Biogeosciences Discuss., https://doi.org/10.5194/bg-2019-163
Manuscript under review for journal Biogeosciences
Discussion started: 20 May 2019

The Holocene Evolution of a Sedimentary Carbon Store in a Mid Latitude Fjord.

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Key Points

1. Fjord sediments have been sustained hotspots for carbon burial during the Holocene
2. Climate change and humans have driven the evolution of fjord carbon stores
3. Terrestrial carbon in fjord sediments reaches up to 70% during the last millennium

Abstract

Fjord sediments are recognized as hotspots for the burial and storage of organic carbon, yet little is known about what drives the formation of these coastal carbon stores and how this has altered over time. Here we show that fjords can act as sustained hotspots for carbon burial and storage over Holocene timescales. Further we investigate the role of North Atlantic climate and humans in the evolution of a coastal carbon store using sediment records from a temperate Scottish fjord. Our findings indicate that climate and anthropogenic activity have independently driven increases in terrestrial carbon to the marine environment. When both these drivers were coupled, the terrestrial response was pronounced and the relative proportion of terrestrial OC in the marine sediments increases from 5% up to 70%. We hypothesize that sustained human disturbance through the late Holocene sensitized the catchment to abrupt climate reorganizations. The results highlight the importance of fjords for carbon burial and the significance of terrestrial carbon subsidy to the long-term carbon store.

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1. Introduction

Early work suggested that fjords are location of high sediment deposition and that despite their representation of a relatively small volume of the global continental margin (<0.1%), they contain ~ 12% of the sediments deposited over the past 100,000 yr (Syvitski et al. 1987). It was also shown that fjords contain some of the highest organic carbon (OC) found in coastal sediments (Skei, 1983), and that these systems may be global sink of OC - largely derived from terrestrially materials (Burrell, 1988). Later work suggested that temperate fjords may contain as much 12% of the global OC buried in margin sediments 100,000 yr (Newer and Keil, 2005), in support of the previously estimated high global sediment deposition in the margin (~ 12%) during this period (Syvitski et al. 1987). More recently, there has been a renewed interest in the role of fjords as global hotspots for long-term carbon (C) burial (Hinjosa et al. 2014; Smith et al. 2015; Cui et al. 2016a; Smeaton et al. 2016, 2017). To date, the mechanisms of enhanced carbon burial in these systems, which are likely related to high productivity (Simo-Matchim et al., 2016), rapid sedimentation (Syvitski et al., 1987), redox (Hinjosa et al. 2017), abundance of mineral surfaces (Keil and Mayer, 2014), and inputs of terrestrial C from litter to petrogenic materials) (Cui et al., 2016a, Smeaton and Austin, 2017) have yet to be fully explored.

In the North Atlantic region atmospheric forcing of Atlantic meridional overturning circulation (AMOC), which is reflected by the strength and position of the North Atlantic Current (NAC), coupled to persistent mode changes in the strength of the winter North Atlantic Oscillation (NAO) (Thornalley et al. 2009; Ortega et al. 2015), is a plausible underlying climate forcing mechanism that drives the sedimentary C composition of fjords during the post-deglacial Holocene epoch (<11.5 ky) in Europe. In fact throughout the Holocene, decadal and millennial-scale climatic variability has been well-documented in ice-core, marine sediment & terrestrial archives (Meeker and Mayewski, 2002). In particular, North Atlantic Holocene archives are amongst the most intensively studied records available, yet the climatic linkages between the ocean, atmosphere and terrestrial environment remain poorly understood on these timescales. Recently the impact of AMOC and NAO variability in the Holocene on terrestrial ecosystems sensitive to climate forcing (Seddon et al. 2016) has gained attention, the challenge lies in understanding the long-term interactions of climate and humans on C in the terrestrial environment and how this impacts the development of C stores at the land-ocean interface.

In the North Atlantic region previous attempts to separate the effects of climatic and human forcing on past alterations in terrestrial ecosystems have focused on the Baltic Sea (Zillén & Conley 2010); these authors showed that in the last two millennia hypoxic events mirrored the expansion and contraction of regional human population density, suggesting anthropogenic disturbance is a significant driver of coastal hypoxia through the introduction of terrestrial organic matter. While this work was successful in reconstructing the regional effects of humans and climate within the Baltic watershed and it's C cycle, an aquatic repository situated farther to the west, should provide a more suitable location to investigate the link between humans and North Atlantic climate forcing. Fjord systems store C effectively due in part to their deep glacial geomorphology, high sedimentation rates and often low oxygenated bottom waters; these features make these systems ideal for reconstructing regional climate change (Cage & Austin 2010; Faust et al. 2016) while also accumulating stores of C (Smeaton et al. 2017). Scottish fjords have also been shown to be ideally situated for coupling to North Atlantic climate forcing (Austin et al. 2006; Gillibrand et al. 2005) due to their proximity to the major North Atlantic currents and the westerlies (Fig.1). Additionally, there are long regional records of human occupation and environmental disturbance (e.g. Smout. 1993; Tipping. 2013).

Here, we present a sediment record from Loch Sunart, a fjord on the west coast of Scotland (Fig.1) in an attempt to unravel the roles played by North Atlantic climate and human disturbance play through the mid to late Holocene (7-0 ky) in the development of fjord sedimentary C stores and the wider significance of long-term carbon burial.

2. Study Site

Loch Sunart is a temperate non-glaciated fjord on the west coast of Scotland (Fig.1). The fjord is 30.7 km long and has an areal extent of 47.3 km^2 with a maximum depth of 145 m and consists an outer, middle and upper basin separated by shallow rock sills at depths of 31m and 6m respectively. Loch Sunart's catchment covers 299 km^2 with the main tributaries, the rivers Carnoch and Strontian entering close to the fjord head. The topographic nature of the catchment results in rivers having flashy flow regimes (Gillibrand et al., 2005).

The oceanographic conditions of Loch Sunart and most other Scottish fjords result in well ventilated bottom waters and generally they experience only minor seasonally hypoxic events (Gillibrand et al., 2005). Recent calculations estimate the post-glacial sediments of Loch Sunart hold 9.4 ± 0.2 Mt of OC (Smeaton et al., 2016), with an estimated 42.0 ± 10.1 % of the OC held within the surface sediments being terrestrial in origin (Smeaton and Austin, 2017).

Loch Sunart's catchment is dominated by shallow (mean depth: 50cm) C-rich, peaty gley soil and a land cover largely consisting of acid grasslands, commercial coniferous and deciduous woodlands (Smeaton and Austin, 2017). The physical characteristics of Loch Sunart and its catchment are largely representative of fjords across mainland Scotland (Smeaton et al. 2017); further Syvitski and Shaw's (1995) table of generalised fjord characteristics indicates that the fjords of mainland Scotland have comparable physical characteristics to the non-glaciated fjords found on the Norwegian mainland, Canada and in New Zealand.

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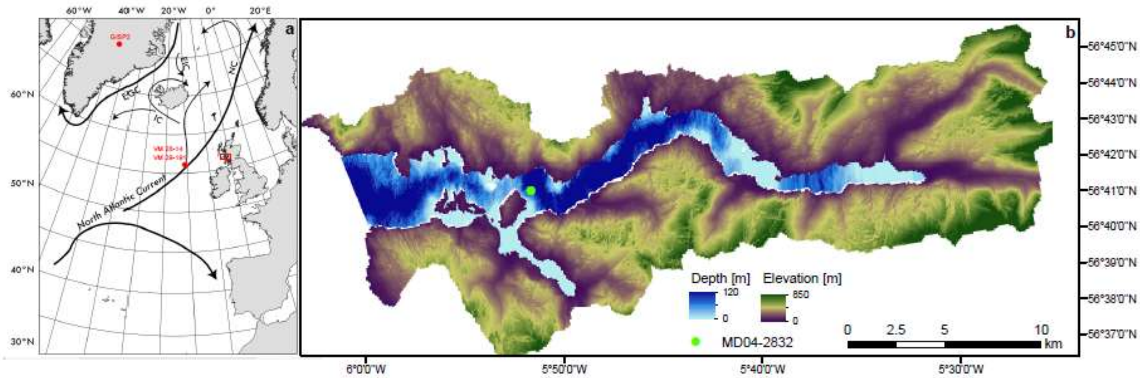

Figure.1 (**a**) North Atlantic Ocean circulation. Location of Loch Sunart (Red Box), GISP2 and cores VM 28-14 and VM29-191 (Bond et al. 1997) presented alongside the principal surface currents: **NAC**, North Atlantic Current; **IC**, Irminger Current; **NC**, Norwegian Current; **EGC**, East Greenland Current and **EIC**, East Iceland Current. (**b**) Location map detailing the catchment topography and seabed bathymetry with the sampling site of core MD04-2832.

Loch Sunart's catchment and the surrounding areas have a long record of human occupation (e.g. Smout. 1993; Tipping. 2013) extending back to the early Viking presence (~850 AD) and events which potentially caused regional environmental disturbance, such as the intensification of grazing (1350 AD), wide-spread woodland removal (1400-1600 AD), the introduction of lead mining (1722 AD) and start of industrial forestry (1919 AD). Key events in the catchment's history are summarized in Figure 2.

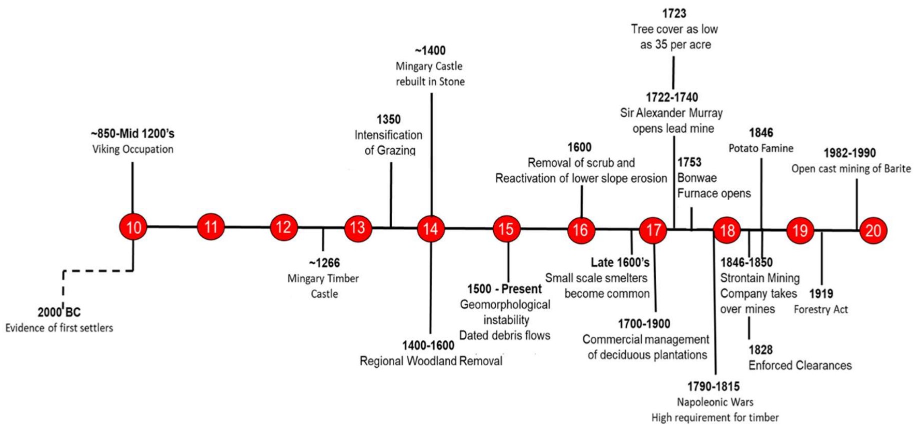

Figure 2. Timeline of key events pertaining to human occupation and disturbance in the Sunart region. Based upon Smout (1993) and Tipping (2013).

2.1 Regional Climate and Oceanography

The northward transport of warm, salty surface waters in the North Atlantic which is a fundamental element of the AMOC and one that asserts a fundamental control upon NW European climate. Flowing into Nordic Seas, the warm surface waters cool and sink promoting the formation of deep-water, which flows back into the Atlantic across the Greenland-Iceland-Scotland Ridge, becoming a major component of North Atlantic Deep Water (NADW). The surface NAC element of the AMOC draws waters from both the cold, fresh subpolar (SPG) and warm, saline subtropical (STG) gyres during its northward flow (Fig. 1). Relative contributions from these two gyres change through time and are primarily controlled by the dynamics of the SPG. Strong SPG circulation provides colder, fresher water to the NAC whilst weaker SPG circulation decreases its contribution to the NAC therefore making it warmer and saltier via enhanced STG contribution.

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3. Materials and Methods

3.1 Sampling

A 22.5 m giant piston core MD04-2832 (56.669833, -5.868667) was collected from the research vessel *Marion Dufresne* in the middle basin of Loch Sunart in 2004 (Fig.1). In addition to core MD04-2832, a 6m gravity core PM06-GC01 (56.670000, -5.871833) and a multi-core PMO6 MC01 (56.670000, -5.871667) were collected from the research vessel *Prince Madog* at same site in 2006. Core MD04-2832 is characterised by stiff silty mud with very little change throughout its 22.5m length, detailed sedimentological description and photographs can be found in the supplementary material.

3.2 Core Chronology

In-situ paired bivalve shells (*Corbula varicorbula and Nucula sulcate)* were collected from the length of the core for Accelerator Mass Spectrometry (AMS) dating. In total 22 samples underwent ^{14}C measurement from MD04-2832, PM06-GC01 and PM06-MC01C (Sup Table.1). Ages were calibrated using OxCal 4.2.4 (Bronk Ramsey and Lee, 2013) with the Marine13 curve (Reimer, 2013) and a regional correction of ΔR value of −26 ± 14 yr (Cage et al., 2006). Additionally, a ^{210}Pb chronology was developed for core PM06-MC01 (Sup Table.2). The ^{210}Pb dating was carried out in Copenhagen University following the Appleby and Oldfield (1992) and Appleby (2001) methodology. The ^{210}Pb and ^{14}C ages were combined to create a full chronology for core site MD04-2832. The methodology used magnetic susceptibility and geochemical (zinc) signatures to splice these core chronologies, this methodology is fully discussed in Cage and Austin (2010). To further test the splicing two calibrated ^{14}C ages were acquired from benthic foraminiferal (multi-species) samples from depths of 305cm and 440cm which agree well with adjacent mollusc shell dates (Sup Table 1), suggesting that the reworking of older sediment and foraminifera is not a significant problem.

An age model was created using the BACON software (Blaauw and Christensen, 2011) representing the 8000 year record (Sup Fig.3). The calibrated model for site MD04-2832 shows that the last 1000 years is contained within the uppermost 280 cm of the sediment record, which has a resolution of ~1 yr cm^{-1} for the top 50 cm and 4–6 yr cm^{-1} for the remainder of the core.

3.3 Geochemical Analysis

Elemental (OC, N) and isotope analyses ($\delta^{13}C_{org}$) of the sediments were carried out. Briefly, the samples were dried at 60°C for 24 hours, milled to a powder, with ~12 mg placed into both tin and sliver capsules. The tin capsules were analysed to determine N concentration while the silver capsules undergo acid fumigation (Harris et al., 2001) to remove carbonate. Acid fumigation involves placing the silver capsules in a desiccator with a beaker of 12 M HCl for 8hrs to remove carbonate and prevent the loss of water soluble C. Prior to analysis these samples were dried at 60°C for 24 hours. Measurements were made using an elemental analyser interfaced with an isotope ratio mass spectrometer (IRMS). C and N isotope ratios were calculated in δ notation relative to the Vienna Pee Dee Belemnite (VPDB) and Air standards respectively.

3.4 Physical Properties Analysis

Water content, wet and dry bulk density and porosity of the sediment were measured at 10 cm intervals following standard methods (Dadey et al., 1992; Danielson, 1986). Magnetic susceptibility (SI units) measurements were taken at 2 cm resolution. Particle size analysis (PSA) was undertaken, where the organic and carbonate fractions are removed by Hydrogen Peroxide (H_2O_2) and Hydrochloric Acid (HCl) treatments, respectively. The treated samples were then analyzed by laser granulometry using a Beckman Coulter LS230 to measure the particle size (<2 mm) of the reaming mineralogical fraction.

Sedimentation rates (cm yr^{-1}) were calculated using the output from the Bayesian age-depth model. OC accumulation rates (OCAR) where calculated using the approach outlined in Smith et al., (2015). Briefly, the average bulk density, porosity and % OC was calculated allowing the OCAR to be calculated as follows:

$$OCAR = \%OC \text{ x sedimentation rate x } (1 - \text{porosity}) \text{ x bulk density}$$

3.5 Biomarkers

Analysis of alkanes and fatty acids was based on a modified method of Cui et al. (2016b). Briefly, ~1 g samples were extracted on accelerated solvent extractor (ASE) using dichloromethane (DCM): methanol (MeOH) (9:1 v:v). After being saponified with KOH in MeOH, "neutral" and "acid" fractions were sequentially extracted with hexane and hexane:DCM (4:1 v:v). The former fraction containing alkanes were analysed on the gas chromatographer – flame ionization detector (GC-FID) for alkane concentrations. The latter fraction containing fatty acids (FA) were then derivatized using boron trifloride (BF_3) in MeOH, re-extracted using DCM, and eluted using DCM on a Pasteur pipette column. Fatty acid methyl ester (FAME) samples were analysed on the same GC-FID as above.

The concentrations of alkanes and fatty acids were calculated and corrected with internal standards (C_{34} alkane isomer, C_{19} FA) and mix standards of alkanes and FAMEs. ALK $_{C25-35}$ is calculated as the sum of the odd chain C_{25} to C_{35} alkanes, while ALK C_{24-36} is the sum of even chain C_{24} to C_{36} alkanes. ALK P_{aq} is the ratio of C_{23} and C_{25} alkanes over the sum of C_{23}, C_{25}, C_{29}, C_{31} alkanes. Short-chain fatty acids (SCFA) is calculated as the sum of C_{12} to C_{18} fatty acids, while long-chain fatty acids (LCFA) is calculated as the sum of C_{24} to C_{32} fatty acids. Terrestrial to aquatic ratios of fatty acids (TARFA) is the ratio of C_{24}, C_{26} and C_{28} fatty acids over the sum of C_{12}, C_{14}, C_{16}, C_{24}, C_{26}, C_{28}. Finally, the ratio of fatty acids to alkanes (FA/ALK) is the ratio of C_{24-32} fatty acids to C_{24-36} alkanes.

Analysis of glycerol dialkyl glycerol tetraethers (GDGTs) was based on the method of Liu et al. (2016) and Smith et al. (2010). Shortly, ~1 g of sediment samples were sonicated and extracted using DCM: MeOH (9:1 v:v) using an ultra-sonicator. The extracts were re-concentrated in hexane and analysed on a liquid chromatographer – mass spectrometer (LC–MS). Quantification of GDGTs was achieved by using a synthesized tetraether surrogate standard and focusing on targeted ions (e.g., m/z 1292) on the LC–MS. Branched/isoprenoid tetraether (BIT) index is calculated as the ratio of three branched GDGTs (I, II, and III) to the sum of branched and crenarchaeol GDGTs. The targeted m/z of the four compounds are 1022, 1036, 1050, and 1292 for branched I, II, III and crenarchaeol GDGTs.

3.6 Paleoclimate and Paleo-Environmental Reconstruction

Stable oxygen ($\delta^{18}O$) isotopes were measured on between 20-30 hand-picked individuals of the infaunal benthic foraminifera species *Ammonia beccarii*. Samples were cleaned in ethanol, dried at 40°C and run on a Gas-bench coupled to an IRMS. Stable isotope compositions are reported relative to the Vienna Pee Dee Belemite (VPDB) using the NBS-19 standard with precision of 0.07‰ based on an internal Carrara marble standard.

We estimate past regional vegetation composition using the regional estimates of vegetation abundance from large sites (REVEALS) modelling approach (Sugita, 2007). REVEALS is a Landscape Reconstruction Algorithm (LRA) (Sugita, 2007) which transforms pollen proportions into estimates of vegetation cover. REVEALSinR implements the REVEALS model (Theuerkauf et al., 2016) and integrates Lagrangian stochastic dispersal modelling (Kuparinen et al., 2007). Data for Gallanech Beg were taken from the European Pollen Database, the site was chosen due to its proximity to our study site and strong chronological constraint.

To estimate the proportion of terrestrial OC in the sediments a mixing model approach was utilised. The approach used $\delta^{13}C_{org}$, $\delta^{15}N$, C/N ratios and BIT index as tracers in conjunction with a Bayesian mixing model. The methodological approach used by Smeaton and Austin (2017) was utilised alongside the OC source characteristics, which are specific to Loch Sunart (Sup Table 3). This approach does not completely overcome the problems associated with post-depositional alteration of the organic matter, but the use of four tracers, site specific source data and a Bayesian approach gives us confidence in the estimates and associated errors to be largely representative of the sedimentary environment.

4. Regional Climate Reconstruction

Loch Sunart is separated from the coastal ocean by a sill at a water depth of 33 m, which allows for direct sub-surface exchange and communication of shelf salinity and temperature into the main basin (Gillibrand et al., 2005). The middle basin core site (MD04-2832) is therefore directly influenced by the hydrography of the Scottish Shelf, where northward flowing Scottish Coastal Current (SCC) waters are influenced by the intrusion of North Atlantic water onto the shelf (Inall et al. 2009). The $\delta^{18}O$ composition of benthic foraminifera is a measurement of the interplay between the salinity and temperature of seawater (Cage and Austin, 2010). The Holocene $\delta^{18}O$ values of benthic foraminifera at core site MD04-2832 exhibit pronounced millennial-scale variability of up to 1 ‰ around a long-term average of 1.65 ‰. Values range from greater than 2 ‰ to less than 1 ‰, indicating changes from colder and/or more saline conditions to warmer and/or fresher conditions, respectively (Fig.3). Based on the palaeo-temperature equation O’Neill (1969) and assuming no change in salinity, the observed Holocene shifts in $\delta^{18}O$ equate to temperature changes of approximately 4ºC. In contrast, assuming no change in temperature, the observed Holocene 1 ‰ shifts in $\delta^{18}O$ equate to salinity changes of ~5.5 practical salinity units (psu). Such large and sustained changes in main basin salinity are unlikely, given that coastal salinity changes are small (<0.5) over the instrumental period and that modelling studies (assuming constant coastal salinity boundary conditions) show main basin salinity changed by less than 0.02 during recent extreme NAO years (Gillibrand et al., 2005).

The most pronounced feature of the Loch Sunart climate record occurs shortly after 6000 cal. BP, when $\delta^{18}O$ values reach their most depleted (0.9 ‰) and then increase abruptly shortly before 5000 cal. BP (Fig 3). The timing of this event is broadly coincident with the decreasing relative NADW contribution inferred from benthic foraminiferal $\delta^{13}C$ from Ocean Drilling Project Site 980, located at a water depth of 2.1 km on the Feni Drift (Fig.1), which began at ~6.5 cal. BP (Oppo et al. 2003). This event is also broadly time equivalent to reconstructions of warmer and more saline surface water inflows to the sub-polar northeast North Atlantic, suggesting that as NADW formation and the strength of the SPG weakened the STG contribution to the NAC (Thornalley et al. 2009). At the same time, chemical records from the Greenland Ice Sheet Project 2 (GISP2) ice core suggest major atmospheric reorganization (Meeker and Mayewski, 2002), with the expansion of the polar vortex and a shift towards winter-like conditions at high latitudes (Fig.3).

This coincides with an increase in the relative abundance of haematite-stained grains in northeast North Atlantic cores during this 5-6 kyr event (Bond et al. 2001) which suggests that cold, fresh, ice-bearing surfaces waters expanded southwards from North of Iceland and were able to influence the strength of SPG circulation. The dynamics of the SPG are therefore critical to AMOC variability and the Loch Sunart $\delta^{18}O$ data suggest a progressive strengthening (warming) of the STG contribution to the NAC in the build-up to the 5-6 cal. BP event (Thornalley et al. 2009).

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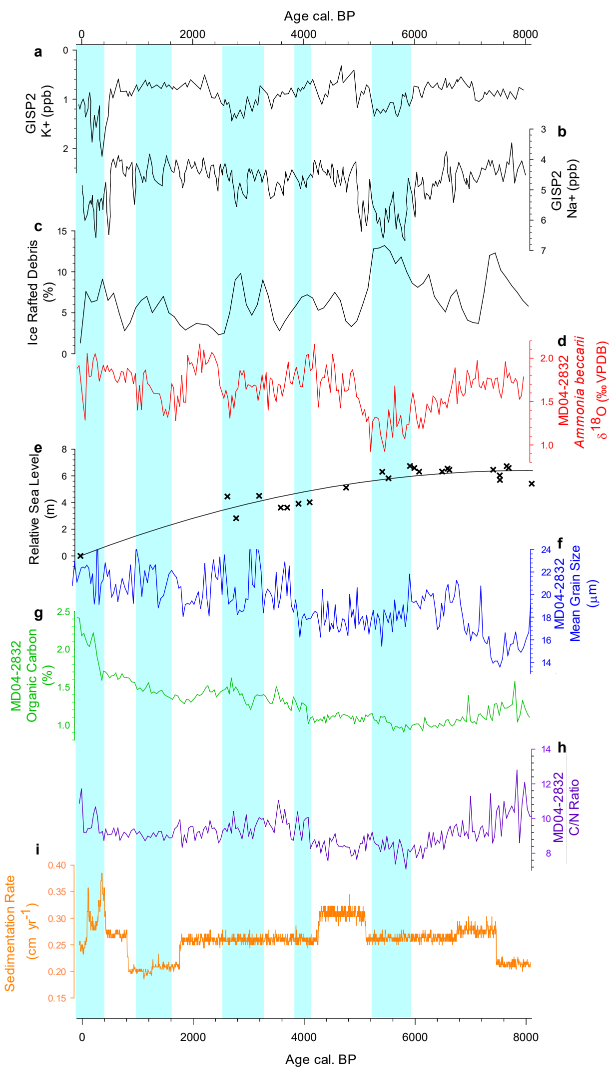

Figure 3. Age calibrated MD04-2832 record in the context of North Atlantic Holocene paleoclimate records (a) Gaussian smoothed (200 yr) GISP2 sodium (Na+; parts per billion, ppb) ion proxy for the Icelandic Low (b) Gaussian smoothed (200 yr) GISP2 potassium (K+ ; ppb) ion proxy for the Siberian High (Mayewski et al., 2004; Meeker and Mayewski, 2002). (c) Pervasive millennial-scale cycle illustrated by ice-rafted debris (%) from cores VM-28-14 and VM-29-191(Bond et al., 2001). (d) MD04-2832 foraminifera (*Ammonia beccarii*) $\delta^{18}O$ (‰

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VPDB) record as a proxy for basin temperature/salinity. (**e**) Regional relative sea level (Shennan et al., 2005) (**f**) MD04-2832 mean grain size (after removal of organic and carbonate components) (**g**) MD04-2832 organic carbon (%) (**h**) MD04-2832 C/N ratio. (**i**) Sedimentation rate (cm yr^{-1}) calculated from the Bayesian age model (Sup Fig.3). Vertical shading represent the Rapid Climate Change timing tuned to the GISP2 chronology (Mayewski et al., 2004)

5. Long-term Evolution of a Sedimentary C Store

5.1 Early to Mid-Holocene

During the early to mid-Holocene the accumulation of sediments and the development of the sedimentary C store is partly governed by ongoing post-glacial Relative Sea Level (RSL) change. The relatively steady, long-term fall in Holocene RSL (Fig. 3) since 7500 cal. BP (Shennan et al. 2005) reduced sill depth from ~41 m (early to mid-Holocene) to 33 m (present) and modified sub-surface exchange and bottom currents with the coastal waters. The increased openness of the fjord in the early Holocene allowed greater exchange between the coastal and fjord waters, resulting in a greater proportion of marine derived OC being buried, as illustrated by the lower C/N ratios found during the early to mid-Holocene (Fig.3). Falling RSL over the entire Holocene may have gradually restricted the input of marine derived OC and allowed for the relative increase in the proportion of terrestrial OC; changes in C/N ratio and the coarsening of mean grain size throughout the Holocene may well reflect these RSL drivers (Fig.3).

Climate reorganizations, such as those recorded in the sedimentary record of Loch Sunart, are likely to have triggered a terrestrial ecosystem response (Frank et al., 2015), in turn increasing the proportion of terrestrial OC input to the fjord. During the early to mid-Holocene the C/N ratio remains relatively stable which indicates little or no change in the sources of OC, suggesting that there was no or only a minor terrestrial ecosystem response to the major climate reorganizations. Regional landscape vegetation reconstructions (Fig.4) further support this conclusion - during this period the woodland coverage remained stable and did not respond to millennial-scale climate reorganizations, as shown by previous pollen analysis from the neighbouring Loch Etive (Cundill and Austin 2010).

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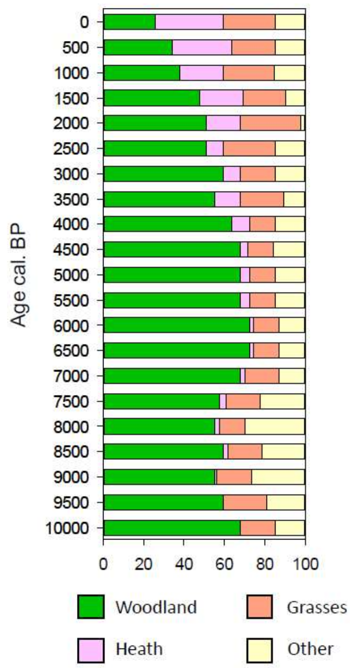

Figure 4. Estimated Holocene regional vegetation cover types for Gallanech Beg (Davies, 2009) a bog 40 km South of Loch Sunart derived from the REVEALS modelling. Pollen counts available from the European Pollen Database (www.europeanpollendatabase.net).

5.2 Mid- to Late Holocene

The west coast of Scotland has been occupied since the Mesolithic period (Bishop et al. 2015) and it is believed that the region surrounding Loch Sunart was permanently settled at approximately 4000 cal. BP (Tipping 2013; Bishop et al. 2015). The earliest anthropogenic alteration to leave a lasting legacy on the environment was the removal of woodlands (Smout 1993). The Gallanech Beg record (Fig.4) shows a decline in woodland cover from 4500 cal. BP, corresponding to the start of a long-term gradual increase in OC content (weight %) in the fjord sediments (Fig. 3). The removal of these woodlands initiated landscape reorganization of vegetation with the appearance of more pioneering plant species and grasslands, which initially dominated the deforested landscape before being replaced by lowland heath plants (Fig. 4). This succession of vegetation is common throughout Scotland and the North Atlantic region (Fyfe et al. 2013). Heath plants, such as heather, have the ability to stabilize soils (Panagos et al. 2015), but are vulnerable to grazers (i.e. sheep and deer) and less adaptable to climate alterations compared to the earlier pioneering plant species (Hartley & Mitchell 2005). The mid- to Late Holocene is characterized by slightly coarser sediment and a gradual increase in C/N ratios, which coincide with periods of rapid North Atlantic climate oscillations (Fig. 3). We hypothesis that the gradual underlying change in vegetation cover superimposed by these rapid shift in the North Atlantic climate may have facilitated the erosion of terrestrial C and its transfer to the marine environment.

The mid-Holocene, from around 5000 to 2000 cal. BP, is characterized by largely stable OC accumulation rates (OCAR) (Fig 5a). To maintain this stability in OCAR, we speculate (but cannot prove) that a decrease in marine OC input due to falling sea level may have been balanced by an increase in terrestrial OC input due to natural and anthropogenically induced vegetation change (Fig.4). Previous estimates from Loch Sunart of Holocene OCAR ranged between 1.89 and 25.68 g C m^{-2} yr^{-1} (Smeaton et al., 2016) with the upper range of these estimates originating from the vicinity of core MD04-2832. In comparison to the published values, the OCAR of the early Holocene (8-6 kyrs BP) are significantly higher (38 ± 6 g C m^{-2} yr^{-1}), but still fall within the range of OCAR observed in non-glaciated North Atlantic fjords (Smeaton et al., 2016). By the mid to late Holocene (6-1 cal. kyrs BP) the OCAR in MD04-2832 decreased to 26 ± 3 g C m^{-2} yr^{-1} which is equivalent to the rates observed in previous studies at this location within the fjord (Smeaton et al. 2016). This millennial-scale record of OCAR suggests that Loch Sunart has been a hotspot for the burial and storage of C throughout

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the Holocene, highlighting the long-term significance of fjords as globally important environments for C burial (Smith et al. 2015).

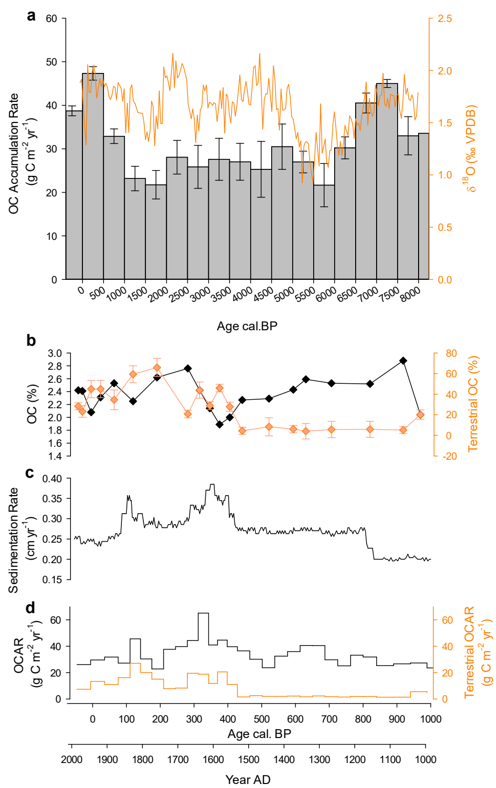

Figure 5. (**a**) Holocene OC accumulation rates with standard deviation for site MD04-2832 overlain by the foraminifera $\delta^{18}O$ record and records for the last millennium (**b**) % OC and % terrestrial OC with uncertainties (**c**) Sedimentation rate and (d) OCAR and terrestrial OCAR.

5.3 The Last Millennium

The last millennium witnessed a 48 % rise in OCAR above the mid-Holocene average of the preceding 6500 years; these late Holocene OCARs are similar to those observed between 7500-6500 cal. BP. During the last millennium the concentration of marine OC has remained stable, as indicated by constant crenarchaeol concentrations (Sup Fig. 5) suggesting that the increase in OCAR in Loch Sunart's sediment are primarily driven by changes in terrestrial inputs (Fig. 5d). The observed increases in terrestrial OC (Fig. 5b) do not directly correspond to any major change in climate (Fig.3) and RSL change had slowed significantly by this time (Shennan et al., 2005). Therefore the increase in terrestrial OC seems to be de-coupled from either driver (i.e. climate, RSL), suggesting another mechanism had become dominant during this period of time. The last millennium witnessed an unprecedented anthropogenic pressure on the catchment to provide resources for a growing local and national population in Scotland (Sup Fig. 6). Our sediment records show that anthropogenic disturbances to the catchment were initiated at approximately 1520 ± 63 AD, when there is a significant increase in terrestrial input (Fig. 6). Between 1000 – 1520AD, terrestrial OC rose from less than 5% to a maximum of 70% by 1520AD. During this time period (16th Century) scrub vegetation was being removed from the landscape to improve grazing and to supply local charcoal production (Tipping 2013), which in turn reactivated lower slope erosion (Brazier and Ballantyne 1989), the mobilization and transport of soil materials to the loch. It should also be noted that earlier slope instability during the 15th Century and recorded nearby was also likely due to phased woodland and scrub removal (Ballantyne 1991).

In Loch Sunart, the initial phase of the 16th Century disturbance resulted in a pulse of coarse grained mineralogical material being delivered to the sediments, most likely due to the erosion of deep soils (Brazier & Ballantyne 1989; Ballantyne 1991); an observed increase in grain size and magnetic susceptibility is noted at this time (Fig. 6). This high mineralogical input resulted in a dilution effect, lowering the OC in the fjord sediment. The initial pulse was followed by an increased input of terrestrial OC (1584 ± 58 AD), as evidenced by a decrease in $\delta^{13}C_{org}$ from -18.5 ‰ to -25.0 ‰ and an increase in C/N from 10.5 to a peak of 16.8 (Fig.6). This pattern is comparable to a similar record in Loch Etive (Nørgaard-Pedersen et al. 2006), where in the last 1000 years there is a marked increase in OC coupled to an increase in magnetic susceptibility which is indicative of higher mineralogical input suggesting a terrestrial source. This shift towards greater terrestrial input (Fig. 5b) is further supported by the biomarker profiles which all indicate an increase in terrestrial OC input to the sediments (Fig.6), these complementary records are presumably reflecting regional terrestrial responses across NW Scotland.

A decline in terrestrial C inputs to Loch Sunart sediments in the early to mid-1800's suggests the fjord system is potentially returning to pre-1520 conditions based on chemical biomarkers and bulk OC proxies. This change could be due to the exhaustion of erodible soil materials, or more likely that the depopulation of the catchment during the 19th century allowed the recovery of vegetation and stabilization of the soils within the catchment. More recent disturbance of the catchment has also impacted the quantity of C held within fjord sediments. In particular, widespread planting of coniferous woodland during the 1950's is associated with increased terrestrial inputs from this time onward (Fig. 6). A pulse of slightly coarser-grained material diluted the bulk OC concentration in the sediments and was followed rapidly (1964 ± 8 AD) by a marked increase in terrestrial OC. Interestingly, this initial response of OC dilution, via coarse lithic material input from eroding soils, is similar to that observed during the 1520 event.

The pressure humans have exerted and the associated disturbance of vegetation and soils within the catchment from the late 18th Century to the present day through Lead, Zinc and Copper mining (Sup Fig.7) alongside commercial forestry is several orders of magnitude more intense than prior to the 1520 AD event. For example, the concentrations of these metals increases dramatically from around the mid-1700's, intensifying in the 1900's and corresponds to the written records for mining activity in and around Stontian, within the Loch Sunart catchment (Smout. 1993; Tipping. 2013). Yet the terrestrial response to these later catchment alterations are muted in comparison (Fig. 5). It is therefore unlikely that human activity was the sole driver of this increased terrestrial C storage within the sediments during the 16th Century. Here, we hypothesis that abrupt climatic change may have been the contributing factor responsible for this heightened terrestrial response. For example, at approximately 1525 AD (within chronological uncertainty of 1520 ± 63AD) there was a rapid reorganization in the NAO recorded in the sediments of Trondheimsfjord, Norway (Faust et al. 2016), where the NAO switched to a positive phase after a sustained period (~315 years) in its negative phase. This positive switch was short (10-15 years) but would have created a wetter atmosphere over NW Europe. Moreover, peatbog water table (Charman et al. 2006; Langdon et al. 2003) and tree ring temperature reconstructions (Rydval et al. 2017) from Scotland confirm this widespread atmospheric reorganization and corroborate a transition to a wetter environment. Loch Sunart has been shown to be sensitive to NAO-forcing (Gillibrand et al. 2005), whereby the switch in the phase of NAO recorded in the $\delta^{18}O$ record may also have driven increased runoff, and increased C loss through soil erosion. This link between regional climates, oceanography and $\delta^{18}O$ was outlined in Scottish sea lochs by Cage & Austin (2010) in their interpretation of a millennial-scale record from Loch Sunart. In particular, during the dry negative NAO phases of the Holocene the catchment would build and store soil materials, which could then quickly be lost during the shift to a positive, wetter phase of the NAO. There have, of course, been similar changes to climate and phase of the NAO throughout the Holocene (Fig.3), but not with the same underlying destabilizing effects of humans on the landscape. The major reorganization in the mode of the NAO over the North Atlantic during the late Holocene may

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Manuscript under review for journal Biogeosciences
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have triggered this enhanced terrestrial response. We hypothesize that the long-term modification of the terrestrial environment by humans sensitized the catchment to abrupt climatic reorganization. The shift in the NAO with the associated anthropogenic destabilization resulted in a more vulnerable terrestrial ecosystem, allowing for the mobilization and transfer of terrestrial OC to the fjord sediments (Fig.6).

The last millennium has seen two significant increases in sedimentation rate (SR) (Fig. 5c) associated with the 16th century climate event and the post-industrial era (~1850 AD) disturbances of the catchment. Both increases in SR are mirrored by an increase in OCAR; these increases can be large and rapid but they also return to the Holocene background rates within 20-30 years of each major event. This demonstrates that fjords not only record changes in climate and the catchment, but that they also have the capacity to capture and bury OC at greater rate than the long-term Holocene norm (Smeaton et al. 2016). This adaptability of the coastal ocean C, and fjords in particular, in trapping terrestrial OC released by climatic and anthropogenic activities may represent an unrealized yet significant long-term buffer in the global carbon cycle.

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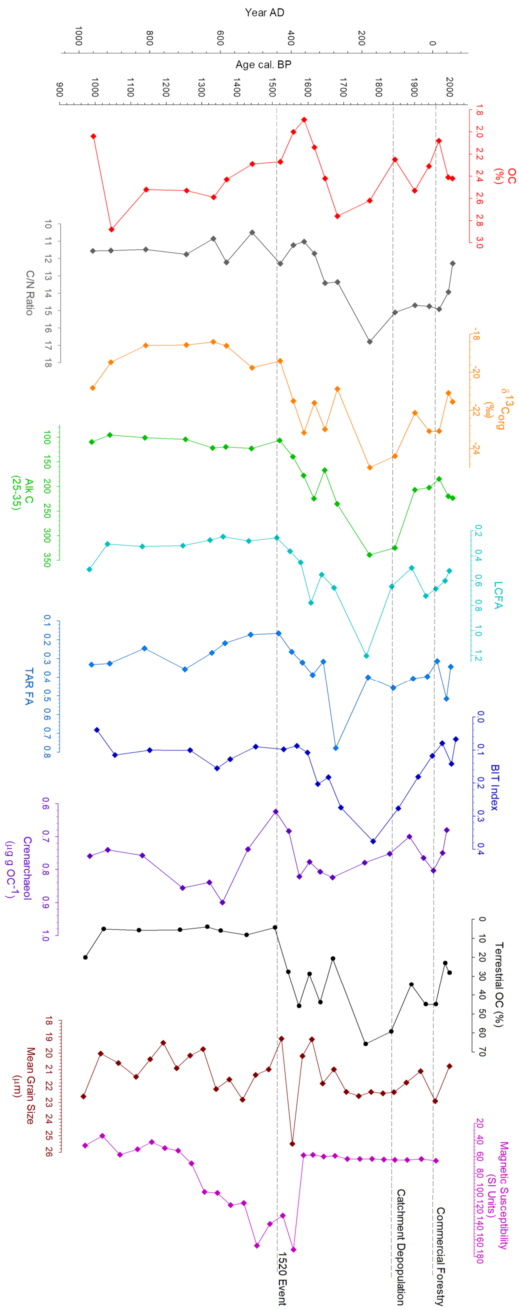

Figure 6. Bulk elemental, biomarker and physical property profiles of core MD04-2832 spanning the last 1000 years. Further biomarker and isotope profiles can be found in the supplementary material.

6. Conclusion

While fjords are known hotspots for C burial (Smith et al. 2015) and storage (Smeaton et al. 2016, Smeaton et al., 2017) the effectiveness of these environments as OC sinks over Holocene timescales is now evident from this study. It is clear that both climate and humans are important drivers in the development of such coastal C stores, with the results indicating that both climate change and human activity independently drove changes in the terrestrial environment and transport of OC to the coastal ocean. When climatic and human forcing is coupled, the terrestrial response is heightened. The long-term importance of fjords as hotspots for carbon burial and the potential role of terrestrial carbon subsidy to the coastal ocean as an overlooked component in the global carbon cycle is evident. The capacity of fjords to capture significant quantities of OC and to effectively store both terrestrial and marine C are significantly overlooked services within the earth system.

Acknowledgments

This work was finically supported by the Natural Environment Research Council (grant number: NE/L501852/1), the EU FPV HOLSMEER project (EVK2-CT-2000-00060) and the EU FPVI Millennium project (contract number 017008), Biotechnology and Biological Sciences Research Council (grant number: BB/M026620/1) with additional support from the NERC Radiocarbon Facility (Allocation 1154.1005). We acknowledge Jon L. and Beverly A. Thompson Endowed Chair of Geological Sciences held by T.S. Bianchi in the Dept. of Geological Sciences at University of Florida, and China Scholarship Council for supporting part of this research. Further we thank both the SAGES (Scottish Alliance for Geoscience, Environment, Society) and MASTS (The Marine Alliance for Science and Technology for Scotland) pooling initiatives in funding this collaborative research. The CALYPSO long core was acquired by W. Austin within the frame of the French ECLIPSE programme with additional financial support from NERC, SAMS and the University of St Andrews. W. Austin and J. Howe would like to thank Marion Dufresne's Captain J.M. Lefevre, the Chief Operator Y. Balut (from IPEV). Lastly, we would like to thank Charlie Wilson (Scottish Association of Marine Science) and Chris Wurster (University of St-Andrews) for laboratory support.

Author Contribution

C.S and W.E.N.A conceived the research and wrote the manuscript in collaboration with T.S.B to which all co-authors contributed data and provided input. The analytical work was undertaken by C.S, X.C and A.G.C under the supervision of W.E.N.A, J.A.H and T.S.B.

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