glacial period 13500 to 17000 cal BP (Clark et al., 2010, Scourse et al., 2009, Wilson et al. 2002).

Through comparison of the chronologies to the seismic stratigraphy we can test the interpolation and further constrain the age of each seismic unit. The seismic unit for the equivalent depth of each of the radiocarbon samples has been compiled (Table.2), then compared to the seismic unit that the sample would fall into based on age alone as per the Baltzer et al. (2010) chronostragraphy. Of the 18 samples tested, 15 have a matched pair of seismic units; the three that do not have corresponding seismic units are all from GC023, suggesting a problem with the dating of this core rather than the interpolation of the seismic geophysics. This test signifies that our interpolation of the seismic geophysics is accurate and that the chronostratigraphy developed for MD04-2833 (Baltzer et al., 2010) can be applied throughout Loch Sunart. The seismic interpolation and the dated samples confirm that both U2 and U3 are postglacial in origin. We can further constrain the age of the seismic units with U2 representing the early to mid-Holocene and U3 mid to late Holocene in age.

SARs vary between the sedimentary basins, with the most rapid rates in the middle basin as indicated by GC023 (0.12 ± 0.026 cm yr^{-1}). The inner basin has a similar SAR (0.089 ± 0.026 cm yr^{-1}) to the middle basin, but accumulation rates were significantly slower in the outer basin, as illustrated by GC011 (0.017 ± 0.006 cm yr^{-1}).

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

3.3 Sediment Analysis

3.3.1 Bulk Density Measurement

Mean DBD was calculated for U1, U2 and U3 from each core. Figure 4 displays the DBD results, which are arranged to mirror the spatial distribution of the cores, from the inner basin to the outer basin. U1 sediment is characterised by the single section of MD04-2833, which has a mean DBD of 2.19 ± 0.09 g cm^{-3}. This is within the range of other northern hemisphere fjords (Pedersen et al. 2012, Forwick et al. 2010 and Baeten et al. 2010). DBD increases down each core as a result of sediment dewatering in response to compaction. GC011 is the only core where U3 has a higher DBD than U2 most likely due to large quantities of shell in the upper part of the core. U1 has the highest DBD; this reflects both the type of sediment deposited during glacial retreat and long-term compaction over the post-glacial period.

3.3.2 Bulk Elemental Analysis

The mean quantity OC and IC has been calculated for U1, U2 and U3 (Fig.5). Again values for U1 have been calculated using basal sediments of MD04-2833 (Section VIII). Clear trends emerge from the data, with U3 always containing a greater quantity of OC than U2, while the proportion of sedimentary OC generally decreases away from the inner basin. The opposite is true for sedimentary IC, which generally increases away from the inner basin. Sediment Quantification & Characterisation

3.3.3 Digital Terrain Models (DTMs)

The interpolation of the seismic profiles led to the creation of four DTMs (Fig.6) which represent horizons H1 to H4. Both H4 and H3 follow the trajectory with minimal deviation, primarily because U3 has a uniform depth of 1 m throughout the loch. Differences in depth between H3, H2 and H1 are far more variable. The inner basin shows the least change in depth between the horizons, although both H2 and H1 deepen before the sill (Fig.5). The middle basin displays the greatest depth differences between the horizons and also where the majority of the sediment is stored. Patterns in the outer basin are similar to those in the middle basin, especially where the two meet. Horizon depths become less variable towards the seaward direction.

To determine the accuracy of the models, the DTM for H4 was compared to an existing high-resolution bathymetric model of the loch (Bates et al. 2004). The coordinates (x,y,z) of key

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

high and low points (n=12) were compared between surveys; the mean divergence between surveys were calculated as *x*: -0.56 m , *y*: -0.81 , *z*: 0.21. Although the H4 DTM slightly negatively offsets the *x*,*y* and overestimates the *z* coordinates of these points, the general location and pattern of these seabed features compare favourably.

3.3.4 Volumetric Modelling

The DTMs and numerical integration algorithms were combined to calculate the volume of sediment held within each seismic unit. This was further broken down by basin and into post-glacial (U2 & U3) and glacial (U1) derived sediment (Table.3). The loch as a whole contains a greater volume of glacial (599731882 m^3 ± 1.89 %) than post-glacial sediment (530872293 m^3 ± 7.39 %). The outer basin is the only area where this trend is reversed. Comparison of the three basins indicates that the middle basin contains the greatest combined (post-glacial + glacial) volume of sediment (30409301.04 m^3 ± 5.30 %) followed by the outer (16039257.2 m^3 ± 5.74 %) and inner basins (4171662.46 m^3 ± 4.48 %).

3.3.5 Sediment Mass Quantification

The mean DBD for U2 and U3 were modelled (Fig.7) to determine the variability in spatial distribution throughout the loch. A similar spatial pattern of DBD is found in both U2 and U3; the DBD is lowest in the inner basin (U2: 0.47 g cm^{-3}, U3: 0.59 g cm^{-3}) rising out through the middle basin where it peaks at 1.75 g cm^{-3} and 1.67 g cm^{-3} for U2 and U3 respectively. The transition between the middle and outer basins is characterised with low DBD values (U2: 0.72 g cm^{-3}, U3: 0.91 g cm^{-3}); from this low point the DBD rises towards the seaward end of the loch.

The model output was integrated with the volumetric data to calculate the mass of sediment held within post-glacial sediment (Table 4). Since we have a single mean value for DBD for U1 we applied this throughout the loch to calculate the mass of sediment held within this unit. The loch holds a total of 1928.26 ± 7.29 Mt of sediment which is split into 652.09 ± 6.62 Mt of post-glacial and 1276.17 ± 8.93 Mt of glacial sediment. The inner basin holds the least sediment followed by the outer basin with the middle basin acting as the main store of sediment in Loch Sunart.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

3.3.6 Sedimentary Carbon Quantification

Using a similar approach, the mean OC and IC were spatially modelled throughout the loch. The output for U3 is illustrated in Figure 8. As before, the model outputs for U2 and U3 were integrated with the sediment mass data in order to quantify the mass of TC, OC and IC held within the post-glacial and glacial sediment (Table.4). Single mean values for TC, OC and IC were again used to calculate their respective mass of C within the sediment of U1.

The sediment of Loch Sunart holds a significant quantity of C (26.88 ± 0.52 Mt) split between OC (11.05 ± 0.23 Mt) and IC (15.02 ± 0.35 Mt). Though a greater mass of sediment is held within the glacial sediment component, it is the post-glacial sediments which hold the largest quantity of C (19.88 ± 0.27 Mt). The quantity of C held within each of Loch Sunart's basins varies; the lowest amount is found in the inner basin (2.12 ± 0.45 Mt) followed by the outer basin (6.70 ± 0.64 Mt). The sediment of middle basin holds significantly more C than both the inner and outer basins combined; with 18.05 ± 0.66 Mt C stored in these sediments indicating that the middle basin is the main repository for sedimentary C in Loch Sunart.

How effectively the loch stores C is measured by the C_{eff} (Table.5) and the OC:IC ratio. Loch Sunart is characterised by an OC:IC ratio of 0.74 and has a C_{eff} of 0.568 Mt C km^{-2}, which can be further broken down to a post-glacial C_{eff} of 0.42 Mt C km^{-2} and a glacial C_{eff} of 0.148 Mt C km^{-2}. The effective C storage can also be illustrated at the individual basin level with the post-glacial sediments of the inner, middle and outer basins characterised by OC:IC ratios of 0.42, 1.00 and -0.42, illustrating the transition from OC as the dominant component of the sediment in the upper loch to an IC-dominated sediment at the seaward end of the loch. The middle basin is the most effective at storing post-glacial OC followed by the inner and outer basin; similarly the middle basin is most effective at storing IC, but in contrast to the effective storage of OC, the outer basin ranks second followed by the inner basin for IC. The glacial material held within the loch as a whole is characterised by an OC:IC ratio of 0.42 with a mean OC_{eff} 0.044 Mt km^{-2} and IC_{eff} 0.104 Mt km^{-2}.

3.4 A Methodology for Estimating Sedimentary Carbon and Attributing Uncertainty Estimates

The joint geophysical and geochemical methodology outlined (Fig.9) provides a robust approach to allow the first quantification of sedimentary C stocks in a fjord setting. An important part of estimating sedimentary C stocks should be the quantification of uncertainty

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

associated with these estimates. There are several types of uncertainty that can influence sedimentary carbon estimations (Fig.9), including interpolation, algorithmic, analytical, sampling and extrapolation uncertainty. Several of these types of uncertainties are easily dealt with statistically, for example the analytical uncertainties can be quantified through triplicate measurements. The sampling uncertainty of a stratigraphic sequence (i.e. spatial variability of C content in relation to sampling density) can be overcome by calculating the mean and standard deviation to create composite values that are representative of the entire sediment or seismic unit. We integrated the quantifiable uncertainties at each calculation step (Fig.4). By calculating composite standard deviations we are able to propagate the uncertainties throughout the C quantification process. In the interpolation of the seismic geophysics, it is difficult to fully quantify the uncertainty involved in the process. Bond et al. (2007) set out a 5 step framework designed to reduce uncertainty in this process. We utilised the framework of Bond et al. (2007) and additionally integrated a validation step using radiocarbon dating of sedimentary cores (See Section 3.2). This allows us to reduce the uncertainties associated with the seismic interpretation, although we recognise that some uncertainty remains (e.g. highly variable patterns of depth) which cannot be fully quantified. Within this framework of uncertainty, we consider our method to give a robust estimate for the carbon socks present.

4 Discussion: A new Sedimentary C Inventory for Scottish Coastal Waters

The development of this methodology has allowed the estimation of the sedimentary C stocks stored in Loch Sunart. The sediment which has been uncounted for within Loch Sunart holds 26.88 Mt C.

The only directly comparable estimation for sedimentary C stocks is the report by Burrows et al. (2014), where they calculated that 0.34 Mt OC was stored in all Scottish sea lochs. In comparison, our findings estimate that Loch Sunart alone holds 11.05 Mt OC. However, Burrows et al. (2014) focused on the top 10 cm of sediment because data availability and the lack of a robust methodology made it impossible to calculate the entire sedimentary C stock; this has resulted in a significant underestimation of the quantity of C held within the sediment of these lochs. Additionally, Borrows et al. (2014) did not consider IC to be a major component in these sediments; instead the authors focused on Scottish sea lochs largely as OC stores. In contrast, our results demonstrate that Loch Sunart stores 15.02 Mt IC in comparison to 11.05 Mt OC. The general lack IC data for Scottish sea lochs makes it difficult to assess how representative Loch Sunart is of these coastal sedimentary IC stores; our results do

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

highlight the potential significance of IC as a major component of sedimentary C stores in these depositional environments. Our results also highlight that sea lochs (and probably fjords in general) act as an OC-rich sediment transition zone between terrestrial and oceanic environments.

Loch Sunart's sediment currently holds 11.05 Mt OC which has been trapped and prevented from reaching the adjacent shelf sea. This OC trapping in the coastal zone may reduce reworking and remineralisation of the material which would have otherwise resulted in the release of CO_2 through biotic processes (Smith et al., 2015). This 11.05 Mt OC is equivalent to 40.93 Mt CO_2e (carbon dioxide equivalent). As a whole, the sediment within Loch Sunart stores 99.56 Mt CO_2e which is almost double Scotland's total greenhouse gas emission for 2013 which reached an estimated 53 Mt CO_2e (Scottish Government, 2015).

Globally, the terrestrial C stores have received much more attention than their marine counterparts; with significant focus on quantifying the forest (Köhl et al., 2015) and soil C stocks (Köchy et al., 2015, Scharlemann et al., 2014). The work by Duarte et al. (2005) to compile the known stocks and burial rate of C in the coastal environment highlighted that the coastal ocean is a large store of carbon, which remains poorly understood; from this work the concept of Blue Carbon arose (Nellemann et al., 2009). The focus of Duarte et al. (2005) was to highlight that the vegetated coastal zones (i.e. saltmarsh, seagrass and mangroves) bury and store significant quantities of C and that these stores should be further investigated and recognised in policy outputs, but they largely overlooked the importance of what they described as depositional area (estuaries and the shelf sea) as long-term repositories of OC detritus from the vegetated coastal environment (Krumhansl et al. 2012). These authors recognised that coastal (and shelf?) depositional areas are important stores of sedimentary C globally, yet no consideration is given to how these areas vary in terms of their capacity to store C. Conceptually, if we consider the types of estuaries (i.e. fjord, delta, coastal plain, bar-built and tectonic), it is clear that the characteristics of each type of estuary will impact the manner in which C is buried and stored, for example the restricted nature of fjords will be conducive to sediment capture and C storage compared to the more open estuarine types.

Our initial work suggests that the depositional area category could be further expanded upon to include fjords as a separate component and this concept is supported by Smith et al. (2015), who indicated that fjords are "hot-spots for OC burial" and should be considered separately from estuaries when investigating global ocean OC burial. Currently, there is insufficient data

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

globally to advocate fjords being categorised as a separate component in global coastal C stores; the standardised methodology outlined (Fig.4) provides a platform to investigate this concept further.

At the national level there has been a significant focus on quantifying Scottish soil C stocks, with much attention given to the peatlands (Aitkenhead & Coull., 2016, Bradley et al., 2005 & Chapman et al. 2009). Peat and other organic rich soils cover 66% of Scotland and account for 50% of all the United Kingdom's soil C stocks (Cummins et al., 2011). The Scottish peatlands store 1620 Mt C (Chapman et al., 2009) over an area of 17270 km^2, while the other soils hold 2110.9 Mt C over 60215 km^2 (Aitkenhead & Coull., 2016). In comparison to these figures, the quantity of C stored in Loch Sunart is small, but the loch itself only covers an area of 47.3 km^2. When the loch's C_{eff} is compared to how effectively Scotland's soils and peatland store C (Table.5) we can see that on a C amount per unit area basis Loch Sunart stores significantly more C than the soils of Scotland. The loch has a C_{eff} of 0.568 Mt C km^{-2} compared to 0.094 Mt C km^{-2} and 0.035 Mt C km^{-2} for the peatlands and other soils of Scotland. Our results suggest that Loch Sunart is one of the most effective stores of C in Scotland and highlights the potential of the sediment in these sea lochs to hold a significant quantity of C which has previously not been recognised. Many of these terrestrial C stores are, of course, vulnerable to rapid and long-term environmental change; the Scottish terrestrial C stocks are at risk from erosion (Cummins et al. 2011) and even fire (Davies et al. 2013), both of which are increasing in pace and frequency by anthropogenic activities. In comparison, a sea loch's geomorphology combined with its depth gives sedimentary C stores a level of protection not afforded to terrestrial C stores. This does not mean that the sedimentary C in sea lochs is invulnerable, but rather that it is buffered from the immediate effects of chemical, biological and physical environmental change. Little is currently known regarding the long-term stability of these stores.

The methodology outlined in this paper has given us a platform to calculate the carbon stocks within Loch Sunart and has the potential to be applied in other fjordic systems as well as environments with restricted sediment exchange processes, such as estuaries and freshwater lakes, as well as artificial systems such as reservoirs and irrigation pools.

5 Conclusion

The integration of the geochemical and geophysical techniques outlined provides a robust and repeatable methodology to quantitatively calculate the volume of sediment and make first

order estimations of carbon stored within fjordic sediments. Using this methodology we have shown that Loch Sunart holds 26.88 Mt C which is almost double the quantity of Scottish CO_2 emissions for 2013. Although this is small in comparison with Scotland's peatland and soil C stocks, per unit area Loch Sunart is a more effective store of both OC and IC than Scotland's soils or peatlands. The results from this study suggest that the sediment in Scotland's 110 sea lochs (Edwards and Sharples et al., 1986) represent a potentially significant, yet currently largely unaccounted for repository for both OC and IC. These coastal settings trap and prevent the remineralisation of OC into the atmosphere. Additionally, the C held within these 110 sea lochs is likely to represent a significant part of Scotland's blue carbon capital that has not been considered at the marine ecosystem, global C cycle and wider policy levels. Without a better understanding of these stores of marine sedimentary C we will remain unable to fully quantify the coastal C cycle or the role that these fjordic environments play in buffering the release of CO_2 through the burial of C in these sediments. The future strategic use of this methodology within different fjord types and locations offers the potential to upscale and quantify the C held within all Scottish sea lochs and possibly begin to estimate the fjordic sedimentary C stores both at a national and global level.

Author Contribution

CS & WA conceived the research and wrote the initial manuscript, to which all co-authors contributed data or provided input. CS conducted the research as part of his PhD at the University of St Andrews, supervised by WA, AD and JH.

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Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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Manuscript under review for journal Biogeosciences
Published: 17 June 2016

Table.1. Details of the sediment cores extracted from Loch Sunart that were used in this study.

Core ID	Basin	Position (Lat, Long)	Water Depth (m)	Recovery (m)
GC009	Middle	56.672056, -5.867083	107	1.41
GC011	Outer	56.759861, -5.969639	91	2.45
GC013	Inner	56.681306, -5.629528	58	1.67
GC016	Inner	56.680944, -5.642333	58	0.56
GC020	Middle	56.704278, -5.751333	105	2.38
GC022	Middle	56.680333, -5.804944	120	2.46
GC023	Middle	56.665917, -5.840361	87	2.89
GC081	Middle	56.668972, -5.863278	58	3.63
GC01	Middle	56.696806, -5.704972	42	0.21
MD04 2833	Middle	56.665500, -5.859667	38	12

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

Table.2 Radiocarbon ages from Loch Sunart cores. Ages were calibrated using OxCal 4.2.4 (Bronk Ramsey., 2009 & Bronk Ramsey & Lee., 2013) with the Marine13 curve (Reimer et al. 2013) and regional correction of ΔR value of -26 ± 14 yr (Cage et al. 2006) . All ages are calibrated at 95.4% probability and the mean age has been determined from the minimum and maximum calibrated ages. Additionally; we list the seismic unit assigned to each equivalent (eqv.) depth of each sample and compare this to the age equivalent seismic unit based on Baltzer et al. (2010).

Laboratory Code	Core ID	Depth (cm)	^{14}C Age, BP (No Correction)	Calibrated ^{14}C Age (cal BP)	Seismic Unit	
					Depth eqv.	Age eqv.
AA-48108	GC009	140	9827 ± 49	10801 ± 93	U2	U2
SUERC 65990	GC011	60	2837 ± 35	2625 ± 66	U3	U3
SUERC 65991	GC011	120	9890 ± 38	10878 ± 87	U2	U3
SUERC 65992	GC011	170	11266 ± 40	12760 ± 61	U2	U2
AA-48109	GC011	231	12181 ± 58	13658 ± 90	U1	U1
AA-48107	GC013	113	1716 ± 32	1294 ± 35	U3	U3
SUERC 65995	GC016	30	1865 ± 35	1438 ± 51	U3	U3
SUERC 65994	GC020	9	683 ± 35	357 ± 44	U3	U3
SUERC 65993	GC020	19	3067 ± 37	2864 ± 57	U3	U3
AA-48106	GC020	126	11652 ± 74	13160 ± 90	U2	U2/U1
AA-51569	GC023	30	340± 60	64 ± 51	U3	U3
SUERC-681	GC023	49	1215 ± 47	788 ± 58	U3	U3
SUERC-677	GC023	58	1322 ± 43	886 ± 55	U3	U3
AA-51570	GC023	73	1430 ± 55	1011 ± 66	U3	U3
SUERC-679	GC023	111.5	1695 ± 57	1274 ± 59	U2	U3
SUERC-680	GC023	250	2180 ± 61	1801 ± 80	U2	U3
CAMS-82821	GC023	286	2425 ± 40	2099 ± 70	U2	U3
UL 2853	MD04-2833	745	14420 ± 210	17041 ± 312	U1	U1

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

Table.3 Sediment volume calculated as the mean of the three numerical integration algorithms; the error is reported as relative standard deviation (%RSD) which integrates the uncertainty in the seismic interpolation and the standard deviation of the numerical integration algorithms. The data is reported for the post-glacial (PG) and glacial (G) sediment at the basin level.

Basin	**Layer**	**Volume**	
		Mean (m^3)	%RSD
Inner	PG	2869825.90	6.48
	G	1301836.56	1.89
Middle	PG	23046267	7.26
	G	7363034.04	1.89
Outer	PG	13371884	7.90
	G	2667373.2	1.89
Loch Sunart	PG	530872293	7.39
	G	599731882	1.89
	Total	**1130604175.55**	**3.61**

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

Table.4 Mass of sediment held within Loch Sunart and the mass of total carbon (TC), organic carbon (OC) and inorganic carbon (IC) held with Loch Sunart's Sediment.

Basin	Layer	Mass (Mt)	TC (Mt)	OC (Mt)	IC (Mt)
Inner	PG	27.05 ± 2.98	1.31 ± 0.16	1.05 ± 0.14	0.26 ± 0.15
	G	126.65 ± 7.15	0.81 ± 0.62	0.24 ± 0.2	0.57 ± 0.41
Middle	PG	421.53 ± 7.26	14.08 ± 0.30	7.05 ± 0.25	7.03 ± 0.17
	G	738.31 ± 9.62	3.97 ± 0.88	1.17 ± 0.28	2.80 ± 0.57
Outer	PG	203.52 ± 11.10	4.49 ± 0.34	1.32 ± 0.14	3.15 ± 0.24
	G	411.22 ± 9.78	2.21 ± 0.84	0.65 ± 0.14	1.56 ± 0.57
Loch Sunart	PG	652.09 ± 6.62	19.88 ± 0.27	8.98 ± 0.21	10.09 ± 0.17
	G	1276.17 ± 8.93	7.00 ± 0.82	2.07 ± 0.27	4.93 ± 0.55
	Total	**1928.26 ± 7.29**	**26.88 ± 0.52**	**11.05 ± 0.23**	**15.02 ± 0.35**

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
Published: 17 June 2016

Table.5 The effective C storage (C_{eff}) of Loch Sunart's postglacial and glacial sediment in comparison to Scottish terrestrial C stores.

C Inventories	Area (km^2)	TC (Mt)	C_{eff} (Mt km^{-2})	OC_{eff} (Mt km^{-2})	IC_{eff} (Mt km^{-2})	Reference
			Postglacial			
Inner Basin	5.5	1.31	0.238	0.191	0.047	
Middle Basin	24.7	14.08	0.57	0.285	0.284	
Outer Basin	17.1	4.49	0.263	0.077	0.184	
			Glacial			
Inner Basin	5.5	0.81	0.147	0.044	0.104	
Middle Basin	24.7	3.97	0.161	0.047	0.113	
Outer Basin	17.1	2.21	0.129	0.038	0.091	
Postglacial	47.3	19.88	0.42	0.189	0.213	
Glacial	47.3	7.00	0.148	0.044	0.104	
Loch Sunart	47.3	26.88	0.568	0.234	0.318	
			2 m Depth			
Peatlands*	17270	1620		0.094		Chapman et al., 2009
Organo-Mineral Soil*		754				Bradley et al., 2005
Mineral Soil*		498				
			1 m Depth			
Peat	17369	813.9		0.047		Aitkenhead & Coull ,2016
Alluvial Soil	1657	40.8		0.025		
Alpine Soil	3825	145.7		0.038		
Bare Ground	1672	50.5		0.030		
Brown Earth	15971	590.3		0.037		
Gley	15963	645.4		0.040		
Podzol	18159	536.6		0.029		
Ranker	2531	82.6		0.033		
Regosol	437	19.0		0.044		

*Both studies calculated the soil C stocks excluding IC data therefore the stocks only represent the OC held within these stocks.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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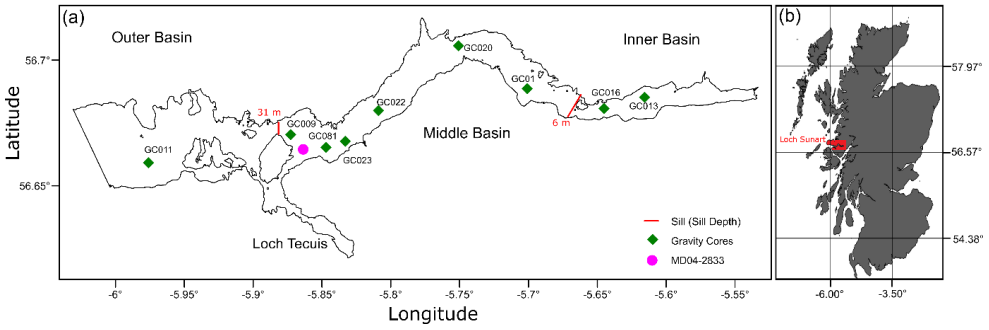

Figure 1. Maps of Loch Sunart illustrating (a) the three basins and the sediment core locations (b) Loch Sunart in a Scottish context.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
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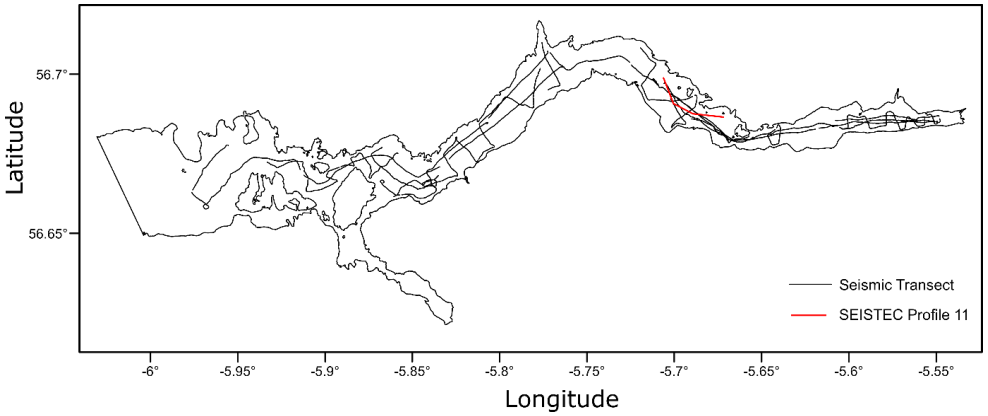

Figure.2. Map of the 34 Seismic transects undertaken in Loch Sunart with Siestec Profile 11 highlighted.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
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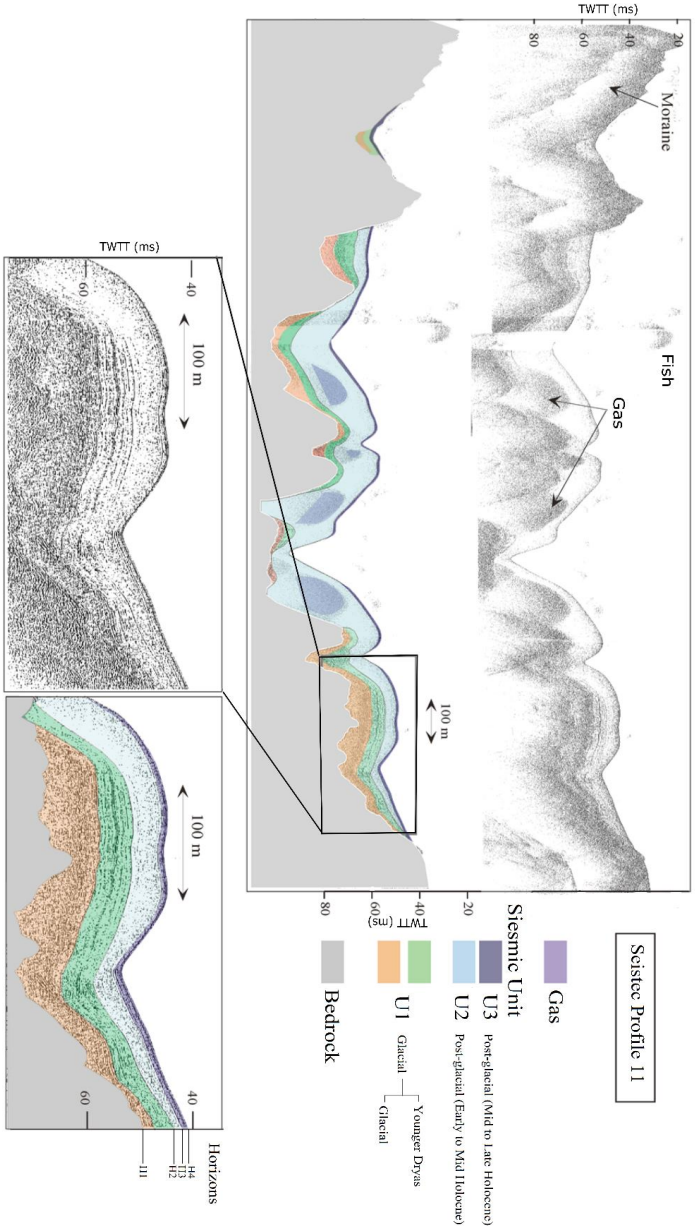

Figure 3. SIESTEC Profile 11: A characteristic seismic profile displaying the four seismic horizons (H1, H2, H3 and H4) and the three seismic units (U1, U2 and U3) adapted from Baltzer et al.,2010.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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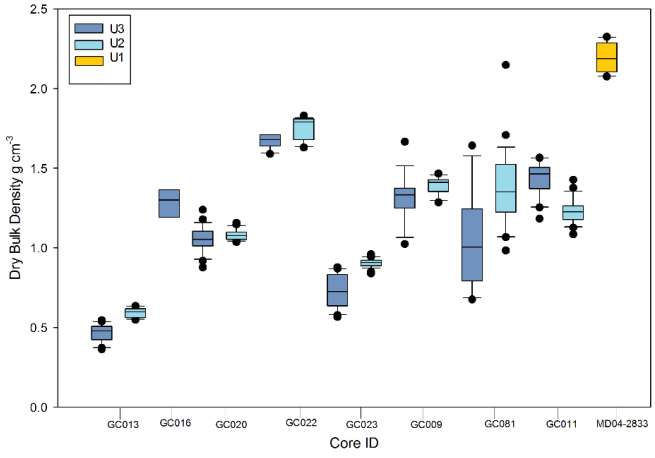

Figure 4. Dry bulk density values from each sediment cores corresponding to seismic units 1, 2 and 3.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
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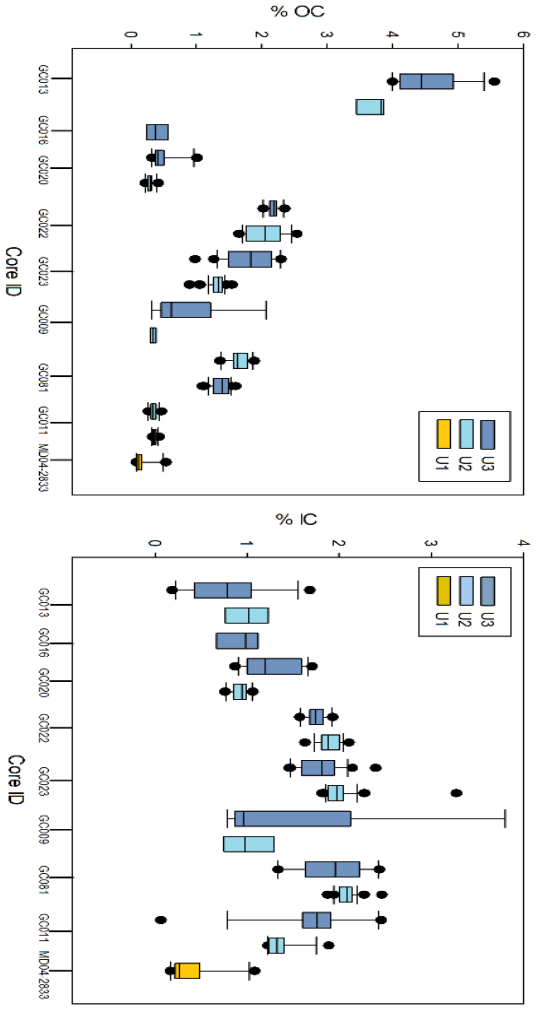

Figure 5. %OC and %IC values from each sediment cores corresponding to seismic units 1, 2 and 3.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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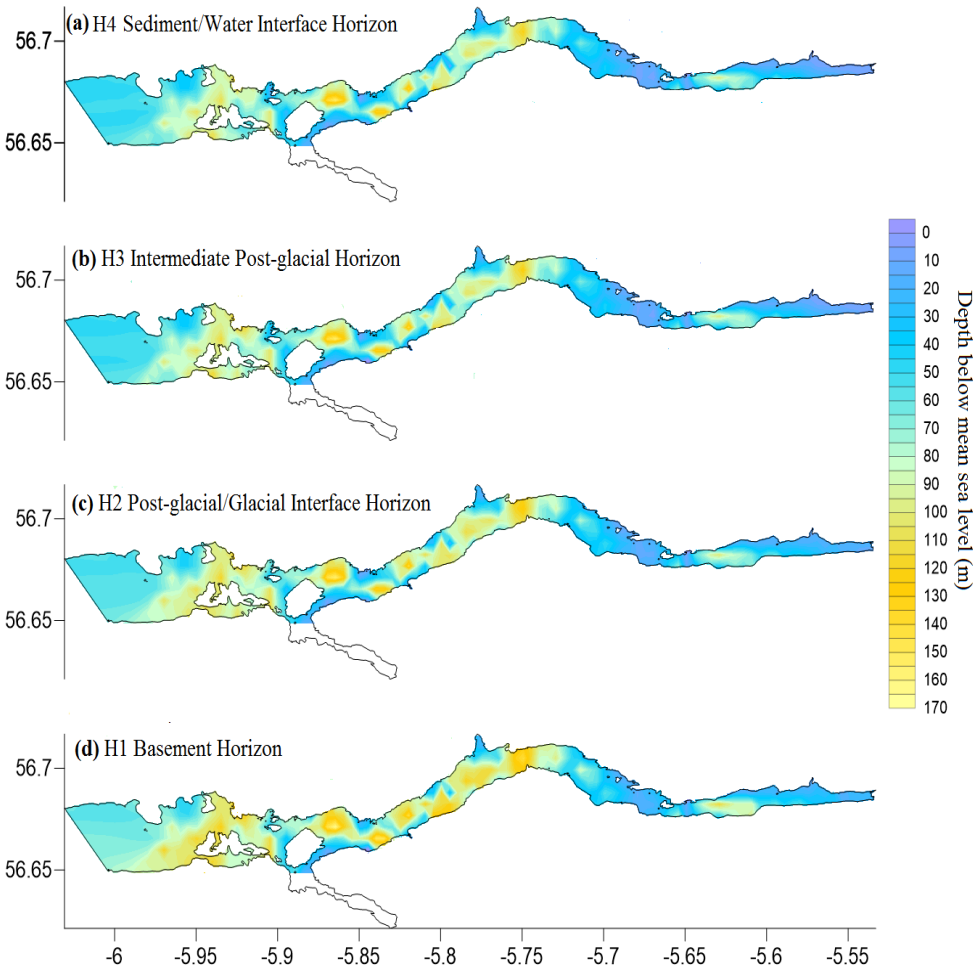

Figure 6. Contour maps defining the topography of each seismic horizon. **(a)** H4 sediment/water interface. **(b)** H3 intermediate post-glacial horizon. **(c)** H2 post-glacial/glacial interface. **(d)** H1 basement.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
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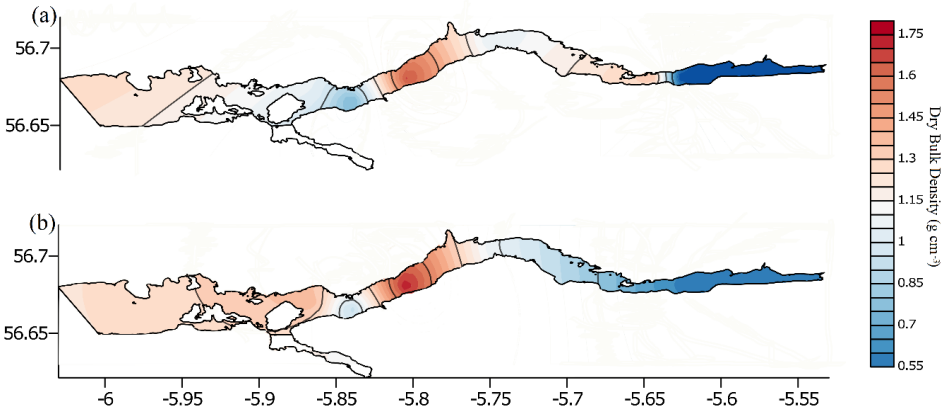

Figure 7. Contour maps showing the output of the spatial distribution model for the mean dry bulk density of **(a)** U3. **(b)** U2.

Biogeosciences Discuss., doi:10.5194/bg-2016-245, 2016
Manuscript under review for journal Biogeosciences
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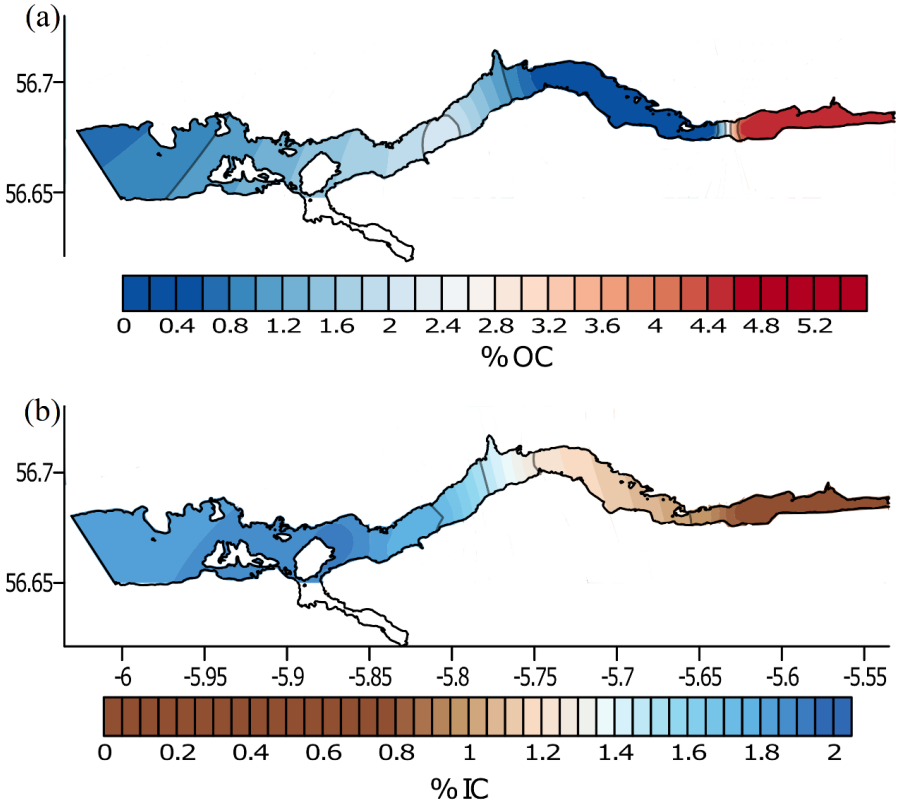

Figure 8. Output of U3 spatial distribution model for **(a)** Total carbon**. (b)** Organic carbon. **(c)** Inorganic carbon.

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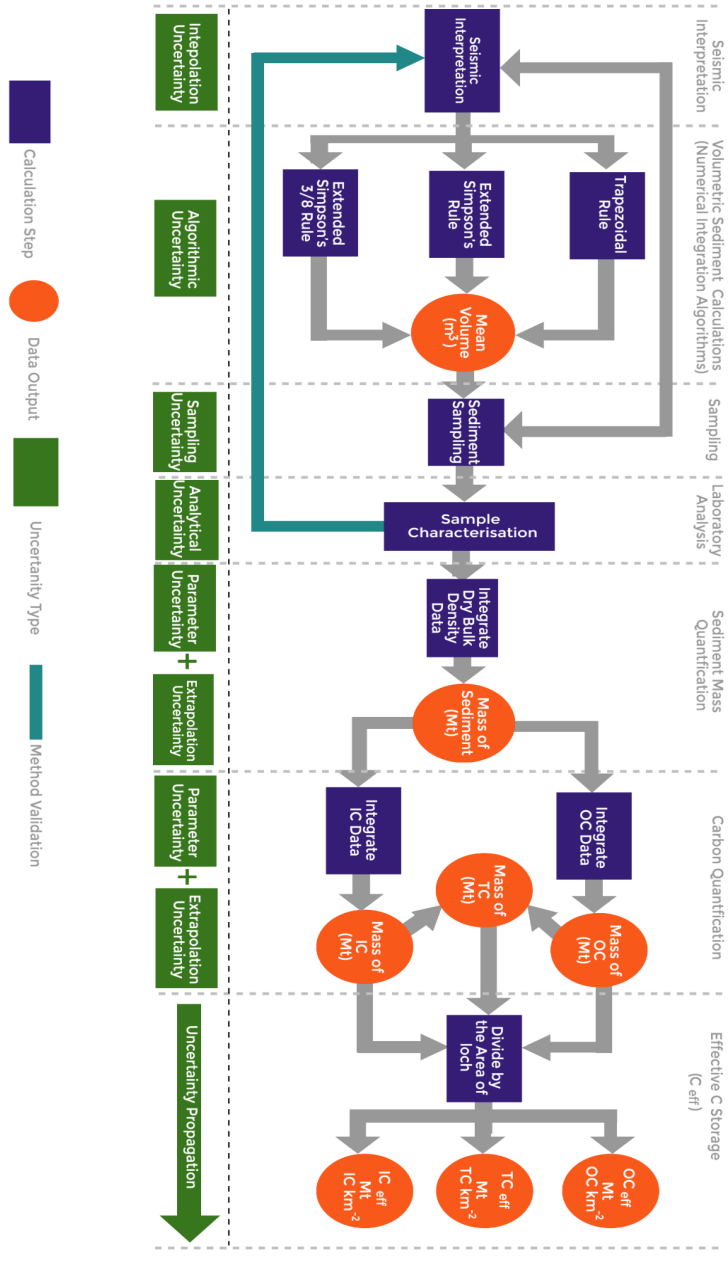

Figure. 9. Flow diagram detailing the steps towards calculating the sedimentary C stocks within a fjord with the known uncertainties specified.