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Organic carbon rich sediments: benthic foraminifera as bio-indicators of depositional environments

Elena Lo Giudice Cappelli[1], Jessica L. Clarke[1,2], Craig Smeaton[1], Keith Davidson[3], William E. N. Austin[1,3]

[1]School of Geography and Sustainable Development, University of St Andrews, St Andrews, KY16 9AL, Scotland, UK
[2]School of Natural and Environmental Sciences, Newcastle University, Newcastle, NE1 7RU, UK
[3]Scottish Association for Marine Science, Scottish Marine Institute, Oban, PA37 1QA, Scotland, UK

Correspondence to: Elena Lo Giudice Cappelli (elena.logiudice.cappelli@gmail.com)

Abstract. Fjords have been described as hotspots for carbon burial, potentially playing a key role within the carbon cycle as climate regulators over multiple timescales. Nevertheless, little is known about the long-term fate of the carbon that may become stored in fjordic sediments. One of the main reasons for this knowledge gap is that carbon arriving on the seafloor is prone to post-depositional degradation, posing a great challenge when trying to discriminate between an actual change in carbon deposition rate and post depositional carbon loss. In this study, we evaluate the use of modern benthic foraminifera as bio-indicators of carbon content in six voes (fjords) on the west coast of Shetland. Benthic foraminifera are sensitive to changes in carbon content in the sediments, and changes in their assemblage composition therefore reflect synchronous variations in the quantity and quality of carbon reaching the seafloor. We identified four environments based on the relationship between benthic foraminiferal assemblages and carbon content in the sediments: 1) Land-locked regions influenced by riverine/freshwater inputs of organic matter, namely the head of fjords with a restricted geomorphology; 2) Stressed environments with a heavily stratified water column and sediments rich in organic matter of low nutritional value; 3) Depositional environments with moderate organic content and mild or episodic current activity; 4) Marginal to coastal settings with low organic content, such as fjords with an unrestricted geomorphology. We conclude that foraminifera potentially provide a tool to disentangle primary carbon signals from post-depositional degradation and loss of carbon because of their environmental sensitivity and high preservation potential in the sedimentary record.

1 Introduction

Globally, fjords sequester about 18 Mt of organic carbon (OC) annually in their sediments, partly acting to reduce the remineralisation of OC into the environment and thus buffering the release of carbon dioxide (CO_2) to the atmosphere (e.g. Smith et al. 2015; Cui et al., 2016). A recent study by Smeaton et al. (2017) estimated that the sediments of Scotland's 111

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major fjords hold 295 ± 52 Mt OC. In Shetland, these fjordic systems are different from their mainland counterparts, notably in that they are shallower and less glaciated (Edwards and Sharples, 1986) and peat or peaty soils dominate their catchments (Soil Survey of Scotland, 1981). Erosion of terrestrial organic matter (OM) is known to be a major source of OC in fjordic sediments (Cui et al., 2017) and Scottish peatlands hold 1620 Mt of OC (Chapman et al., 2009). Shetland peatlands cover over one third of the archipelago's total area and their erosion accounts for a loss of 1 to 4 cm yr^{-1} from peat surfaces (Birnie, 1993). Eroding peatlands may therefore hold the potential to contribute significantly to OC burial and accumulation in Shetland's voes and to the overall coastal OC store of this region. Nevertheless, little is known about the long-term fate of the OC that may accumulate and become stored in these sediments. One of the main reasons for this knowledge gap is that OC is prone to post-depositional modification and degradation. It is therefore difficult to discern whether a down-core decrease in OC content is due to an actual change in carbon deposition over time or simply to this post depositional loss (Hülse et al., 2017; Stolpovsky et al., 2017).

Benthic foraminiferal assemblages have long been used as indicators of environmental change and to reconstruct the intrinsic characteristics of marine ecosystems. A recent study (Duffield et al., 2017) explored the relationship between foraminiferal assemblages and variations in OC content in the surficial and recent sediments of Norwegian fjords, highlighting temporal trends in OC fluxes and ecological quality status. In this study, we evaluate the use of modern benthic foraminifera as bio-indicators of OC content in six voes (fjords) on the west coast of Shetland whose sedimentation dynamics are likely influenced by the erosion of OC rich peat from the surrounding catchments (Fig. 1). As benthic foraminifera are sensitive to changes in OC content in the sediments (Alve et al., 2016 and references therein), changes in their assemblage composition should reflect synchronous variations in the amount of OC reaching the seafloor. Given the high preservation potential of their tests in marine sediments (e.g. Murray, 2003; Thomas et al., 1995), foraminifera potentially offer a means to discriminate primary OC signals from post-depositional OC degradation and may provide a tool to validate studies and models reconstructing past changes in the carbon cycle (Hülse et al., 2017; Stolpovsky et al., 2017). Nevertheless, the relationship between OC and foraminiferal assemblages is not straightforward and warrants further investigation.

Early studies developed conceptual models explaining benthic foraminiferal microhabitat preferences based on changes in the organic flux reaching the seafloor and bottom water oxygenation (Jorissen et al., 1995; Van Der Zwaan et al., 1999). These authors found that in well-oxygenated bottom waters, food is usually the limiting factor controlling the distribution of benthic foraminifera, whereas under eutrophic conditions, bottom water oxygenation may become the dominant control on the composition of foraminiferal assemblages (Jorissen et al., 1995; Van Der Zwaan et al., 1999). Typically, food availability is deeply connected to OC fluxes and the transport of organic matter to the seafloor. In this context, it becomes crucial to understand not only food availability in terms of the quantity of OM reaching the seafloor, but also to understand the quality of this food source. For example, refractory organic matter (ROM), often derived from reworked terrestrial sources, is known to be of low nutritional value for benthic foraminifera compared to the more bio-available form of labile organic matter (LOM) (e.g. Fontanier et al., 2008; Jorissen et al., 1998). It should also be noted that OC accumulation rate and bottom water oxygen concentrations are often related, with low oxygen concentrations reducing the reworking/remineralisation of OM and thus

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favouring its preservation in marine sediments. In certain environments, this implies that similar OC fluxes could result in different OC accumulation rates depending on bottom water oxygenation, the latter defining the composition of benthic foraminiferal assemblages (Van Der Zwaan et al., 1999).

To investigate the relationships between sedimentary OC in six west Shetland voes and the associated changes in benthic foraminiferal assemblages, this study combines geochemical and micropalaeontological techniques to: 1) Fingerprint the source (terrestrial vs. marine) and quality (refractory vs. labile) of organic matter and the form (organic vs. inorganic) of sedimentary carbon. 2) Establish benthic foraminiferal biogeography in Shetland's voes from recent surficial sediments. 3) Investigate the use of benthic foraminifera as bio-indicators of OC content in coastal sediments and their potential for palaeo-OC reconstruction purposes.

2 Material and Methods

2.1 Area of study and sampling strategy

Shetland is a subarctic archipelago covering a total area of 1466 km^2, lying about 170 km north of mainland Scotland, UK (Fig. 1). Several voes dissect Shetland's coastline, providing an extended route for the transport of OC rich material from land to sea (Cui et al., 2016). The current study focuses on six voes on Shetland's west coast: Clift Sound, Sand Sound, Olna Firth, Aith Voe, Busta Voe and Vaila Sound (Fig. 1, Table 1). These six voes are characterised by different geomorphologies, freshwater inputs, proximity to the open sea and intensity of local currents; together these characteristics determine the amount of carbon deposited in the sediments. For a detailed description of each voe, please refer to the Supplementary Material.

The MV Moder Dy 2015 cruise in west Shetland surveyed the six voes in August 2015 (Table 1). Marine surface sediments were sampled using a Duncan and Associates Van Veen grab with a sampling area of 0.1 m^2. Twenty-three surface sediment samples were obtained by scraping the top layer of each grab with a domestic spoon, samples were then stored in a cold-box in sealed plastic bottles. For foraminiferal analysis, aliquots of surface sediments were stained with Rose Bengal to allow the identification of living foraminifera, as detailed in Schönfeld et al. (2012). It should be noted that foraminiferal counts are 'total' (live + dead) because the sampling technique may lead to underrepresentation of 'live' foraminifera. At two sites, MD15-01 and MD15-05, replicate samples were taken (-01A and -01B, -05A and -05B, respectively) to check measurement reproducibility and seafloor heterogeneity.

An earlier field survey of Shetland voes carried out in August 2009 measured bottom water temperature (BWT), salinity (BWS) and oxygen (O_2) at the same locations as this study (Fig. 2); however, no data were collected in Vaila Sound. Oceanographic measurements were made using a SeaBird SBE 19 plus conductivity-temperature-depth (CTD) profiler with a dissolved oxygen probe. Bottom water salinity remains almost constant across these voes ranging between 35 and 35.2 p.s.u. with the exception of MD15-06 where BWS reached a minimum of 34.7 p.s.u (Fig. 2b). Similarly, bottom waters are mostly well-oxygenated in all the voes (O_2 > 5mg l^{-1}) with the exception of Olna Firth where lower O_2 were observed (Fig. 2c).

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2.2 Particle size measurement

Particle size analyses were performed on a sub-set of surface sediments using a Coulter LS230 particle size analyser to quantify the volume (%) of the various grain sizes in each sample. We identified three categories based on the International Organisation for Standardisation (ISO) scale: 'Clay' includes particles < 2 µm, 'Silt' particles between 2-63 µm, and 'Sand' particles > 63 µm (upper limit of 2000µm). Before analysis, sediment samples were digested using 30 % hydrogen peroxide to remove organic matter, and 10 % hydrochloric acid to remove carbonate, following a similar methodology to that outlined by Austin and Evans (2010). Digestions were carried out to obtain an insoluble residue useful for comparing sediments having very different carbonate and organic matter content, which otherwise might skew the measurements because of *in situ* processes.

2.3 Loss on ignition (LOI) analysis

Loss on ignition measurements were carried out to quantify the percentage of total organic matter (TOM), and the respective amounts of labile (LOM) and refractory organic matter (ROM). The initial combustion temperature was set to 250˚ C, as significant mass loss of LOM has been recorded at this temperature (Mook and Hoskin, 1982). Sediments were successively heated to 550˚ C to quantify ROM, as refractory terrestrial and aquatic OM will likely burn off at this temperature (Kristensen, 1990). For the detailed steps of this analysis, please refer to the supplementary information.

2.4 Bulk carbon elemental & stable isotope analyses

Surface sediment samples were analysed to determine bulk elemental C and stable isotope ($\delta^{13}C$) values. Each sediment sample was freeze dried and homogenized and approximately 12 mg of milled sediment were placed into silver capsules and a further 10 mg into tin capsules.

The samples encapsulated in silver underwent an acid fumigation step (Harris et al., 2001) to remove inorganic carbon (IC). After drying for 24 hours at 40 °C, both OC and $\delta^{13}C$ were measured using an elemental analyser coupled to an isotope ratio mass spectrometer (IRMS) at the NERC Life Science Mass Spectrometer Facility (Lancaster, UK). The standard deviation of $\delta^{13}C$ triplicate measurements was 0.07 ‰ and $\delta^{13}C$ values are reported in standard delta notation relative to Vienna Pee Dee Belemnite (VPDB).

The samples in tin capsules were analysed for total carbon (TC) using a Elementar Elemental Analyser (EA) at the School of Geography and Sustainable Development, University of St Andrews (Verardo et al., 1990). Precision of the analysis is calculated based on repeat measurements of standard reference material B2178 (medium Organic content standard from Elemental Microanalysis, UK) with C = 0.08%.

These results were combined together to calculate the quantity of IC in each sample; the percentage of OC was subtracted from the percentage of TC to obtain the percentage of IC.

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2.5 Binary mixing model

To discriminate between marine-sourced and terrestrially-sourced OC, we used a two end-members (binary) mixing model assuming a two-point source of OC, marine and terrestrial, following Thornton and McManus (1994). We used $\delta^{13}C$ measurements in the sediment samples as tracer of the source of OC in west Shetland voes and calculated the fraction of terrestrially-sourced OC (OC_{terr}) as follows:

$$OC_{terr} = (\delta^{13}C_{mar}-\delta^{13}C_{terr}) / (\delta^{13}C_{mar}-\delta^{13}C_{sample})$$

$$OC_{terr} + OC_{mar} = 1$$

where the end-member value for marine-sourced OC ($\delta^{13}C_{mar}$) was taken from Smeaton and Austin (2017) based on phytoplankton, zooplankton, macroalgae, and benthic microalgae carbon stable isotope values, while the end-member for terrestrially sourced OC ($\delta^{13}C_{terr}$) was based upon the work of Thornton et al. (2015) who studied the distribution of carbon stable isotope in Scotland's topsoil; OC_{mar} indicates the fraction of marine-derived OC. We assumed no isotope discrimination between the source and the surface sediment samples.

2.6 Benthic foraminiferal counts and statistics

Rose Bengal stained surface sediments were wet sieved over a 63 μm mesh. The residues were oven-dried at < 60 °C, then weighed and sieved into 63-150 μm and >150 μm size fractions. Both size fractions were analysed independently as part of an associated study which demonstrated that a more holistic picture of environmental change is obtained when benthic foraminiferal assemblages of both size fractions are compiled together; this improves the representation of the entire assemblage and results in more reliable environmental reconstructions. Similar findings were also reached by Weinkauf and Milker (2018 and references therein). Depending on sample volume, each sample was divided into a number of splits using a standard splitter and, when possible, at least 300 specimens were picked to ensure statistical significance when discussing benthic foraminiferal assemblages' diversity and composition (Schönfeld et al., 2012). Species were identified following Austin (1991) and counted using a tally sheet; notes on tests preservations were made when signs of etching were observed. As the main objective of this study is to provide a tool for the interpretation of fossil foraminiferal assemblages and their relationship with changes in OM and OC content in sediments over time, total assemblages (live + dead) were analysed (e.g.: Conradsen, 1993). Our priority is to understand how benthic foraminifera respond to prevailing (long-term) environmental conditions, rather than to seasonal variability.

Relative benthic foraminiferal abundances were calculated for each sample (Supplementary Table 1). Ten taxa of benthic foraminifera were observed in each one of the six voes and had relative abundances of more than 10 % in at least one of the 21 stations studied (Table 2): *Ammonia* spp., *Bulimina marginata*, *Buliminella elegantissima*, *Cibicides* spp., *Eggerelloides scaber*, *Elphidium gerthi*, *Elphidium margaritaceum*, *Elphidium excavatum*, *Rosalina* spp. and *Stainforthia fusiformis*. It should be noted that we grouped under the name *E. excavatum* both forma *selseyense* and forma *clavata* despite them being

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recognised as genetically different species (Darling et al., 2016). It was not possible to consistently identify and discriminate between these two species, hence the use of a common identifier. These ten dominant taxa were imaged with a scanning electron microscope (SEM) at the Scottish Oceans Institute, University of St Andrews (Fig. 4). Altogether, these ten taxa account for more than 70% of the total assemblage at each station (Table 2).

The Palaeontological Statistics software package PAST (version 3.16, Hammer et al., 2001) was used to analyse relationships between benthic foraminiferal assemblages through cluster analysis and non-metric multidimensional scaling (MDS) based on the Bray-Curtis similarity index and relative abundance data. Cluster analysis showed good reproducibility of benthic foraminiferal assemblages (Supplementary Fig. 1) with the two pairs of replicate stations exhibiting assemblage reproducibility of 90% (MD15-01A and 01B) and 94% (MD15-05A and -05B).

3 Results

3.1 Sediment analyses

3.1.1 Particle size distribution in west Shetland voes

A highly variable pattern is evident in the particle size distribution of the sediments of west Shetland voes (Fig. 3). Clift Sound, Olna Firth, stations MD15-08 and-10 in Sand Sound, and station MD15-13 in Aith Voe have silt dominated sediments (>65%), despite having very different geomorphologies (unrestricted vs. restricted) and circulation patterns (high vs. low energy) (Fig. 3). Similarly, stations MD15-06, -07,and -09 in Sand Sound, MD15-14 in Aith Voe, MD15-15 and -17 in Busta Voe, and stations MD15-20 and -21 in Vaila Sound are dominated by sandy sediments (>50%) despite their different environmental settings (Fig.3).

3.1.2 Organic matter content in west Shetland voes

Total OM is generally higher in proximity to land than in sites more exposed to the open sea (Fig. 4b). Site MD15-20 (Vaila Sound) has the lowest percentage of total OM (3.05%), ROM (2.24%) and LOM (0.81%), whereas site MD15-08 (Sand Sound) has the highest percentage of total OM (20.85%) and ROM (12.37%). LOM is the highest (8.81%) at site MD15-11 (Olna Firth) (Table 3; Fig. 4b).

3.1.3 Carbon content and source in west Shetland bulk sediments

The percentage of total carbon (TC) in west Shetland voes varies between 4.31 and 14.32%, with the highest percentage found at station MD15-05 (Clift Sound) and the lowest at station MD15-09 (Sand Sound). Organic carbon (OC) content ranges

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between 2.0-8.51%, peaking at station MD15-11 (Olna Firth) and reaching a minimum at site MD15-17 (Busta Voe) (Fig. 4a; Table 3). The percentage of inorganic carbon (IC) varies between and 0.35-7.51% and is the highest in station MD15-03 (Clift Sound) and the lowest in station MD15-14 (Aith Voe) (Fig. 4a). We excluded from the data set site MD15-15, as this sample contains coal, which is believed to have been shed accidentally from a commercial vessel. In general, there is a tendency for high OC and low IC at the head of the voes and the reverse occurs where the voes connect to the sea (Fig. 4a).

In terms of carbon source, OC_{terr} is generally higher at sites closed to land than in proximity to the open sea (Fig. 4c). It reaches a peak of 72% at station MD1-06 in Sand Sound, and a minimum of 21% at station MD15-03 (Fig. 4c).

3.1.4 Carbon stable isotopes ($\delta^{13}C$)

Bulk sediment $\delta^{13}C$ varies between -25.41 and -21.26 ‰ with an average value of -22.67 ‰ (Table 3), in range with carbon stable isotope measurements of other Scottish sea lochs: Sunart (average -22.37 ‰), Teacius (average -24.79 ‰) (Smeaton and Austin, 2017), Creran (-24.7 to -21.3 ‰), Etive (average -25.8 ‰) (Loh et al., 2008). The lowest value (-25.41 ‰) is recorded at site MD15-06 (Sand Sound) and the highest (-21.26 ‰) at station MD15-03 (Clift Sound); a clear seaward trend of increasing $\delta^{13}C$ values is evident in all six voes (Fig. 4c).

3.2 Benthic foraminiferal assemblages in west Shetland voes

We observed a total of 64 species of benthic foraminifera in the 21 stations analysed in this study, including the two replicates samples (Supplementary Table 1, Supplementary Fig. 2). Nine of the observed species are agglutinated: *Ammoscalaria runiana*, *E. scaber*, *Connemarella rudis*, *Haplophragmoides* sp., *Reophax fusiformis*, *Reophax scotii*, *Spiroplectinella wrightii*, *Textularia earlandii*, *Trochammina* sp. Of these, only *E. scaber* and *Trochammina* sp. were observed in all 21 stations (Supplementary Table 1). However, *E. scaber* relative abundance varies from 0.4 % in Vaila Sound (MD15-20) to 43.0 % in Sand Sound (MD15-10) (Fig. 5), whereas *Trochammina* sp. relative abundance reaches a maximum of 2.3 % in Clift Sound. Other common agglutinated species are *Haplophragmoides* sp. and *S. wrightii*, with relative abundance ranging from 0.1 % (Vaila Sound) to 2.9 % (Busta Voe), and 0.1% (Olna Firth) and 3.9 % (Clift Sound), respectively (Supplementary Table 1). The distribution of the remaining agglutinated species is patchy and their relative abundance varies around the average value of 1 % (Supplementary Table 1).

Six of the 64 observed species belong to the porcelaneous group Miliolina: *Cornuspira* sp. *Miliolinella subrotunda*, *Quinqueloculina bicornis*, *Quinqueloculina seminula*, *Quinqueloculina* sp., *Spiroloculina rotunda*. Of these, the most abundant species is *Q. seminula*, which is present in low numbers in five of the six studied fjords, it does not occur in Aith Voe. The general distribution of Miliolina is patchy and their relative abundance varies around the average value of 0.5 %, with the highest value of 2.4 % observed at the mouth of Clift Sound (MD15-05B) (Supplementary Table 1).

The remaining 49 species are hyaline foraminifera of which 9 species belong to the ten dominant taxa having relative abundances of > 10 % in at least one of the 21 stations studied (Table 2; Fig. 5). Additionally, of all the hyaline species, 20 are

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rare having relative abundances < 1 %, while 18 species are frequent having a relative abundances between 1-5 % and only 2 species are common having relative abundances ranging between 5-10 % (Supplementary Table 1, Supplementary Fig. 2). In general, the most abundant taxa observed in west Shetland's six voes is the hyaline *Cibicides* spp., followed by the agglutinated *E. scaber* (Table 2 and Fig. 5).

4 Discussions

4.1 Particle size distribution in west Shetland voes

The particle size data from the voes of Shetland exhibit a large range with the silt and sand fractions, but little variation in the proportion of clay (Fig. 3a). These samples suggest a significant size sorting which is consistent with the tidal and estuarine dynamics of these voes. In Vaila Sound, an unrestricted geomorphology (Fig. 1), intensified bottom currents and winnowing of finer sediments (Cefas, 2009) appear to control local grain size distribution (Fig. 3). Here, stations exposed to the open sea and intensified bottom currents are characterised by coarser sediments (MD15-20 and 21), whereas more sheltered and calm locations (MD15-18 and -19) are characterised by finer sediments (Fig. 3a). In contrast, sediments in Clift Sound are silt-dominated (Fig. 3a) despite the unrestricted geomorphology (Fig. 1) and vigorous bottom currents (Cefas, 2007a). It appears that a different mechanism than winnowing of finer sediments by intensified bottom currents drives particle size distribution here. In Clift Sound, the overall low percentage of OM together with the relatively high IC content and $\delta^{13}C$ values (Fig. 4) point to a reduction in terrigenous input as the main driver of particle size distribution (Fig. 3). Conversely, in Sand Sound, an increased terrigenous input appears to be responsible for the coarser sediments found in stations MD15-06, -07 and -09 (Fig. 3). The head of the voe is a sheltered area that receives freshwater from a significant number of rivers draining the surrounding land (Cefas, 2007b, 2008), resulting in an overall high percentage of OM and OC, low $\delta^{13}C$ values (Fig. 4) and coarser sediments (Fig. 3a). In contrast, station MD15-08 at the head of Sand Sound is characterised by finer sediments (Fig. 3a) despite the high terrigenous supply at this location (Fig. 4). It is possible that, due to the restricted geomorphology of Sand Sound (Fig. 1), weak currents are unable to transport the coarse material to station MD15-08, the deepest station at the head of the voe (11 m vs. 6 m). In Sand Sound inner basin (MD15-10), silty sediments dominate (Fig. 3a). Here, a combination of reduced terrigenous input and/or weaker currents could be driving particle size distribution. Nørgaard-Pedersen et al. (2006) found similar drivers of particle size distribution in Loch Etive, a sea loch in mainland western Scotland. Vigorous bottom currents and/or enhanced terrigenous supply would result in locally coarse sediments, whereas weaker currents and/or limited terrigenous supply would result in finer sediments. Due to the duality of drivers determining particle size distribution in west Shetland voes (bottom currents intensity vs. terrigenous supply), it is challenging to consistently group locations based on granulometry data (Fig. 3a) or identify a relationship between grain size distribution and OC content in the sediments (Fig. 3b). For these reasons, we have not included these data in the following discussion characterising Shetland's sediments and benthic foraminiferal biogeography.

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4.2 Spatial distribution, quantity, quality and source of organic matter, and carbon in west Shetland voes.

A seaward gradient is evident with regard to the quantity, quality and source of organic matter and carbon measured in the six west Shetland voes (Fig. 4). Typically, carbon is present in the sediments as IC and OC and the distribution of these two forms of carbon tends to mirror one another, with low IC and high OC percentages towards land while the reverse occurs seaward (Fig. 4a). Organic matter follows the same seaward pattern of OC distribution regardless of its quality (refractory vs. labile); however, ROM content in the sediments is always higher than LOM (Fig. 4b) owing its resilience to degradation.

About the source of OM and OC, low $\delta^{13}C$ and high OC_{terr} values characterise the head of the voes where high ROM and OC are accumulated, indicating that the source of this material is terrestrial (Fig. 4). Terrestrial OM is more stable and resistant to decay than marine OM (Batten, 1996) and it is enriched in ^{12}C from soils resulting in lower $\delta^{13}C$ values than marine OM derived from algae (e.g. Balasse et al., 2005; Ficken et al., 1998; Marconi et al., 2011; Schiener et al., 2014; Schmidt and Gleixner, 2005). Sand Sound is dominated by organic rich and terrestrially sourced material as it receives freshwater and terrestrial OM from the numerous streams draining the surrounding land (Fig. 4). In contrast, Clift and Vaila Sounds are characterised by marine sourced material and high IC content in their sediments (Fig. 4) as the inflow of Atlantic Ocean waters improves the voes' potential for IC storage (Burrows et al., 2017; Smeaton et al., 2016, 2017). Olna Firth has the highest LOM percentage as this relatively deep and micro-tidal basin (Cefas, 2013) is prone to water mass stratification which can lower bottom water oxygenation (Fig. 2) and slow down the reworking/remineralisation of OM favouring the preservation of the more labile material (Fig. 4).

A slight deviation from this overall trend was recorded at station MD15-05. We speculate that the unusually high TC content at station MD15-05 at the mouth of Clift Sound (Fig. 4a) could relate to the MV Braer oil spill of 1993. It appears that after the spill, due to the adverse weather, the oil became deposited in the sediment of Burra Haaf, just west of Clift Sound (Fisheries Research Services). Despite the fact that oil levels in the contaminated sediments have been decreasing steadily over the intervening years, it is plausible that the OC measurements in this study are detecting remnants of this event.

4.3 Modern benthic foraminiferal biogeography in Shetland.

We used non-metric MDS based on the Bray-Curtis similarity index and relative abundance data to analyse resemblances between foraminiferal populations in the 21 studied stations. We defined four groups with 75% confidence (Fig. 5). Group 1 includes assemblages dominated by the genus *Elphidium*, and by *E. scaber*; the head of Clift Sound falls into this cluster (MD15-06 to -09). Group 2 contains assemblages dominated by *E. scaber* with *S. fusiformis* and *B. marginata* as associated species; the inner basin of Sand Sound (MD15-10), Olna Firth (MD15-11 and -12), and MD15-16 in Busta Voe are part of this cluster. Group 3 consists of assemblages characterised by relatively high diversity and no obvious dominant species; MD15-01A and -01B (head of Clift Sound), MD15-13 and -14 (Aith Voe); MD15-15 (Busta Voe); MD15-18 and -19 (head of Vaila Sound) belong to this group. Group 4 comprises assemblages dominated by the genus *Cibicides* with *Rosalina* as associated taxa and includes stations MD15-02 to MD15-05 in Clift Sound and stations MD15-20 and -21 in Vaila Sound (Fig. 5). Station

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MD15-17 in Busta Voe falls just outside the 75% confidence interval defining Group 4; however, cluster analysis (Supplementary Fig. 1) shows ~82% similarity between the assemblage of this site and station MD15-20 and more than the 70% similarity with the rest of the locations belonging to Group 4. Therefore, for the purposes of this discussion, we include station MD15-17 in Group 4. In general, foraminiferal assemblages do reflect the geomorphology of the six voes (restricted vs. unrestricted basins) and the seaward gradient in OM and OC distribution (Figs. 4 and 5).

4.3.1 Group 1 – *Elphidium* spp. and *E. scaber*

The genus *Elphidium* becomes the dominant taxa (relative abundance ~38%) at the head of Sand Sound (Fig. 5), a very shallow (max depth 11m) and organic rich basin (OC 5.2%, ROM 7.3% and LOM 4.9%; Fig. 4). Specifically, *E. excavatum* is the most abundant *Elphidium* (~21%, Fig. 5), as it can live close to freshwater outflows, in low energy environments, and tolerates high OC and OM concentrations (Alve and Murray, 1999; Jennings, 2004; Korsun et al., 2014; Mendes et al., 2012). This is consistent with streams draining into the head of Sand Sound transporting terrestrial and organic rich material into the voe (Figs. 1 and 4). Additionally, the high relative abundance of *E. scaber* (26%) at the same locations purports a low energy environment, possible water mass stratification and high ROM (Evans et al., 2002; Fontanier et al., 2002; Murray, 1992; Nørgaard-Pedersen et al., 2006). Thus, assemblages dominated by *Elphidium* spp. and *E. scaber* are likely to be found in land-locked regions influenced by riverine/freshwater inputs like the head of fjords with a restricted geomorphology.

4.3.2 Group 2 –*E. scaber, B. marginata* and *S. fusiformis*

Eggerelloides scaber, *B. marginata* and *S. fusiformis* are species typical of organic rich sediments, stratified waters and possibly low oxygen concentrations, often suggesting stressed environments (Evans *et al.* 2002; Fontanier *et al.* 2002; Murray 1992; Alve 1994; 2003; Nørgaard-Pedersen *et al.* 2006). *Bulimina marginata* (~9%) seems to thrive in regions with high ROM (9.6%) due to its ability to use OM of low nutritional value as food (Klitgaard-Kristensen & Buhl-Mortensen 1999) (Fig. 4). *Stainforthia fusiformis* (~21%) is a known opportunistic species which follows high OM gradients at stratification fronts (Alve, 2003; Alve and Murray, 1997; Scott et al., 2003). In general, high organics in the sediments (Fig. 4) can result in low oxygen concentrations at the seafloor (Fig. 2) due to eutrophication and increased respiration (Bianchi et al., 2016). Additionally, the production of humic acids as a by-product of organic matter biodegradation may lower the pH (Bauer and Bianchi, 2012), overall making the environment more hostile for foraminifera to calcify, which could result in the dominance of agglutinated species like *E. scaber* (~31%) (Fig. 5). Etching of *Ammonia* spp. shells (Supplementary Fig. 2) in Olna Firth supports local acidification of bottom waters, which is substantiated by the low IC content (2.6%) of stations MD15-11 and -12 (Fig. 3). Therefore, assemblages characterised by *E. scaber*, *B. marginata* and *S. fusiformis* reflect a stressed environment with a heavily stratified water column - possibly low oxygen levels, and organic rich sediment especially in ROM of low nutritional value.

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4.3.3 Group 3 – no obvious dominant taxa

The assemblage of Group 3 is characterised by high relative abundance of taxa with affinities for OC (4.9%) and OM (6.3%) like *Elphidium* (8%), *E. scaber* (13%) and *B. elegantissima* (5%) (e.g.: Mendes *et al.* 2012; Alve & Murray 1999) as well as high energy taxa like *Cibicides* (22%) and *Ammonia* (5%) (Scott et al., 2003) (Figs. 4 and 5). The low relative abundance of *B. marginata* (2%) and *S. fusiformis* (5%) additionally suggests that OM at these locations has good nutritional value and current activity is present at these sites (reduced stratification, possibly vertical mixing). Therefore, assemblages showing this composition are likely to reflect depositional environments with mild or episodic current activity and moderate organic content.

4.3.4 Group 4 – *Cibicides* spp. and *Rosalina* spp.

In Group 4, the dominant taxa is *Cibicides* (~57%) followed by *Rosalina* (8%) (Fig. 5), which reflect marginal to open sea conditions and vigorous bottom currents (Alve & Murray 1999; Jennings 2004; Klitgaard-Kristensen & Sejrup 2002). A clear relationship is evident between low organics (OC 4.2%, ROM 3.2% LOM 1.5%) (Fig. 4) and the dominance of *Cibicides* (Fig. 5). All locations falling in Group 4 are on the path of Atlantic Ocean waters entering the fjords meaning that this assemblage composition is most likely found in areas dominated by sea processes.

4.3.5 Other drivers influencing modern benthic foraminifera distribution

Seawater properties can compete with OC and OM in influencing the distribution of benthic foraminifera across the six voes, especially BWS and O_2 concentration may exert a second-order control on the composition of foraminiferal assemblages (Jorissen et al., 1995; Van Der Zwaan et al., 1999). A highly variable BWS generally influences the distribution of stenohaline (strictly marine species) and euryhaline species (able to adapt to a wide range of salinity) along BWS gradients; however, in Shetland, BWS is almost constant across the voes (Fig. 2) having very minimal influence on benthic foraminiferal assemblages. Similarly, bottom waters are mostly well-oxygenated ($[O_2] > 5$ mg l^{-1}) in all the voes (Fig. 2), meaning that food availability is the main driver of changes in the composition of benthic assemblages (Jorissen et al., 1995; Van Der Zwaan et al., 1999). In turn, food availability is deeply connected to organic fluxes and transport of organic matter to the seafloor, hence the relationships found between benthic foraminiferal assemblages and depositional environments (Fig. 5). Only one station in Olna Firth (MD15-11) has poorly oxygenated bottom waters ($O_2 < 5$ mg l^{-1}), which could represent the onset of hypoxic conditions (Vaquer-Sunyer and Duarte, 2008) potentially affecting the distribution of benthic foraminifera (Fig. 2). The benthic foraminiferal assemblage of this location reflects not only organic rich sediments, but also stratified waters and low oxygen concentrations, pointing to a stressed environment. However, this is far from the dysoxic/anoxic conditions required for a switch from food-limited to oxygen-limited conditions (Fig. 2). Therefore, the quantity and quality (labile vs. refractory) of organic matter reaching the seafloor is still the major control on the benthic foraminiferal assemblage found at station MD15-11.

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4.4 Comparison with other fjords.

4.4.1 Spatial distribution and sources of OM and C.

The processes governing OM and C inputs in west Shetland voes are comparable with those reported for other Scottish fjords by Smeaton et al. (2017) who found a relationship between the OC content in Scottish fjords and their physical characteristics such as tidal range, precipitation, catchment area and runoff. However, as catchments in west Shetland transport sediment from the adjacent C rich peatlands into the marine system, west Shetland voes overall store more total C compared to other Scottish fjords (Smeaton et al., 2016). On average, the total C content of the six west Shetland voes is 8.8%, whereas total C in other Scottish fjords ranges between 0-8% (Smeaton et al., 2017 in Fig. 5). Nevertheless, as stated in Smeaton et al. (2017), there are similarities between Scotland's mainland fjords and these six voes in west Shetland. For example, fjords like Loch Broom and Little Loch Broom in Scotland have relatively unrestricted geomorphologies which makes them highly efficient in capturing IC, similar to Clift Sound and Vaila Sound in Shetland (Fig. 4). Transient low oxygen concentrations in Scotland's mainland fjords seem to be an important factor in the burial and preservation of OC (Gillibrand et al., 2006). Similarly, low oxygen levels in Olna Firth (Fig. 2), also indicated by benthic foraminiferal assemblages (Fig. 5), are associated with the preservation of labile organic matter and OC (Fig. 4).

4.4.2 Benthic foraminiferal assemblages

Our knowledge of benthic foraminiferal assemblages in Scottish fjords is limited to studies based on fauna from Loch Etive (Murray et al., 2003; Nørgaard-Pedersen et al., 2006) and their response to changes in sea level and terrigenous input entering the catchment. Species in common with west Shetland voes include *Cibicides lobatulus*, *Ammonia batavus* and *E. scaber*. Assemblages dominated by *C. lobatulus* and *A. batavus* in Loch Etive indicate marine conditions and intensified bottom currents possibly associated with deep water renewal events (Nørgaard-Pedersen et al., 2006). This agrees well with the assemblages dominated by *Cibicides* spp. and *Rosalina* spp. observed in Clift Sound and Vaila Sound suggestive of high-energy environments (Fig. 5). The dominance of agglutinated species such as *E. scaber* occurs in Loch Etive when OM deposition increases eventually resulting in low oxygen levels in bottom waters and carbonate dissolution (Murray et al., 2003; Nørgaard-Pedersen et al., 2006). We observed similar environmental conditions and assemblages in Olna Firth, the middle of Busta Voe and inner basin of Sand Sound (Fig. 5).

Similarities can also be observed between assemblages from the Scottish coast and shelf and west Shetland voes. Klitgaard-Kristensen and Sejrup (2002) and Murray (2003) found high abundance of *C. lobatulus* and *Rosalina* sp. in areas with strong currents, low organics and coarse-grain sediments. We do not see a correlation between grain size and *Cibicides* or *Rosalina* in our samples due to the multiple forcings driving particle size distribution in Shetland (bottom current intensity vs. terrigenous supply) (Fig. 2); however we do observe a clear correlation between high relative abundance of *Cibicides* and *Rosalina* and low organics and high energy environments (Figs.4 and 5). Klitgaard-Kristensen and Sejrup (2002) and Murray (2003) also

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found a relationship between the dominance of *B. marginata* and low oxygen levels, high organics and fine sediments. Additionally, Murray (2003) also observed *S. fusiformis* dominated assemblages in association with high availability of LOM and low oxygen levels. Overall, this compares really well with the assemblage we described in Group 2 (Fig. 5).

A recent study by Murray and Alve (2016) synthesise benthic foraminiferal biogeography in NW European fjords, mostly focusing on Norwegian fauna. Similarities with west Shetland voes include *C. lobatulus* as an "Atlantic species" pointing to the influence of Atlantic Ocean waters on local estuarine/fjordic circulation. Assemblages dominated by *E. scaber* have been observed in Oslofjord and Drammensfjord in Norway (Alve 1995; Alve & Nagy 1986) and Gullmar Fjord in Sweden (Qvale et al. 1984), generally associated with organic rich sediments and shallow waters. Additionally, Klitgaard-Kristensen and Buhl-Mortensen (1999) found high relative abundance of the opportunistic species *S. fusiformis* in Norwegian fjords with a high per cent of sand and organic carbon, and assemblages characterised by *B. marginata* in environments rich in ROM.

Overall, benthic foraminiferal assemblages do respond to variations in the amount of OM and OC becoming deposited in fjord sediments, and comparisons can be drawn between fjordic systems. We recognise that this is a qualitative approach and we are not able to quantify changes in OM or OC content reaching the seafloor based on foraminiferal assemblages. However, this study provides an initial insight into the use of foraminifera to discriminate between post-depositional OC degradation and actual OC burial and accumulation in fjords, as foraminifera would only preserve the latter information in their assemblage composition. We reckon however that extreme conditions at the seafloor may result in post depositional dissolution of benthic foraminiferal tests, which would obviously compromise the assemblage composition. For this reason, we recommend to carefully consider the level of preservation of each assemblage and, when necessary, make appropriate remarks on etching of tests and other possible signs of tests dissolution (e.g. Khanna et al., 2013). Additionally, assemblages dominated by agglutinated species like in Group 2 might have a lower diagnostic potential in their application down-core compared to their hyaline and porcelaneous counterpart due to a tendency of being poorly preserved in the fossil record. Nevertheless, foraminifera are generally abundant in the fossil record and this approach shows down-core potential to support reconstructions of changes in OC burial and accumulation over time and could be paired with statistical and spatial model studies to tease out post depositional OC degradation.

5 Conclusions

Our records from west Shetland voes provide new insights into sediments provenance and type, modern benthic foraminiferal spatial distribution across environmental gradients, and their use as indicators for OC enrichment in fjordic systems. Our results support a terrestrial source of both OM and OC in land-locked areas and calm depositional settings in west Shetland voes. Additionally, in locations characterised by low bottom water oxygenation, LOM is present in higher percentages than in well ventilated sites. Conversely, marine processes govern IC storage, with higher concentrations in marginal to open marine settings. Benthic foraminiferal assemblages reliably reflect changes in OC content reaching the seafloor and show great

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potential as bio-indicators to trace these changes over time. We were able to identify four groups (75% confidence) based on foraminiferal relative abundance data, which reflect four types of depositional settings: 1) Land-locked with high organics and very weak currents; 2) Stressed environment with high organics, heavily stratified water column and low oxygen concentrations; 3) Transitional environment characterised by moderate concentrations of organics and mild or intermittent currents; 4) Marginal to coastal settings with low organics and a high-energy environment.

Therefore, studies aiming to reconstruct past variations in the amount of OC reaching the seafloor and becoming stored therein could benefit from the use of benthic foraminifera assemblages as a qualitative approach to understand OC preservation in the sediment column and, for example, ground truth model predictions.

Author contribution. WENA and ELGC designed the study. ELGC, JC and CS analysed sediment physical properties, while ELGC and WENA worked on benthic foraminiferal assemblages. KD provided hydrographic data. ELGC prepared the manuscript with contributions from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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Figures Captions

Figure 1: Location Map of Shetland and sampling stations.

Figure 2: CTD profiles. a) Temperature profiles and bottom water tempreature (BWT) at each sampling station. b) Salinity profiles and bottom water salinity (BWS) at each sampling station. c) Dissolved oxygen profiles and oxygen concentration in bottom waters at each sampling station. Dashed line at 5 mg l^{-1} indicate possible onset of hypoxic conditions.

Figure 3: Particle size distribution. a) Ternary plot. b) Trends between particle size and organic matter and carbon content.

Figure 4: Sediment physical properties. a) Inorganic and organic carbon percentage at each site. b) Refractory and labile organic matter content at each site. c) Carbon stable isotope and fraction of terrestrially-source OC (OC_{terr}) at each site.

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Figure 5: Non metric MDS-ordination plot of benthic foraminifera assemblages from each site based on relative abundance data.

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Table 1. Station Data

Location	Sample ID	Longitude	Latitude
Clift Sound	MD15-01	-1.277267	60.11943
Clift Sound	MD15-02	-1.284533	60.09987
Clift Sound	MD15-03	-1.2975	60.07995
Clift Sound	MD15-04	-1.307783	60.06635
Clift Sound	MD15-05	-1.315883	60.04765
Sand Sound	MD15-06	-1.40295	60.24485
Sand Sound	MD15-07	-1.404183	60.24477
Sand Sound	MD15-08	-1.379517	60.24237
Sand Sound	MD15-09	-1.35835	60.24178
Sand Sound	MD15-10	-1.370517	60.22822
Olna Firth	MD15-11	-1.295883	60.36277
Olna Firth	MD15-12	-1.32315	60.36425
Aith Voe	MD15-13	-1.374883	60.28945
Aith Voe	MD15-14	-1.373333	60.3055
Busta Voe	MD15-15	-1.361217	60.39075
Busta Voe	MD15-16	-1.358883	60.38063
Busta Voe	MD15-17	-1.370517	60.3694
Vaila Sound	MD15-18	-1.564083	60.21943
Vaila Sound	MD15-19	-1.5624	60.213
Vaila Sound	MD15-20	-1.582167	60.21428
Vaila Sound	MD15-21	-1.567467	60.20567

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Table 2. Relative abundance of the ten dominant taxa.

Location	Sample ID	*Ammonia* spp.	*B. marginata*	*B. elegantissima*	*Cibicidoides* spp.	*E. scaber*	*E. gerthi*	*E. margaritaceum*	*E. excavatum*	*Rosalina* spp.	*S. fusiformis*	Total %
Clift Sound	MD15-01A	0.2	0.5	2.1	14.5	22.0	10.7	20.1	7.8	3.8	3.0	**84.6**
Clift Sound	MD15-01B	0.2	0.6	2.1	15.0	19.9	10.7	24.3	2.1	3.2	4.4	**82.5**
Clift Sound	MD15-02		0.7	0.3	47.6	10.6	5.4	3.1	1.9	6.6	3.8	**80.3**
Clift Sound	MD15-03	0.2	1.2	0.7	53.7	3.3	10.5	0.8	1.2	5.4	4.6	**81.5**
Clift Sound	MD15-04	0.3	0.9	0.3	48.9	5.2	13.9	0.2		4.7	2.9	**77.3**
Clift Sound	MD15-05A		0.3		57.6	2.1	0.4	0.1		9.7	0.7	**71.0**
Clift Sound	MD15-05B		0.6		56.7	2.1	0.2	0.0	0.5	10.4	1.5	**72.0**
Sand Sound	MD15-06	4.8	0.5	4.6	2.0	25.5	15.6	6.1	15.3		1.3	**75.7**
Sand Sound	MD15-07	6.5	0.8	4.8	5.5	27.7	11.7	7.6	21.5	0.7		**86.7**
Sand Sound	MD15-08	3.0	1.7	11.4	2.0	29.7	7.5	1.4	22.4	0.2	6.3	**85.6**
Sand Sound	MD15-09	5.4	0.9	8.8	1.9	19.7	9.8	10.1	25.6	0.2	1.6	**83.9**
Sand Sound	MD15-10	0.9	3.8	1.9	8.3	43.0	4.7	1.3	5.2	1.4	11.3	**81.8**
Olna Firth	MD15-11		14.8	0.8	10.9	31.9	4.4	1.8	0.8	0.5	23.1	**88.8**
Olna Firth	MD15-12	8.5	8.6	0.7	13.7	22.9	2.2	0.9	2.4	0.3	26.0	**86.2**
Aith Voe	MD15-13	3.0	5.6	12.6	14.2	11.7	9.3	4.0	10.0	0.3	7.3	**78.1**
Aith Voe	MD15-14	12.6	4.1	4.6	26.1	10.0	8.2	0.9	4.3	1.8	8.2	**80.8**
Busta Voe	MD15-15	1.0	2.8	2.0	30.1	12.9	7.0	5.1	5.6	0.8	8.2	**75.5**
Busta Voe	MD15-16	5.7	8.5	0.9	7.6	26.4	5.0	1.2	5.1	0.2	23.1	**83.7**
Busta Voe	MD15-17	2.2	1.2	0.1	70.1	2.8	1.4	0.8	1.5	1.1	4.8	**86.1**
Vaila Sound	MD15-18	6.1	0.9	11.5	24.4	9.5	7.6	9.0	9.2	2.3	4.0	**84.5**
Vaila Sound	MD15-19	10.2	2.1	1.8	29.6	8.8	8.3	3.4	9.2	4.5	4.3	**82.3**
Vaila Sound	MD15-20		0.4	0.1	68.7	0.4	1.2	0.9	1.2	14.7	0.5	**88.2**
Vaila Sound	MD15-21	0.3	1.4	0.3	56.8	1.9	1.9	1.0	2.9	11.3	1.4	**78.9**

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Table 3. Sediment physical properties

Location	Sample ID	Water depth (m)	$\delta^{13}C$	%IC	%OC	%LOM	%ROM	$\%OC_{terr}$
Clift Sound	MD15-01A	13	-23.16	3.55	5.44	4.35	6.99	45
Clift Sound	MD15-01B	13	-23.16	3.55	5.44	3.94	7.29	45
Clift Sound	MD15-02	15	-21.92	7.07	2.80	1.59	2.95	30
Clift Sound	MD15-03	23	-21.26	7.51	3.48	1.60	3.38	21
Clift Sound	MD15-04	21	-21.51	4.72	3.58	1.78	3.79	25
Clift Sound	MD15-05A	20	-21.35	7.12	7.20	1.32	4.47	23
Clift Sound	MD15-05B	20	-21.35	7.12	7.20	1.40	4.09	23
Sand Sound	MD15-06	6	-25.41	2.41	4.33	5.32	8.50	72
Sand Sound	MD15-07	6	-24.07	0.53	4.51	2.63	4.26	56
Sand Sound	MD15-08	11	-23.84	1.67	8.28	8.48	12.37	53
Sand Sound	MD15-09	6	-23.64	0.53	3.78	3.07	4.24	50
Sand Sound	MD15-10	21	-22.87	5.57	4.80	4.92	6.68	41
Olna Firth	MD15-11	31	-22.36	3.02	8.51	8.81	10.89	35
Olna Firth	MD15-12	35	-22.65	2.15	7.54	8.07	11.83	38
Aith Voe	MD15-13	12	-23.55	4.70	5.44	5.54	7.07	49
Aith Voe	MD15-14	36	-23.38	0.35	4.48	2.06	3.92	47
Busta Voe	MD15-15	18	-	-	-	2.99	8.27	-
Busta Voe	MD15-16	26	-23.06	7.40	3.27	5.48	9.19	43
Busta Voe	MD15-17	24	-22.59	5.47	2.06	2.54	3.93	38
Vaila Sound	MD15-18	14	-21.82	3.70	4.79	3.03	6.00	28
Vaila Sound	MD15-19	21	-21.54	3.91	3.51	2.86	6.38	25
Vaila Sound	MD15-20	19	-21.6	4.07	4.07	0.81	2.24	26
Vaila Sound	MD15-21	21	-21.75	5.52	2.89	1.16	3.03	27

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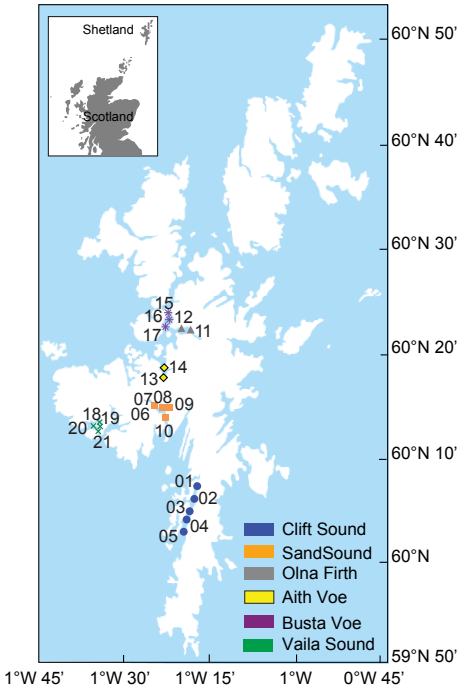

Fig. 1

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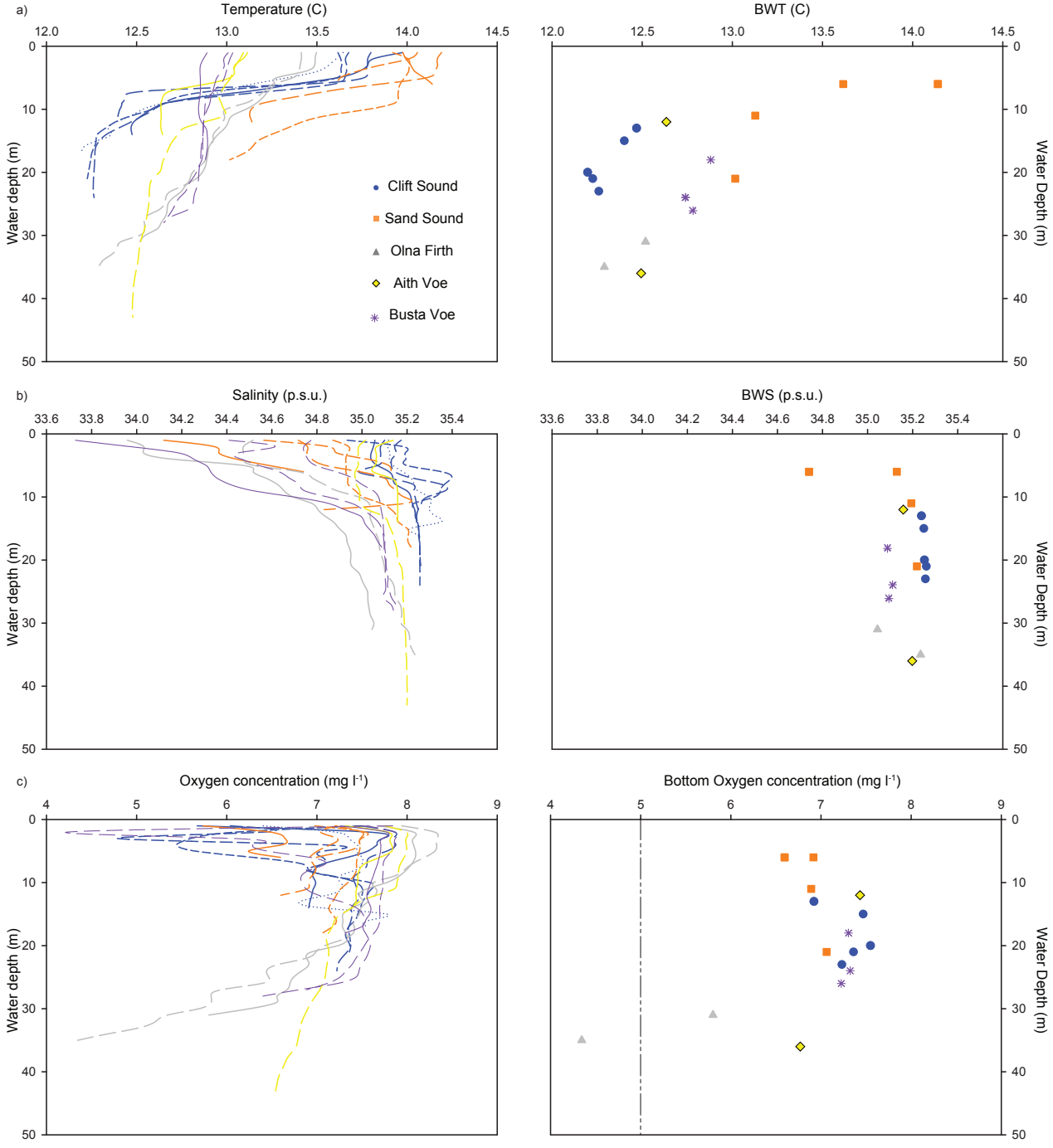

Fig. 2

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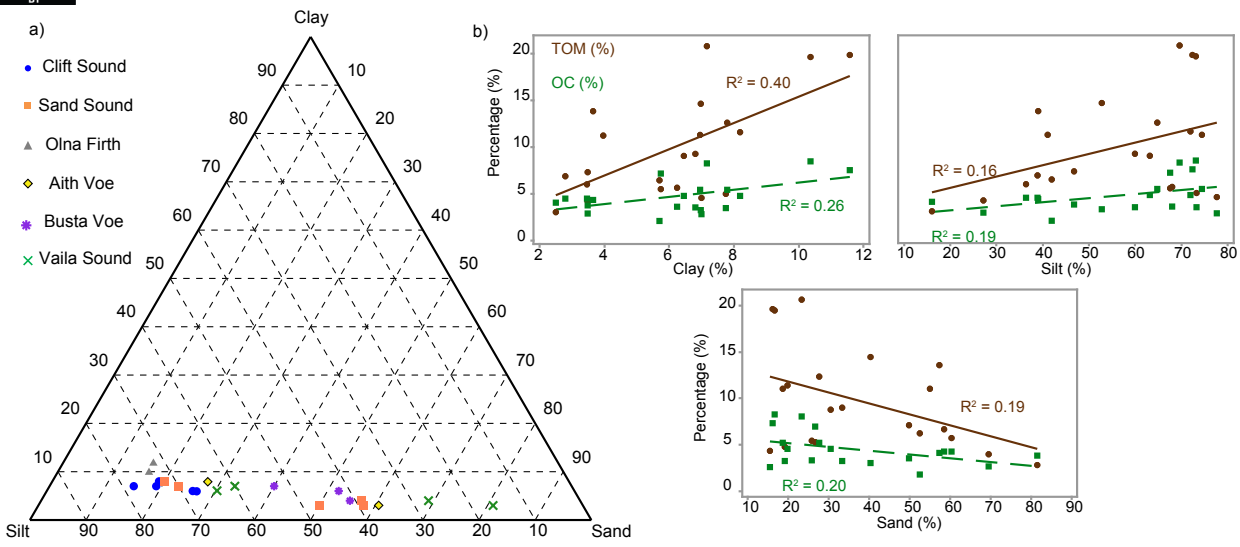

Fig. 3

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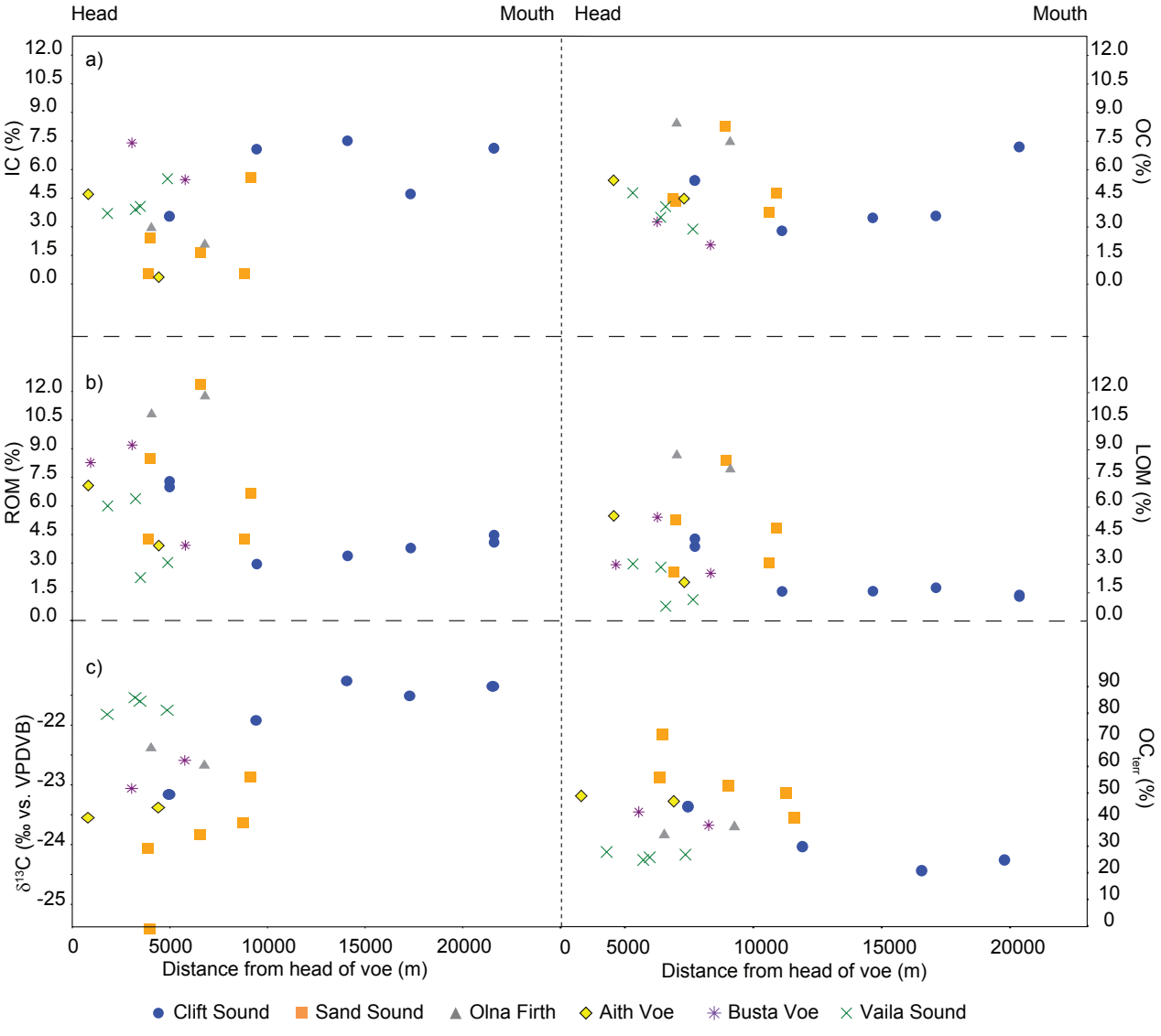

Fig. 4

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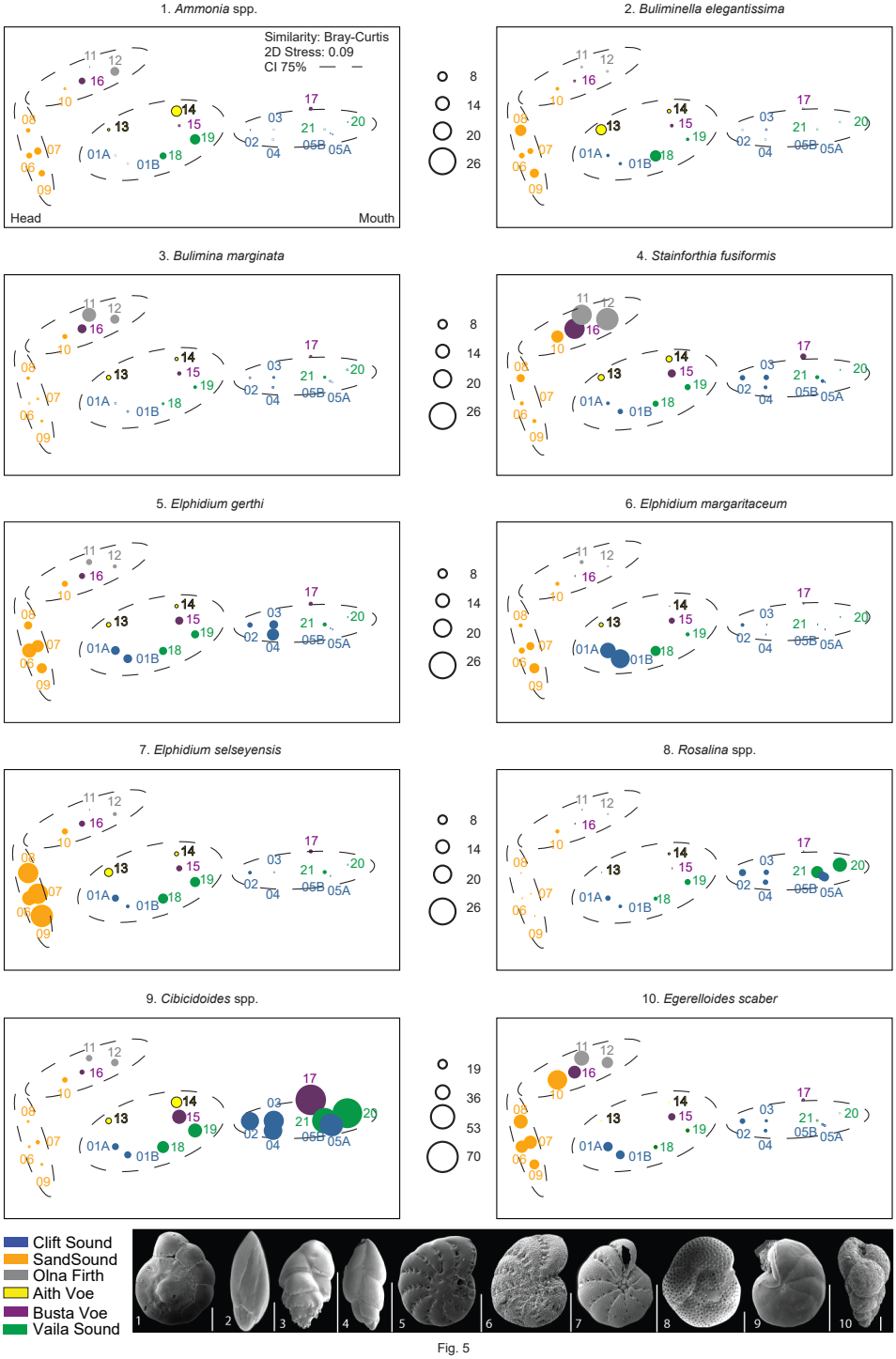

Fig. 5