

Anonymous Referee #3

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This study attempts to reveal benthic foraminiferal responses to organic carbon in fjords system. Previous studies have revealed that benthic foraminifera are useful proxies for various environmental factors, such as oxygen content and food supply in many field. Each voe may have each sediment system because their bottom topography, surrounding environment, and river and current systems are different. These differences control the distributions of grain size, organic carbon, and benthic foraminifera. However, detailed environmental factors of each voe are not described in the manuscript. The authors discuss the “unrestricted” or “restricted” geomorphology, but figure 1 is quite small and detailed characteristics of each voe and sampling points are missing. There is no information about spatial distribution of grain size within each voe in Figure3, so it is difficult to evaluate the distribution. Figures of spatial distributions of grain size, organic carbon, and benthic foraminiferal assemblage can assist in understanding the data interpretation. Grain size is another important parameter controlling benthic foraminiferal distribution in marine environments. However, the authors omit the grain size data from discussion of benthic foraminifera vs. environmental parameters for the reason that grain size distributions don't show obvious trend. Complex factors affect the benthic foraminiferal distributions in shallow marine environments, so I strongly recommend that the authors perform statistical analysis concerning the relationship between benthic foraminifera and environmental parameters (not only carbon but also grain size, water depth, BWS, and DO: :).

Following the referee's comments ,we have included (under the area of study section) a detailed description of available environmental parameters for each voe and modified Fig.1 to better represent the geomorphology of each sea loch (Page 3 lines 17 to page 4 line 18 and Fig. 1, additional info on soil types in Shetland are now illustrated in Suppl. Fig. 1).

We modified Figs. 3 and 4 to better illustrate the spatial distribution of grain size, IC, OC and OM within each voe.

We agree with the referee that grain size is an important parameter, generally co-varying with a number of other variables and therefore linked to foraminiferal distribution in marine environments; however, in our dataset we do not see this simple correlation, most likely because of the complexity of coastal systems compared to open marine settings. For example, *Cibicides* spp. occur in both fine and coarse sediments despite typically being a species that favours coarse substrates for its epilithic mode of attachment. To provide a more thorough representation of the relationship between benthic foraminiferal assemblage distribution and environmental parameters we performed on the full dataset (forams + 10 environmental parameters) canonical correspondence analysis including the % clay as one of the sedimentological parameters (Page 10 line 10 to page 11 line 3 and Figs. 3a and 4).

Introduction Pg 2 Line 24-28: The authors mention the TROX model of Jorissen et al. (1995). This model is mainly applied to the deep sea setting because of its relatively stable environment. As mentioned above, shallow marine environment is affected by various environmental factors, so it is difficult to apply the simple relationship.

Under the light of the referee's comment, we added that TROX models are typically used in deep-sea settings (Page 2 line 24) and that other environmental parameters like salinity and grain size distribution may be more variable and therefore affect the composition of benthic foraminiferal assemblages in shallow marine settings (par 4.3.5). This is further illustrated in Fig. 4.

Pg 3 Line 7 (also for pg 8 Line 31, title of section 4.3, and pg 13 Line 4): The authors use the term “biogeography”, but I think this study in not biogeographical study. “distribution” is more adequate than “biogeography”.

Changed into distribution pg.3 line 7 and recurrence.

Materials and methods Pg 3 Line 17: As mentioned above, the detailed descriptions of each voe should be included in the main body of manuscript.

Done. Page 3 line 17 to page 4 line 18.

Pg 3 Line 21-25: This part is methodology of foraminiferal analysis. I recommend to move this part to section 2.6.

Done. We revised paragraph 2.6 following the referee's recommendation and also the comments of Referee #2.

Pg 3 Line 22-23: Why do you consider that the method of Schönfeld et al. (2012) may lead to underrepresentation of living foraminifera and the method is problematic? The authors explain that the reason for analyzing total (live + dead) assemblage is underrepresentation of living foraminifera in this part. However, the authors mention that the reason 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" in page5, line 23-24.

We understand the referee's confusion and we have rephrased this part of the ms to clarify our points. We do not think that the method of Schoenfeld et al. (2012) may lead to underrepresentation of living foraminifera; what we meant is that scraping the top layer (~ 1 cm thick) of each grab with a domestic spoon may lead to a potential underestimation of living fauna if mixing of the top layer occurred when sampling. In this scenario, the number of living foraminifera at the sediment surface will be "diluted" due to the presence of dead/fossil specimens from deeper sediments. Having said this, a recent study by Rillo et al. (2019) reported that historical sediment samples collected in a way that could have caused disturbance of the sediment surface (sounding and dredge) are still representative of surface conditions and their foraminiferal assemblages can be used to reconstruct environmental changes reliably (Pg. 6 lines 18-29).

The aim of this study is to provide a tool for the interpretation of fossil foraminiferal assemblages and their relationship with OC, therefore using total assemblages over living fauna is a more appropriate tool as total assemblages will better represent downcore fossil fauna found under comparable environmental conditions (revised par. 2.6).

Pg 3 Line 26-27: The authors don't mention BWT. Please describe also about BWT (Fig. 2a).

Done. Pg. 4 line 26.

Pg 4 Line 14: I think the detailed methodology of LOI analysis should be included in the main body of manuscript.

Done. Pg. 5 Lines 13-19.

Pg 6 Line 3: "Fig. 4" should be "Fig. 5".

Changed. Page 9 Line 16.

Results Pg 6 Line 26-27: TC data are not shown.

We changed Fig. 4 and TC data are now shown (Pg. 8 line 5 and Fig. 3d).

Pg 7 Line 5: The authors argue that high OC and low IC at the head of the voes, but I can't find this trend in figure 4a. Please also see comment below (comment to Pg 9 Line 2-6).

We agree with the referee and rephrased this part of the ms (Pg. 8 lines 10-11) and revised Fig. 4 (now Fig. 3). Due to the geomorphology and geometry of the voes, the pattern we observe in the distribution of OC and OM follows a proximity to land/freshwater input trend rather than a seaward gradient (Figs. 3d and 4).

Pg 7 Line 7, 8: The authors use “*Quinqueloculina seminulum*” in supplementary table.
Changed to *seminula* in Supplementary Table 1.

Pg 8 Line 3-4: Please add relative abundance after *Cibicides* spp. and *E. scaber*.
Done. Pg. 9 line 18.

Discussion Pg 9 Line 2-6: The authors argue that a seaward gradient is evident, but I don't agree this argument. First of all, the authors analyze only two stations in Olna Firth and Aith Voe, and three stations in Busta Voe. So, you can't discuss the gradient in these voes. In addition, it seems that samples from Vaila Sound are not collected along the environmental gradient (Fig. 1). Moreover, it seems that there is no obvious seaward gradient in Clift Sound and Sand Sound.

We agree with the referee and rephrased this part of the ms and revised Fig. 4. Due to the geomorphology and geometry of the voes (now better illustrated in Fig. 1), the pattern we observed in the distribution of OC and OM follows a proximity to land/freshwater input trend rather than a seaward gradient. We rephrased the discussions accordingly (par 4.2).

Pg 9 Line 18-22: The authors argue that the high TC content at MD 15-05 is the effect of oil spill. If so, benthic foraminifera may change in response to the effect. Lei et al. (2015_Marine Pollution Bulletin) suggest that *E. scaber* is indicator of oils. Your foraminiferal data don't show the effect of oil or high TC.

We agree with the referee that foram assemblages should reflect the oil spill; it was just a speculation given the surprisingly high levels of OC at this location – not reflected in the *Cibicides* dominated assemblages. We revised this speculation and left an open question regarding what could be driving the high TC content at station MD15-05 (Pg. 11 lines 21-28).

References: Some articles are overlooked in references. Soil Survey of Scotland, 1981 Alve, 1994 Alve, 1995 Alve and Nagy, 1986 Qvale et al., 1984
Fixed.

Figure 1: Please add space between “Sand” and “Sound”. As mentioned above, figure 1 is quite small and detailed characteristics of each voe and sampling points are missing.
Changed. Fig. 1.

Figure 2: Please add legends for each profile (i.e. please add “station number”).
Done. Fig.2.

Figure 3: Please add legends for each plot (i.e. please add “station number”). It is difficult to identify the relationship between data and sampling point. I think figure 3b is not needed because the data are not discussed enough in the manuscript.
We substantially revised Fig. 3 to include the referee's suggestions, also based on previous comments.

Figure 4: Distance of OCTerr of Aith Voe (left one) don't match with OC and LOM. Third plot from the right (Clift Sound) is missing in figure of OCTerr.
We substantially revised Fig. 4 to include the referee's suggestions, also based on previous comments. This info is now in the revised Fig. 3.

Figure 5: What are “Head” and “mouth” in the figure of Ammonia? Do you mean horizontal axis of MDS is gradient from head to mouth? Fig. 5-9 is *Cibicoides*, but

the authors use *Cibicides* in the text. The authors mention that “we grouped under the name *E. excavatum* both forma *selseyense* and forma *clavata*: :”, but “*selseyensis*” is used in figure 5-7. There is no indication of legend unit. Please add the unit (%?).

There are no indication about four circled groups, so please add the name of each group near the dashed circles. It would be better to move SEM figures close to each MDS plot.

We revised Fig. 5 to include the above suggestions; however, it was not possible to include the SEM figure close to each MDS plot due to legibility issues. Foram nomenclature is now consistent throughout the revised manuscript.

Supplementary Material: I can't find supplementary figures, so I can't evaluate supplementary figure 1 and 2.

We apologise for the missing supplementary material. There must have been a problem when we uploaded those files and we were not aware they were missing. We have now included both supplementary figures.

The authors use “Fig. 1a” in supplementary material, but figure 1 is single figure. The authors describe “the island of Vaila” and “the isle of Linga”, but I can't identify the islands because there are no indication of these islands in figure 1.

We revised Fig. 1 to better illustrate the geometry and geomorphology of each voe.

Supplementary Table 1: The authors use *Cibicidoides*, but “*Cibicides*” is used in the text. *Oolina mellow* should be *Oolina melo*. *Trochamina* sp. should be *Trochammina* sp.. There are blanks only in the column of *Reophax fusiformis*. There is “0.0” in the gray cell of *Reophax fusiformis*. Please add picked number of specimens in table 1.

We revised Supplementary Table 1 following the referee's remarks. Upon acceptance of this ms, the full dataset will be made available on PANGEA.

Best regards,

Anonymous Referee #2

Review of the ms "Organic carbon rich sediments: benthic foraminifera as bioindicators of depositional environments" by Elena Lo Giudice Cappelli et al.

The review is based on the version of the manuscript received in April 2019.

The aim of the present study is "To investigate the relationships between sedimentary OC in six west Shetland voes and the associated changes in benthic foraminiferal assemblages..." in order "...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 bioindicators of OC content in coastal sediments and their potential for palaeo-OC reconstruction purposes". This is a very topical theme, an interesting approach, and the manuscript should be of interest to the readers of Biogeosciences.

The manuscript is generally well organized and well written, all figures and tables are necessary, and adequate literature is cited. However, my concern is the weakly described quantitative relationship between the foraminiferal assemblages and the associated geochemical parameters. This relationship is supposed to serve as the baseline for using foraminifera as indicators for OC enrichment (see aims) and, hence, ought to be more clearly addressed. Methodological weaknesses (see examples below) which potentially affect the relationships/correlations should be identified and discussed.

Following the referee's suggestions, we strengthen the discussion regarding the use of benthic foraminifera as indicators of OC content in marine sediments with emphasis on the relationship between forams and geochemical parameters (par 3.2.1 and par. 4.3). We additionally revised the methods and discussed potential weaknesses that could affect the strength of the relationship between forams and OC.

Page 1, lines 13-14: "... evaluate the use of modern benthic foraminifera as bio-indicators of carbon content in six voes (fjords) on the west coast of Shetland." I guess the authors do not mean any kind of carbon? Please specify. The same applies other places in the manuscript.

We added organic before carbon (Pg. 1 line 14 and following instances).

Page 1, lines 14-16: "Benthic foraminifera are sensitive...." Please make it clear to the reader if these statements are based on previous studies or results of the present study.

We rephrased this sentence to make clear that the statement is based on previous studies (Pg. 1 lines 14-16).

Page 3, line 20: "...sub-sampling the top layer of each grab,..." What was the thickness of the "top layer"? How do the authors know if the sampled top layers in the grabs were intact and had not lost some of the fines from the sediment-water interface, i.e. that the samples were comparable? Since no replicates were collected, how do the authors know how representative the OC and OM data were for each site?

The thickness of the top layer is ~ 1 cm (Pg. 4 line 21). We did not have a way of controlling for loss of fine material at the sediment-water interface when collecting grab samples; however, our results compare well with the grain size distribution found in other Scottish sea lochs (Pg. 10 lines 28-31) suggesting that if there were a loss of fines, it was most likely negligible. We did not have replicate samples for OC measurements, but we did have two pairs of replicate samples for OM which resulted in a mean relative error of $\pm 0.07\%$ for LOM, $\pm 0.06\%$ for ROM and $\pm 0.03\%$ for TOM (Pg. 5 lines 17-19), pointing to a very good reproducibility of data and representation of local conditions.

Page 3, lines 22-23: "...foraminiferal counts are 'total' (live + dead) because the sampling technique may lead to underrepresentation of 'live' foraminifera." This needs some explanation.

Following this suggestion and comments of Referee #3, we revised this part of the ms to improve clarity (Par. 2.6).

Page 3, lines 26-27: “An earlier field survey of Shetland voes carried out in August 2009 measured bottom water temperature (BWT), salinity (BWS) and oxygen (O₂) at the same locations as this study (Fig. 2)”. If this implies that the present foraminiferal data collected in 2015 were only compared with 2 hydrographic data from 2009, it should be addressed in the discussion; particularly the statements postulating “low” or “poor” oxygen concentrations in Olua Firth, should be modified throughout the ms.

We agree with the referee in that BWT, BWS and O₂ in 2009 may be different from those of 2015 and we have now discussed this possibility in our ms (Pg. 9 lines 25-27; Pg. 12 lines 27-29; Pg. 13 lines 22-24).

Page 5, lines 14-17: “Both size fractions were analysed. Depending on sample volume, we subdivided each sample into a number of splits using a standard splitter and, when possible, picked at least 300 specimens ...”. The samples were dry-sieved and dry-split? Please clarify. It is not clear why the samples were sieved into two size fractions? How did the authors ensure that the proportion between the two size fractions was the same in the counted splits as in the original sample? This is essential and needs to be explained.

Samples were dry-sieved and dry-split. We added in the methods that samples were split in the two size fractions prior to this study and that we analysed both size fractions to make sure that no environmental information was lost due to changes in biodiversity and/or in the composition of benthic foraminiferal assemblage between the two size fractions (Pg. 6 line 30 to Pg. 7 line 9).

Page 5, line 21: total assemblages (live + dead) were analysed. Please explain how you distinguished *in situ* tests from tests transported into the sites. This is particularly relevant in the more high-energetic environments and deserves some comments.

We had no means to systematically distinguish *in situ* tests from advected ones when counting unstained specimens. What we did was to compare our assemblages with published data from other Scottish locations and fjords to detect possible inconsistencies that may suggest advection of material from other locations, especially in the more energetic environments. Our data overall compare well with other studies, suggesting that advection of tests from other locations is negligible in our sample set. We included this information in Pg. 15 lines 19-21.

Page 5, lines 23-26: “Ten taxa” This belongs to results.
Moved. Pg. 9 Lines 12-15.

Page 6, lines 3-5: = results.
Moved. Pg. 9 Lines 15-16.

Page 6, lines 11-12: “...despite having very different geomorphologies (unrestricted vs. restricted) and circulation patterns (high vs. low energy) (Fig. 3).” This belongs to discussion.
Removed.

Page 7, line 3: “...at sites closed to land ...” close to land
Changed. Pg. 8 Line 13.

Page 7, section 3.1.4: Most of this belongs to discussion. How meaningful is the average stable isotope values of the different lochs? Would you not expect that the average values depend on how many samples are collected and analysed from different parts of the land-sea transect?

Moved to discussion and included range of isotope values and/or number of measurements when possible (Pg. 14 lines 17-20).

Page 8, line 7: "In Vaila Sound, an unrestricted geomorphology (Fig. 1), ..." It is not obvious, based on Fig. 1, that Vaila Sound has an unrestricted geomorphology; please explain, and perhaps modify Fig. 1.

We modified Fig. 1 and this part of the discussion.

Page 8: Section 4.1 may be shortened, particularly since the data are not used in the further discussion. Following the comments of Referee #3 we now included % Clay in our discussion, revised Figs. 3 and 4 accordingly, and moved to the supplement Fig. 3b (now supplementary fig. 2).

Page 9, lines 24-32: These are results.

Moved into a new paragraph in the results (par 3.2.1).

Page 10, lines 2 and 26: I cannot find the Supplementary Fig. 1 and Fig. 2.

We apologise for the missing of supplementary material. There must have been a problem when we uploaded those files as we were not aware they were missing. We have now included both supplementary figures.

Page 10, lines 4-5: "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)." The links between the foraminiferal assemblages and the distribution of OM and OC are neither easily seen from Figs 4 and 5, nor from the descriptions in the following sections. If the authors can show that the statement above actually holds, they should provide some clearer justifications.

Under the light of the referee's comment, we added canonical correspondence analysis of our dataset (foraminifera relative abundance + 10 env parameters: WD, BWT, BWS, O2, IC, OC, LOM, ROM, $\delta^{13}\text{C}$, % Clay) to better constrain the relationship between environmental parameters and benthic foraminifera assemblage distribution and better illustrate the links between forams and OC (par. 3.2.1 and par. 4.3 and Fig. 4)

Page 13, Conclusions, lines 28 and 32: The usefulness of benthic foraminifera as bio-indicators for OC is mentioned in the abstract, in the aims of the study, and in the conclusions but it is not addressed in the discussion. Hence, the importance of foraminifera as bio-indicators for OC in the present study should either be tuned down, or it should be thoroughly addressed in the discussion with concrete, quantitative, examples illustrating how they can be used.

Following the referee's comment, we expanded the discussion about the relationship between forams and OC to better illustrate how benthic foraminiferal assemblages can be used as indicators of past changes in OC deposition and accumulation in marine sediments.

Page 17, Fig. 2 caption. Please add that the CTD-data are from August 2009, whereas the sediment samples for the present study were collected in August 2015.

Done. Pg. 20 Line 11.

To summarize, this is a generally well written manuscript on a timely topic which should be of interest to the readers of Biogeosciences. The figures and tables are all needed and well-presented but some should be adjusted to show the postulated relationships between the benthic foraminiferal assemblages and the associated geochemical data. If possible, it would be helpful for the reader if Fig. 1 is modified so it indicates the difference between unrestricted and restricted geomorphologies of the voes.

We thank the referee for constructive comments and this commendation. We have modified Fig. 1 to better illustrate the geomorphology of each voe and extended the discussion regarding the links between foram assemblages and environmental parameters. Additionally, Suppl. Fig. 1 illustrates type of soils in Shetland and % TOC in soils.

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

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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 organic carbon content in six voes (fjords) on the west coast of Shetland. Benthic foraminifera are known to
10 be ~~are~~ sensitive to changes in organic 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 organic 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)
15 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 organic carbon signals from post-depositional degradation and loss of organic carbon because of their environmental sensitivity and high preservation potential in the sedimentary record.

1 Introduction

25 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 (CO₂) 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

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; n.d.](#))~~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 [in deep sea settings](#) 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 ~~strongly~~[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 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-distribution 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.

10 2 Material and Methods

2.1 Area of study and sampling strategy

Shetland is a subarctic archipelago covering a total area of 1466 km², 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. Clift Sound lies south of Shetland, the islands of Trondra and East Burra to the west, has steep-sides and a relatively unrestricted geomorphology (Fig. 1). Two main types of soil are present in the catchment of Clift Sound: humus-iron podzols cover the islands of East Burra and Trondra, while peaty soils dominate the eastern coast of Clift Sound (Supplementary Fig. 1). Winds are generally from the southwest, can be channelled and result in intensified currents due to the geometry of the sound and the surrounding highlands. Tidal energy can also be channelled into more vigorous currents (Cefas, 2007a).

Sand Sound is somehow T-shaped, located in the southwest of Shetland (Fig. 1) and comprises three areas: the head of the voe, the inner basin and the outer basin (Fig. 1). The head of the voe, almost perpendicular to the inner and outer basins, is a sheltered area that receives freshwater from a significant number of rivers draining the surrounding land. Three main types of soil characterise this area: peaty gleisgleys, peaty podzols and peaty rankers (Supplementary Fig. 1). Here, the voe is very shallow and drying can occur in its eastern and western periphery (Cefas, 2007b, 2008b). In contrast, the rest of the voe is much deeper. The inner basin gently slopes away reaching more than 20 m at its maximum depth; drying can still occur where this basin meets the head of the voe (Cefas, 2007b). The outer basin has steep sides, is 42 m deep at its deepest point and completely open to sea. A shallow sill divides the inner from the outer basins (Cefas, 2007b)

Busta Voe, Olna Firth and Aith Voe are part of a major inlet on the southern coastline of St Magnus Bay on the west coast of Shetland (Fig. 1). Olna Firth represents the eastern branch of the inlet and is roughly oriented east – west, it is the furthest away from the open sea (Fig. 1). A large area of this voe exceed 30 m water depth; the northern coastline gradually slopes into the voe, whereas the south side shows a steeper gradient. Olna Firth is classified as micro-tidal and due to the generally low level of energy in this system, stratification may occur, especially during summer time (Cefas, 2013).

Aith Voe is the southern component of the inlet, with a north – south orientation (Fig. 1). Due to the hilly landscape (up to 100 m) surrounding the voe, Aith Voe is exposed to winds from the north, which can significantly alter local circulation and prevent stratification of the water column. Two types of soil surround Aith voe: peaty and organic soils (Supplementary Fig. 1); few streams drain these soils, with the largest of them discharging on the east coast of Aith Voe (Cefas, 2010).

Busta Voe lies in the northern part of the inlet and is oriented north – south, sheltered and with a maximum water depth of 39 m (Fig. 1). Three main types of soil surround this area: peaty gleisgleys, peaty podzols and peaty rankers (Supplementary Fig. 1); two rivers drain a relatively small catchment area into Busta Voe. Tidal flow is weak in the voe and wind generated currents are predominant; stratification of the water column especially during warm periods can occur (Cefas, 2008a).

Vaila Sound lies on the western coast of Shetland, with the island of Vaila to the southwest and the isle of Linga in the middle (Fig. 1). Three types of soil dominate the catchment of this voe: peaty gleisgleys, peaty podzols and peaty rankers (Supplementary Fig. 1). Most streams discharge in the northern part of the sound; hence, locations in this area might become more affected by terrigenous inputs. Additionally, Linga offers protection from wind and currents, which could facilitate localised accumulation of terrigenous material. Shelter from strong winds is also provided by the island of Vaila to the south. Here the tidal range is small and the associated energy weak; however, Vaila Sound is connected to the Atlantic Ocean west and east of the island of Vaila (Cefas, 2009).~~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 m². Twenty-three surface sediment samples were obtained by scraping the top layer (~ 1 cm thick) 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 (O₂) 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 temperature varies between 14.14 and 12.20 °C being on average 12.74 °C (Fig. 2a). 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 ($O_2 > 5\text{mg l}^{-1}$) with the exception of Olna Firth where lower O_2 were observed (Fig. 2c).

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 $< 2\ \mu\text{m}$, 'Silt' particles between $2\text{-}63\ \mu\text{m}$, and 'Sand' particles $> 63\ \mu\text{m}$ (upper limit of $2000\ \mu\text{m}$). 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). About 1 g of dried sediment was precisely weighted into crucibles of known weight for each surface sample (M_0). These were ashed in a muffle furnace at 250°C for 4 hours, cooled and weighted (M_{250}). Samples were then returned to the furnace at 550°C for 4 hours, cooled and re-weighted (M_{550}). TOM, LOM and ROM were calculated as follow:

$$\text{LOM} = [(M_0 - M_{250}) / M_s] * 100$$

$$\text{ROM} = [(M_{250} - M_{550}) / M_s] * 100$$

$$\text{TOM} = \text{LOM} + \text{ROM}$$

Where M_s is the sample weight minus the crucible weight. Replicate samples were measure at stations MD15-01A and 01B and MD15-05A and 05B which resulted in a mean relative error of $\pm 0.07\%$ for LOM, $\pm 0.06\%$ for ROM and $\pm 0.03\%$ for TOM, pointing to very good data reproducibility and representation of local conditions. 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 ($\delta^{13}\text{C}$) 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 $\delta^{13}\text{C}$ 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 $\delta^{13}\text{C}$ triplicate measurements was 0.07 ‰ and $\delta^{13}\text{C}$ values are reported in standard delta notation relative to Vienna Pee Dee

5 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%.

10 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.

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 $\delta^{13}\text{C}$ 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:

$$\text{OC}_{\text{terr}} = (\delta^{13}\text{C}_{\text{mar}} - \delta^{13}\text{C}_{\text{terr}}) / (\delta^{13}\text{C}_{\text{mar}} - \delta^{13}\text{C}_{\text{sample}})$$

$$\text{OC}_{\text{terr}} + \text{OC}_{\text{mar}} = 1$$

where the end-member value for marine-sourced OC ($\delta^{13}\text{C}_{\text{mar}}$) 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 ($\delta^{13}\text{C}_{\text{terr}}$) 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

25 ~~For foraminiferal analysis, a~~ Aliquots of surface sediments were stained with Rose Bengal to allow the identification of living foraminifera, as detailed in Schönfeld et al. (2012). ~~However, it~~ should be noted that foraminiferal counts are 'total' (live + dead) because 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 (e.g.: Conradsen, 1993). Our priority is to understand how benthic foraminifera respond to prevailing (long-term) environmental conditions, rather than to seasonal variability. Additionally, scraping the top layer (~ 1 cm thick) of each grab with a domestic spoon may lead to a potential underestimation of living fauna if mixing of the top layer occurred during sampling. In this scenario, the number of living

foraminifera at the sediment surface will be “diluted” due to the presence of dead/fossil specimens from deeper sediments. Having said this, a recent study by (Rillo et al., 2019) ~~Rillo et al. (2019)~~ reported that historical sediment samples collected in a way that could have caused disturbance of the sediment surface (sounding and dredge) are still representative of surface conditions and their foraminiferal assemblages can be used to reconstruct environmental changes reliably. ~~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.~~

~~For an early~~As part of an unpublished work study, the Rose Bengal stained surface sediments were wet sieved over a 63 µm mesh. The residues were oven-dried at < 60 °C, then weighed and dry-sieved into 63-150 µm and >150 µm size fractions.

As part of an associated work ~~T~~aiming to understand the influence of different size fractions on the composition of benthic foraminiferal assemblages (Lo Giudice Cappelli et al., in review in *Frontiers*). Both size fractions were analysed independently ~~as part of an associated study work, which and the results~~ 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~~ statistically robust environmental reconstructions ~~(Lo Giudice Cappelli et al., 2019)~~. Similar findings were also reached by Weinkauff and Milker (2018 and references therein). Depending on sample volume, each sample was divided into a number of splits using a standard dry-splitter and, when possible, at least 300 specimens were dry-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 preservation 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 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, canonical component analysis (CCA) 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~~Fig. 4b) with the two pairs of replicate stations exhibiting assemblage reproducibility of 90% (MD15-01A and 01B) and 94% (MD15-05A and -05B).

3 Results

5 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. 3a). 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. 3a). ~~Similarly, s~~Conversely, 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.3a).

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. 34bc). 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. 3c4b).

3.1.3 Carbon content and source in west Shetland bulk sediments

20 The percentage of total carbon (TC) in west Shetland voes varies between 4.31 and 14.32% (Fig. 3d), 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 between 2.0-8.51%, peaking at station MD15-11 (Olna Firth) and reaching a minimum at site MD15-17 (Busta Voe) (Fig. 3d4a; 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. 3d4a). We excluded from the data set site MD15-
25 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-and~~ the reverse occurs where the voes connect to the sea (Fig. 3d4a).

In terms of carbon source, OC_{terr} is generally higher at sites closer to land than in proximity to the open sea (Fig. 3b4e). It reaches a peak of 72% at station MD1-06 in Sand Sound, and a minimum of 21% at station MD15-03 (Fig. 3b4e).

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 higher~~ $\delta^{13}C$ values ~~is evident~~ characterise marine dominated locations ~~in all six within each~~ voes (Figs. 4e 3c and 3d).

10 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. Supplementary Fig. 22~~). 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~~ have relative abundances of > 10 % in at least one of the 21 stations studied: *Ammonia* spp., *Bulimina marginata*, *Buliminella elegantissima*, *Cibicides* spp., *Elphidium gerthi*, *Elphidium margaritaceum*, *Elphidium excavatum*, *Rosalina* spp. and *Stainforthia fusiformis* (Table 2; Fig. 5). Additionally, of all the hyaline species, 20 are 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. Supplementary Fig. 22~~).

~~In general, the most abundant taxa observed in west Shetland's six voes is the hyaline *Cibicides* spp. relative abundance, followed by the agglutinated *E. scaber* relative abundance (Table 2 and Fig. 5).~~

Overall, 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*. These ten dominant taxa were categorised as dominant and imaged with a scanning electron microscope (SEM) at the Scottish Oceans Institute, University of St Andrews (Fig. 5). Altogether, these ten dominant taxa account for more than 70% of the total assemblage at each station (Table 2). In general, the most abundant taxa observed in west Shetland's six voes is the hyaline *Cibicides* spp. relative abundance, followed by the agglutinated *E. scaber* relative abundance (Table 2 and Fig. 5).

3.2.1 Statistical analyses of benthic foraminifera data and environmental parameters

We used cluster analysis of environmental parameters (BWT, BWS, O₂, % Clay, OC, IC, ROM, LOM, δ¹³C and WD) based on Euclidean distance and of benthic foraminifera relative abundance based on the Bray-Curtis index to illustrate similarities between locations (Figs. 4a and 4b), and performed CCA on the combined dataset (benthic foraminifera + environmental parameters) to examine the response of benthic foraminiferal assemblages to environmental gradients (Fig. 4c). Additionally, we run non-metric MDS based on the Bray-Curtis similarity index and foraminifera relative abundance data to analyse for similarities between the populations of the 21 studied stations, independently from environmental forcing (Fig. 5). To note that temperature, salinity and oxygen measurements are from 2009 and may not fully represent the conditions in 2015 when the sediment samples were collected.

Based on both CCA and non-metric MDS, we defined the same four groups with 75% confidence; in the CCA plot axis 1 explains 57 % of total variability and axis 2 the 26 % based on the 10 environmental parameters (Figs. 4c and 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 (Figs. 4c and 5). Station MD15-17 in Busta Voe falls just outside the 75% confidence interval defining Group 4; however, cluster analysis (Fig. 4b) 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 our discussion, we include station MD15-17 in Group 4.

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 owing the influx of Atlantic Ocean waters (Cefas, 2009) appear to control local grain size distribution (Fig. 3a). 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 $\delta^{13}\text{C}$ 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, 2008b), resulting in an overall high percentage of OM and OC, low $\delta^{13}\text{C}$ values (Figs. 3c and 3d4) 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 (Figs. 3c and 3d4). 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, OM and OC content in the sediments (Fig. Supplementary Fig. 3-3b). ~~For these reasons, we have not included these data in the following discussion characterising Shetland's sediments and benthic foraminiferal biogeography.~~

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

A ~~seaward gradient~~clear pattern is evident with regard to the quantity, quality and source of organic matter and carbon measured in the six west Shetland voes (Fig. 34). 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~~close to the open sea (Fig. 4a3d). 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. 34bc) owing its resilience to degradation.

About the source of OM and OC, low $\delta^{13}\text{C}$ 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. 34). 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 $\delta^{13}\text{C}$ 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. 34). In contrast, Clift and Vaila Sounds are characterised by marine sourced material and high IC content in their sediments (Fig. 34) 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. 34).

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. 3d4a) 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. Nevertheless, the benthic foraminiferal assemblage of this location is dominated by *Cibicides* spp. (see par. 4.3 for more details) which ~~and~~ does not reflect the oil spill nor the high OC content, leaving an open question ~~as to~~ regarding what drives OC deposition and accumulation at the mouth of Clift Sound.

4.3 Modern benthic foraminiferal ~~biogeography~~distribution 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 Vailla 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 Vailla Sound (Fig. 5). Station 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).

10 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. 4c; 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 (Fig. 4c) (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. 3d and 4c). Additionally, the high relative abundance of *E. scaber* (26%) at the same locations (Fig. 5) purports a low energy environment, possible water mass stratification and high ROM (Fig. 4c) (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.

20 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 (Fig. 4c), often suggesting stressed environments (Alve, 1994, 2003; Evans et al., 2002; Fontanier et al., 2002; Murray, 1992; Nørgaard-Pedersen et al., 2006) (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) (Figs. 4c and 5). *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. 24c) 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](#) [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. [Despite seawater properties \(T, S and O₂\) were measured in 2009, they seem to be representative of seawater characteristic in 2015 when the sediment samples were collected, as the two sets of measurements \(seawater and sediments\) appear to be in agreement, reflecting long-term of environmental dynamics.](#)

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. [4c](#) 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 ([Fig. 4c](#)) (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. [4c](#)) and the dominance of *Cibicides* (Figs. [4c and 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 O₂ 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 ([Fig. 4c](#)). Similarly, bottom waters are mostly well-oxygenated ([O₂] > 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). [It should be noted that seawater properties reported in Fig. 2 were measured in 2009 and may differ from the actual](#)

conditions in 2015 when sediment samples were collected; nevertheless, our results show that similar dynamics were most likely in place in 2015, as the two sets of measurements compare very well in depicting local environmental dynamics. 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 (Figs. 4c and 5). Only one station in Olna Firth (MD15-11) has poorly oxygenated bottom waters ($O_2 < 5 \text{ mg l}^{-1}$), 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 (Fig. 4c). 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.

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-TC~~ total-TC (Supplementary Fig. 1) compared to other Scottish fjords (Smeaton et al., 2016). On average, the ~~total-TC~~ total-TC content of the six west Shetland voes is 8.8%, whereas ~~total-TC~~ total-TC 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. 34). 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. 4c5), are associated with the preservation of labile organic matter and OC (Figs. 3 and 4c4). Additionally, carbon stable isotope values in Shetland are in range with carbon stable isotope measurements of other Scottish sea lochs: Sunart (average -22.37 ‰, ranging from -24.42 to -21.37 ‰), Teacius (average -24.79 ‰, n=15) (Smeaton and Austin, 2017), Creran (average -21.98 ‰ varying between -24.7 to -21.3 17.3 ‰), Etive (average -25.8 ‰, n=3) (Loh et al., 2008).

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*.

5 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; 10 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 (Figs. 4c and 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* 15 in our samples due to the multiple forcings driving particle size distribution in Shetland (bottom current intensity vs. terrigenous supply) (Supplementary Fig. 1-2); however we do observe a clear correlation between high relative abundance of *Cibicides* and *Rosalina* and low organics and high energy environments (Figs. 4c and 5 and 5). Klitgaard-Kristensen and Sejrup (2002) and Murray (2003) also 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 20 availability of LOM and low oxygen levels. Overall, this compares really well with the assemblage we described in Group 2 (Figs. 4c and 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 25 observed in Oslofjord and Drammensfjord in Norway ((Alve, 1995; Alve and Nagy, 1986) Alve 1995; Alve & Nagy 1986) and Gullmar Fjord in Sweden (Qvale et al., 1984) (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.

30 In our sample set, we had no means to distinguish *in situ* tests from potentially advected tests when picking and counting unstained specimens, and the close match between our benthic foraminiferal assemblages and published data suggests that advection of material from other locations, especially in the more energetic environments, is negligible. Overall, benthic foraminiferal assemblages do respond to variations in the amount of OM and OC becoming deposited in fjord sediments (Fig.

4c), 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 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.

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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.

- 5 *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.

- 40 **Figure 2: CTD profiles from 2009 survey.** a) Temperature profiles and bottom water temperature (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: Sediment physical properties-Particle size distribution. a) Ternary plot Particle size analysis. b) Trends between particle size and organic matter and carbon content Fraction of terrestrially-source OC (OC_{terr}) and marine-source OC (OC_{mar}) at each site. c) Refractory

and labile organic matter content at each site and carbon stable isotope, d) Inorganic and organic carbon percentage at each site and carbon stable isotope.

5 **Figure 4: Statistical analyses-Sediment physical properties.** a) ~~Inorganic and organic carbon percentage at each site~~ Cluster analysis of sediment and seawater physical properties. 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~~ Cluster analysis of benthic foraminiferal assemblages. c) ~~Canonical component analysis of the combined datasets (sediment and seawater + benthic foraminifera relative abundance).~~

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