Dear Editor,
We thank both reviewers for their constructive comments and positive feedback. Both reviewers suggest minor revisions. We have corrected our manuscript following the reviewer's suggestions and address each comment specifically which we document here in detail.

Yours sincerely,
Claire McKay, Jeroen Groeneveld, Helena Filipsson, David Gallego-Torres, Martin Whitehouse, Takashi Toyofuku and Oscar Romero

Reviewer 1: Glock

"One flaw of the manuscript I see is that it's not emphasized in a proper way that *E. exilis* is an infaunal species and that this fact severely affects the Mn/Ca ratios. Usually dissolved Mn2+ should be higher in the pore water at the living depth of the foraminifera than in the bottom water, if the oxygen penetration depth is not very deep. An exception would be if the bottom water already is anoxic. Furthermore, infaunal species are able to migrate within the sediment column and probably experience variable pore water conditions within their lifetime. I don't see this fact as a big disadvantage and would rather use infaunal species for this proxy by myself since they probably incorporate more Mn than epifaunal species. These facts are already introduced in the manuscript but discussed only very sparse and very late (page 18; line 18)."

Response: We agree with this statement
Action: We have now emphasized these points in the introduction section (lines 155-160).

"Another point is a major problem with foraminiferal Mn/Ca ratios itself: Diagenetic overprinting by Mn (oxyhydr)oxide coatings. If I look at your data I do not think you have a problem with these, but not every reader is familiar with details of SIMS analyses and, since in the foraminiferal community Mn/Ca ratios are usually used as tracer for contamination, I think it is worth to be discussed in a bit more detail. Regarding this point there is a mistake in a sentence referring to the paper Glock et al. (2012). At page 8, line 19 you wrote "Therefore, we employed a rigorous pre-treatment cleaning technique to remove possible diagenetic coatings following the method of Glock et al. (2012)." Probably I know these details since I'm the author of the paper: If you used the cleaning method from this paper you did not remove possible diagenetic coatings. In this study I just used oxidative cleaning to get rid of organic contamination. Beforehand I showed that the specimens were generally free from diagenetic oxide coatings with element mapping done by EMP. Please correct this sentence! Furthermore, I am not sure if you used reductive cleaning during your flow through analyses, thus it would be nice to provide some more details about this in the methods section. As I already mentioned I do not think that you have a problem with diagenetic coatings if you chose your SIMS spots within the massive centre of the test walls. Furthermore, I guess if there would be an influence of diagenetic coatings on the foraminiferal bulk analyses the results would be much higher than the SIMS results."

Response: We agree with this statement that not every reader will be familiar with the details of SIMS analyses and acknowledge the mistake.

Action: We have now addressed these points by explaining in more detail how contamination was avoided. and have corrected the mistake regarding organic contamination and stated that the massive centres of the test walls were measured in order to avoid diagenetic coatings (lines 192 – 196 and 253-256). Regarding reductive cleaning, this has now been addressed in lines 278-284.

Page 4, line 3: I would suggest not to write "trace elemental to foraminiferal calcite" ratios and rather write "trace element to calcium ratios". The trace elements are also incorporated into the foraminiferal calcite and thus part of it.

Response: agreed.
Action: amended (line 61).

Page 4, line 14: "You wrote "…. gained more interest is Mn/Ca both as a measure in biogenic foraminiferal calcite and in bulk sediment samples….". As far as I understood you measured Mn/Al ratios in bulk sediment and not Mn/Ca ratios.
Response: True, Mn/Al was analysed, however in the literature Mn alone has also been analysed in sediments (Lenz et al. 2014). It is true that most geochemical data in bulk sediments are presented as Mn/Al, but eventually, core scanner XRF data (being semiquantitative) might be presented as Mn/Ca (maybe even Mn/Ti, but I am not sure right now if this is frequent at all). In any case, both Al and Ca are used as normalization factor, so it makes sense to use Al for bulk sediment normalization and Ca for foram test normalization.
Action: amended so that there is a clear difference between foraminiferal Mn/Ca and bulk (lines 70-72).

Page 5, line 3: "On the other hand, under anoxic conditions the Mn either diffuses upwards and into the overlying water column or when pore waters become supersaturated with respect to Mn, precipitation of MnCO3 (rhodochrosite) occurs (Froelich et al., 1979; Pedersen and Price, 1982; Tribovillard et al., 2006)." I do not understand the argument that under these conditions Mn/Ca should be very low in the forams. Even if all Mn diffuses out of the pore water it is available for the forams during that process. Only within the sediment you probably won't find any accumulation.
Response: We now realise this statement is unclear. We have to assume that Mn abundance affects Mn content in *E. exilis* tests, and this is what we want to highlight. Otherwise, all variations in Mn/Ca would be related to vital effect, and our data does not support this.
Action: We have clarified this by stating the expected Mn/Ca under different redox conditions (lines 91-93).

Page 5 line 15: "Mn/Ca signatures of the ambient bottom water are recorded by benthic foraminifera, for instance, culture experiments have confirmed that the species Ammonia tepida incorporates Mn into the test (Munsel et al., 2010)." I would suggest to reformulate this sentence. Maybe even divide it into two sentences. Furthermore, it might be good to emphasize that "Ammonia tepida incorporates Mn into the test proportional to the concentration in the ambient water masses (Munsel et al., 2010).
Response: agreed
Action: sentence has now been reformulated to clarify our meaning and divided into two sentences to emphasize this point (lines 101-104).

Page 11 line 2: "With cautious positioning of the primary beam on the test walls, such detrital material and potential contaminants were avoided and therefore only the elements actually incorporated into the calcitic tests were measured." How did you do this? I guess it's easy to avoid macroscopic contaminations which are easy to see optically, but a lot of contaminants you won0t see like this. You wrote that you checked the Ca counts and thus assured that you measured within massive calcite which is already good. Do you have any other evidence? Like watching the element distributions during the measurements, for instance (Mn hotspots would probably indicate contaminats), or any pictures which show that you hit the massive centre of the test walls?
Response: Yes we observed the element distributions during measurements and therefore Mn hotspots could be annulled.
Action: these details have now been added to this paragraph (lines 253-256).

Page 14 Line 8: "The slightly higher Mn/Ca determined by FT-ICP-OES in comparison to SIMS derived Mn/Ca perhaps highlights the issue of comparing bulk foraminiferal samples with individual tests comprising of only 6–10 analytical targets. Overall, when a sufficient number (minimum weight of 0.1 mg) of benthic foraminiferal specimens are not available in sediment samples for solution-based analyses (in this case from 35–18 ka), SIMS has the potential to provide reliable results from a few individuals to compensate for this." I just can say it again: This is a great result! You should emphasize it a bit more (maybe in the conclusions), because it0s not trivial that SIMS Mn/Ca ratios on only a few specimens are indeed comparable with foram bulk samples. You won't find this for every element.

Response: agreed.
Action: We have now expanded on this great result and emphasized it in more detail in the conclusion (lines 499-503).

Page 15 line 8: “overlying water column immediately above” I would suggest to remove either “overlying” or “immediately above” because it basically describes the same.
Response: Agreed.
Action: Removed "overlying" (line 371)

Page 15 line 9: “Concurrently, these low Mn/Ca results adhere to the benthic foraminiferal response of a low abundance (ca. 2 specimens cm3) of low oxygen tolerant E. exilis (McKay et al., 2014)” Why concurrently? Both proxies indicate into the same direction (higher oxygen concentrations).
Response: Perhaps "concurrently" was not the best word to use.
Action: This has now been amended to clarify that the Mn/Ca results agree with the faunal data (line 372-373).

Line 15, line 22: “Dissolved Mn available for the precipitation of Mn hydroxides” It would be a bit more correct to speak from Mn (oxyhydr)oxides. Please check the paper to keep at one formulation.
Response: agreed
Action: amended throughout the paper where applicable.

Page 15, line 24: “Based on this increase in sedimentary Mn/Al coinciding with relatively low foraminiferal Mn/Ca, we therefore infer that the Mn/Al enrichment occurred immediately below the oxygenrich pore waters during late H3 and throughout the period 30–25 ka, delimiting the oxygen penetration front and the upward diffusion of Mn.” If I understood right you suggest that the oxygen penetration depth is very deep and thus Mn is precipitating already below the living depth of E. exilis. What is the typical living depth in the sediment for B. exilis? If it doesn’t only follow the oxygen gradient maybe it is even possible to reconstruct the minimum oxygen penetration depth like this or at least to give a rough estimate.
Response: Page 15, line 24: Agreed. *E. exilis* has been shown to live at 1-2 cm sediment depth (Caulle et al., 2014) which might give a rough estimate of oxygen penetration front.
Action: A sentence has now been added to this paragraph for this detail (line 390-392).

Page 16, line 11: “We interpret this greater range in Mn/Ca as a relative decrease in oxygen within the pore water from earlier times within the record” What if you indeed see oxygen fluctuations over the lifetime of the specimens or between different specimens? Could it be that oxygen indeed was highly variable during the LGM at this location? Did you check if there was a trend in Mn/Ca from older chambers until the younger ones?
Response: We agree that the greater range in Mn/Ca could be due to oxygen being more variable. We checked if there were any trends in Mn/Ca from the older to the younger chambers, however we found no trends or systematic shifts in values.
Action: The interpretation of the greater range of Mn/Ca has now been amended (lines 401-404).

Page 16, line 24: During H1, the increase in foraminiferal Mn/Ca; both the greater variability within the individual tests (Fig. 3) and the higher average Mn/Ca per sample depth (Fig. 4) indicate lower oxygen conditions in the pore waters (Fig. 5c)” This sentence is hard to read.
Response: This refers to our two lines of evidence for lower oxygen conditions.
Action: We have reworded this sentence and divided it into two sentences to more clearly express our intended meaning (lines 415-417).

Page 17, line 14: “We interpret this similarity in oxygen conditions as being due to comparable….” Being what? I think you forgot a word in this sentence.
Response: there is no word missing
Action: the tense has now been amended to make this sentence clearer (lines 433-434)

Page 18, line 6: “As MnO2 is rapidly reduced to soluble Mn2+ in hypoxic pore waters (Glock et al., 2012)….” I would suggest to give another reference here….
Response: reference has now been amended (line 452).

Page 18, line 8: “causes low bottom water oxygen concentrations or even anoxia within millimetres of the sediment-water interface, we can expect a high accumulation of redox sensitive trace metals” Different redox sensitive elements react different under variable oxygen concentrations. Vanadium vor example accumulates under anoxic conditions, while Manganese does accumulate under oxic conditions. You cannot generalize “redox sensitive trace metals” within this sentence.
Response: Agreed that this statement is too general as different elements precipitate under different conditions.
Action: This sentence has now been amended and made more specific (Mn/Ca opposed to "redox sensitive elements") (line 454).

Page 19, line 18: “Therefore, whilst high phytodetritus export typically causes low bottom water oxygen concentrations and benthic faunal studies are indicative of such a scenario, redox trace elemental test composition presents a more comprehensive interpretation.” I would suggest to rewrite this sentence.
Response: Agreed
Action: This sentence has been made shorter for better clarification (lines 490-492).

Table 1: Just for clarification: Write 1sd instead of just sd. Is it possible to provide the precision as well?
Response: Agreed.
Action: 1 sd has now been written which is the precision.

Figure 4: The figure is hard to read. Maybe it would be better to present it in horizontal format.
Response: Agreed and I would prefer it in landscape format.
Action: The editor will be informed.

Figure 5: This figure is great and provides all the interpretations of your multiproxy approach in one graphic. Unfortunately it is hard to understand if the reader does not jump between the discussion part and figure. I would suggest to extend the figure caption and to give a short explanation for every time interval and the reason for the interpretation.
Response: We agree with this suggestion.
Action: We have now extended the figure caption to provide a better explanation for each time interval to link with our interpretations.

Reviewer 2: Limburg

“Figure 4 is quite difficult to read”
Response: Agreed
Action: as previously stated, figure 4 should be placed in landscape layout so it can be enlarged on a separate page.

“The author’s findings are consistent with recent work on fish otoliths”
Response: we were aware of these publications but since our work is on infaunal foraminifera, we did not directly compare with fish otoliths or other biota living within the water column.
Action: We have however, cited Limburg et al., (2011) when discussing MnO_2 in hypoxic waters (line 449).

“Perhaps eventually there will emerge a consensus about carbonate-based bio proxies of hypoxia?”
Response: we hope that our data here contributes to such a future consensus.

A comparison of benthic foraminiferal Mn/Ca and sedimentary Mn/Al as proxies of relative bottom water oxygenation in the low latitude NE Atlantic upwelling system

C.L. McKay[1]*, J. Groeneveld[1,2] , H.L. Filipsson[1], D. Gallego-Torres[3,4], M. J. Whitehouse[5], T. Toyofuku[6], O.E. Romero[2]

[1] Department of Geology, Lund University, Sölvegatan 12, 223 62 Lund, Sweden
[2] MARUM – Center for Marine Environmental Sciences, University of Bremen, Klagenfurter Strasse, 28359 Bremen, Germany.
[3] Instituto Andaluz de Ciencias de la Tierra, (CSIC-Universidad de Granada), Avenida de las Palmeras, Armilla, Granada, Spain.
[4] Departmento de Geologia, Universidad de Jaen, Campus Las Lagunillas, Jaen, Spain.
[5] Department of Geosciences, Swedish Museum of Natural History, SE104-05 Stockholm, Sweden.
[6] Japan Agency for Marine-Earth Science and Technology (JAMSTEC), Natsushima-cho 2-15, Yokosuka 237-0061, Japan.

*Correspondence to: C.L. McKay (Claire.McKay@geol.lu.se)

Abstract

Trace element incorporation into foraminiferal shells (tests) is governed by physical and chemical conditions of the surrounding marine environment and therefore foraminiferal geochemistry provides a means of palaeoceanographic reconstructions. With the availability of high spatial resolution instrumentation with high precision, foraminiferal geochemistry has become a major research topic over recent years. However, reconstructions of past bottom water oxygenation using foraminiferal tests remain in their infancy. In this study we explore the potential of using Mn/Ca determined by Secondary Ion Mass Spectrometry (SIMS) as well as by Flow-Through Inductively Coupled Plasma Optical Emission Spectroscopy (FT-ICP-OES) in the benthic foraminiferal species *Eubuliminella exilis* as a proxy for recording changes in bottom water oxygen conditions in the low latitude NE Atlantic upwelling system. Furthermore, we compare the SIMS and FT-ICP-OES results with published Mn sediment bulk measurements from the same sediment core. This is the first time that benthic foraminiferal Mn/Ca is directly compared with Mn bulk measurements, which largely agree on the former oxygen conditions. Samples were selected to include different productivity regimes related to Marine Isotope Stage 3 (35-28 ka), the Last Glacial Maximum (28-19 ka), Heinrich Event 1 (18-15.5 ka), Bølling Allerød (15.5-13.5 ka) and the Younger Dryas (13.5-11.5 ka). Foraminiferal Mn/Ca determined by SIMS and FT-ICP-OES are comparable. Mn/Ca was higher during periods with high primary productivity, such as during the Younger Dryas which indicates low oxygen conditions. This is further supported by the benthic foraminiferal faunal composition. Our results highlight the proxy potential of Mn/Ca in benthic foraminifera from upwelling systems for reconstructing past variations in oxygen conditions of the sea floor environment as well as the need to use it in combination with other proxy records such as faunal assemblage data.

1. Introduction

Foraminifera, being extensively distributed and highly abundant in most marine environments, are essential proxies for reconstructing the chemical and physical properties of past oceans. Several trace element to calcium (Ca) ratios analysed on foraminiferal tests have been developed as proxies in the last decades. Perhaps one of the most conventional approaches is the reconstruction of seawater temperatures using Mg/Ca (e.g. Nürnberg et al., 1996; Elderfield et al., 2006). Other established trace elemental proxies also include Ba/Ca to trace salinity changes due to continental runoff (Lea and Boyle, 1989; Hönisch et al., 2011) and Cd/Ca to reconstruct water masses (Marchitto and Broecker, 2006). Whilst there is a wealth of research applying the geochemistry of foraminiferal calcite for paleoceanographic reconstruction and copious sedimentary redox proxies have been developed (e.g. Gooday et al., 2009), utilising the trace elemental composition of foraminiferal shells (tests) to reconstruct oxygen conditions is still in its infancy. One redox sensitive element that has recently gained more interest is manganese (Mn), both as a trace element in biogenic foraminiferal calcite (Mn/Ca) and in bulk sediment samples (Reichart et al., 2003; Glock et al., 2012, Groeneveld and Filipsson, 2013; Lenz et al., 2014; Koho et al., 2015). Here we aim to explore the potential of Mn/Ca by analysing both benthic foraminiferal tests and comparing it to Mn/Al of bulk sediment samples from an upwelling record to determine if changes in oxygen conditions during different primary productivity regimes are detectable by these methods.

At the sediment-water interface, the concentration of dissolved redox sensitive elements such as Mn vary significantly between oxic and hypoxic (hypoxia defined as <1.42 ml l^{-1} O_2 following Levin et al., 2009) settings. In sea water, redox sensitive Mn is mainly present as Mn^{2+} which under oxic conditions precipitates as Mn oxyhydroxide (Burdige, 1993; Glasby, 2006). The Mn flux across the sediment-water interface is driven by reductive dissolution of reactive Mn oxyhydroxide (Froelich et al., 1979).

Under oxic conditions, dissolved O_2 is present in the pore waters and thus benthic foraminiferal tests are expected to incorporate less Mn into their test. In contrast, under low oxygen conditions, Mn oxyhydroxide is reduced and the Mn^{2+} concentration increases (Tribovillard et al., 2006), becoming available to be incorporated in the foraminiferal tests. Especially under hypoxic conditions, Mn concentrations will be concentrated in the pore water because the Mn cannot escape into the overlying oxic bottom water meaning that benthic foraminiferal Mn/Ca will be highest. Hence foraminiferal Mn/Ca has potential to be used as a proxy of hypoxic conditions.

Accordingly, we expect bulk Mn to be depleted in the sediment and exhibit the opposite trend. On the other hand, under anoxic conditions the Mn is able to diffuse upwards and into the overlying water column or when pore waters become supersaturated with respect to Mn, it is precipitated as $MnCO_3$ (rhodochrosite) (Froelich et al., 1979; Pedersen and Price, 1982; Tribovillard et al., 2006). Therefore, under low oxygen conditions, foraminiferal Mn/Ca is expected to be higher during hypoxic conditions than during anoxic conditions; but still higher than under oxic conditions.

Whilst benthic foraminiferal Mn/Ca has been conventionally used as an indicator of contamination by Mn oxyhydroxide or Mn carbonate (Boyle, 1983; Barker et al., 2003) new studies are pioneering Mn/Ca as a potential proxy of related changes in bottom/pore water oxygen and redox conditions (Ní Fhlaithearta et al., 2010; Glock et al., 2012; Groeneveld and Filipsson, 2013; Koho et al., 2015). Mn/Ca signatures of the ambient bottom water are recorded by benthic foraminifera. For example, culture experiments have confirmed that the species *Ammonia tepida* incorporates Mn into the test proportional to the concentration in the ambient water masses (Munsel et al., 2010). Thus, during benthic foraminiferal calcification under hypoxic conditions, more Mn will be assimilated into their calcite tests, whereas under anoxic conditions or oxic conditions in particular, Mn is expected to decrease, albeit to different amounts (Pena et al., 2005). Hence Mn/Ca used in this study could provide a reliable means of reconstructing the former seafloor oxygen settings at the time of deposition opposed to sediment bulk measurements which can continue to oxidise and be mobilised post deposition.

Traditionally, trace element/Ca is analysed on solution based samples containing larger numbers of foraminifera specimens to give a representative result (Groeneveld and Filipsson, 2013). When a sufficient amount of specimens is not available for solution based techniques, or if diagenesis has affected the tests, a micro-analytical technique upon single specimens such as Secondary Ion Mass Spectrometry (SIMS) is a valuable tool. From an analytical perspective SIMS has enhanced our ability to determine how trace elements are distributed within foraminiferal tests at high spatial resolution and precision on individual foraminifera (Allison and Austin, 2003; Bice et al., 2005; Kunioka et al., 2006; Glock et al., 2012). Recently, SIMS determined Mn/Ca of benthic foraminifera has been found to be representative of Mn/Ca in the top cm of the pore water, confirming that the foraminiferal calcite composition relates to the level of oxygen depletion (Glock et al., 2012).

Where a sufficient amount of foraminiferal specimens are available, we additionally used Flow-Through Inductively Coupled Plasma Optical Emission Spectroscopy (FT-ICP-OES; Haley and Klinkhammer, 2002). Flow-through analysis is a means of determining elemental composition from samples of foraminiferal tests which permits complete monitoring of the effects of cleaning and dissolution (Haley and Klinkhammer, 2002). However, due to the small size of *Eubuliminella exilis*, a larger number (up to 50 specimens in this case) of foraminiferal tests from the same core sample is required to give a representative average signal. Therefore we utilise both SIMS and FT-ICP-OES to explore the potential of Mn/Ca for interpreting down core oxygen studies.

To explore how Mn/Ca works as a potential proxy for bottom/pore water oxygen conditions, we study a site from the low latitude NE Atlantic Upwelling System. Upwelling systems are an ideal environment to test this proxy, as they are renowned for high export rates of labile organic matter from surface waters which provokes severe oxygen depletion in the underlying intermediate waters and at the seafloor (Böning et al., 2004). We study core GeoB7926-2 from the upwelling region off coastal NW Africa (Fig. 1) and compare foraminiferal and sedimentary Mn with published diatom and benthic foraminiferal species composition (Romero et al., 2008; Filipsson et al., 2011; Kim et al., 2012; McKay et al., 2014). In general, coastal upwelling systems are the most productive of the world ocean resulting in vulnerability to oxygen minima within the water column and underlying seafloor (Helly and Levin, 2004; Bakun et al., 2010). At present, the benthic environment of this particular upwelling system is not especially susceptible to low oxygen conditions and is well ventilated with bottom water oxygen of ca. 5 ml l^{-1} (Goretski and Koltermann, 2004). However, based on benthic foraminiferal faunal studies, there is evidence of previous periods of oxygen depletion at the sea floor during the Younger Dryas (YD, 13.5-11.5 ka) and Heinrich Event1 (H1, 18-15.5 ka) in particular. This is inferred from the predominance of the low oxygen tolerant benthic foraminiferal species *Eubuliminella exilis* (synonymised taxa: *Bulimina exilis*) (Filipsson et al., 2011; McKay et al., 2014). Therefore, we selected samples allocated to late Marine Isotope Stage 3 (MIS3, 35-27 ka), the Last Glacial Maximum (LGM), Heinrich Event 1 (H1), Bølling Allerød (BA) and the Younger Dryas (YD) to reconstruct past bottom water oxygen. These climatic intervals were chosen in order to test if Mn/Ca can confirm the low oxygen conditions during different productivity regimes as reported by previous studies from this particular sediment core (Filipsson et al., 2011; McKay et al., 2014). We focus on utilising SIMS and compare this method with FT-ICP-OES where a sufficient number of *E.*

153 *exilis* specimens were available. We also present Mn/Al sediment bulk measurements from the
154 same sediment for further comparison.

155 The low oxygen tolerant, benthic foraminiferal species *Eubuliminella exilis* is an infaunal species
156 and therefore has the ability to migrate within the sediment and experience variable pore water
157 conditions. This migration could affect the Mn/Ca incorporated within the test; however as *E.*
158 *exilis* is so low oxygen tolerant, it possibly migrates less than other infaunal species which are
159 not as tolerant. Therefore *E. exilis* likely incorporates more Mn and is therefore the right
160 recorder, suitable for reconstructing oxygen levels. Furthermore, abundant populations of this
161 taxon are reported from a range of settings, including coastal upwelling sites, sapropels, oxygen
162 minimum zones and other environments for example in the Bering Sea and the Mississippi River
163 mouth (Caralp, 1989; Jorissen, 1999; Jannink et al., 1998; Rasmussen et al., 2002; Khusid et al.,
164 2006). Thus *E. exilis* also has potential to be a useful proxy for other marine environments
165 susceptible to strong oxygen deficiency and high fluxes of organic export. *Eubuliminella exilis*
166 has been found to correlate with diatom accumulation rate and clearly dominates the
167 foraminiferal fauna during very high diatom input (Caralp, 1984; Filipsson et al., 2011; McKay
168 et al., 2014; McKay et al., in revision). Therefore *E. exilis*, being present (albeit in considerably
169 varying abundance) throughout the GeoB7926-2 record is an ideal candidate to record oxygen
170 changes in the environment in which they lived and also provide an opportunity to test if export
171 productivity is causing low oxygen conditions opposed to the dominance of this species merely
172 being a fresh phytodetritus diet signal (Caralp, 1989).

173 We hypothesise that higher foraminiferal Mn/Ca will occur during times of high diatom
174 accumulation rate and lower bottom water oxygen concentrations and accordingly higher *E.*
175 *exilis* abundance, with the opposite effect during times of low surface productivity.

176

177 ## 2. Method

178 Gravity core GeoB7926-2 from the NE Atlantic Upwelling System (20°13'N, 18°27'E, 2500 m
179 water depth) was recovered during R/V Meteor cruise M53/1 (Meggers and Cruise Participants,
180 2003). The age model for the core was published by Kim et al., (2012) and the timing and
181 duration of the climatic intervals were adopted from the $\delta^{18}O$ of NGRIP (NGRIP Members,
182 2004) and Sánchez Goñi and Harrison (2010). We selected well-preserved foraminiferal
183 specimens based on the criteria of high and low surface productivity regimes as demonstrated

from diatom abundance (Romero et al., 2008). Details of sample preparation for benthic foraminiferal faunal analysis have previously been published (Filipsson et al., 2011; McKay et al., 2014).

2.1 SIMS analysis

From the >150 µm size fraction, a total of 48 specimens of the benthic foraminifera species *Eubuliminella exilis* were hand-picked under a binocular microscope for SIMS analysis (Table 1). We acknowledge that the presence of Mn-rich authigenic coatings (e.g. Mn (oxyhydr)oxides and organic matter) can be problematic for trace elemental analysis of foraminifera (Boyle, 1983; Pena et al., 2005; Klinkhammer et al., 2009). Therefore, we employed a rigorous pre-treatment cleaning technique to remove possible organic contamination following the method of Glock et al., (2012) and avoided potential diagenetic coatings during the SIMS analysis by measuring within the massive centre of the test walls (Fig. 2).

For the SIMS analysis, foraminifera from individual sample depths were rinsed over a 63 µm sieve with milliQ water. After this rinsing step, the foraminifera were transferred into vials and sonicated for 20 seconds. Subsequently, the foraminifera were rinsed with methanol and sonicated again for 1 minute. Any residual methanol was then removed with milliQ water. An oxidative cleaning step was performed to remove organic matter which consisted of mixing the following reagents: 100 µl 30% H_2O_2 to 10 ml of 0.1 M NaOH solution. 350 µl of this reagent was added to each individual vial and the vials were put into a water bath at 92ºC for 20 minutes. Afterwards another 20 second sonic bath was undertaken; the foraminifera samples were rinsed again with milliQ water in the 63 µm sieve to remove any residues. For the final step, the specimens were transferred back into their respective vials and 250 µl of 0.001 M HNO_3 was added to each vial. The vials were put into a sonic bath for 20 seconds and finally rinsed one last time with milliQ water. After the cleaning procedure, the specimens were checked under a binocular microscope to ensure sufficient cleaning and that the tests remained intact.

Foraminifera specimens were embedded in low viscosity epoxy resin at JAMSTEC, Japan. The foraminifera were then ground to expose a cross-section across the test wall using 16 µm silicon carbide paper at the Department of Geosciences, University of Edinburgh, UK. Resin pieces were mounted into low viscosity epoxy resin disks (Struers) at the NORDSIM laboratory, Laboratory for Isotope Geology at the Swedish Museum of Natural history, Stockholm, Sweden.

The mounts were polished using a Struers Rotopol-2 at 150 rpm for 1 minute, first with 3 µm diamond suspension and again with 1 µm diamond suspension. Between each grinding and polishing step, mounts were cleaned with ethanol. Each cross-sectioned foraminifera test was examined under high power reflected light microscopy to evaluate the quality of the carbonate and to assist in assessing the progress of polishing until the cross sections were clear. Subsequently, the mounts were cleaned in high purity ethanol and coated in a 20 nm thick high purity Au coat.

The reference material used for the SIMS was a polished piece of OKA calcite crystal supplied from Geomar, Kiel University, Germany (E. Hathorne, pers. comm). This standard was obtained from a matrix matched specimen for which Mn/Ca has been reported by solution ICP-MS (Glock et al., 2012). During calibration, the OKA was analysed n = 16 times, yielding a high sensitivity with 1 standard deviation repeatability of 1.2% for Mn concentration.

The Mn/Ca analyses of the test cross-sections were performed using a Cameca IMS 1280 ion microprobe at the NORDSIM laboratory at the Swedish Museum of Natural history, Stockholm, Sweden. Analysis used a $^{16}O_2^-$ ion beam accelerated at 23 kV impact energy (-13 kV primary beam, +10 kV secondary beam). It is vital to only analyse Mn which is located internally within the original test wall to attain most representative Mn/Ca for developing it as a redox proxy. Therefore, a 50 µm aperture in the primary column was used to shape a slightly elliptical 5 µm spot on the sample surface, which, together with careful placement, reduced the effects of sample contaminiation from the test wall outer surface. Prior to each analysis, the analytical location was pre-sputtered for 2 minutes with the ion beam rastered over 10 x 10 µm raster to remove the Au coat and any remaining surface contamination. During the initial pre-sputtering, the ^{44}Ca distribution was monitored using the ion imaging system of the instrument and maximised to ensure high precision beam targeting on the fine foraminiferal test walls. The mass spectrometer was operated at a mass resolution of $M/\Delta M \sim 6000$ to resolve the ^{55}Mn peak from nearby molecular interferences. A 400 µm contrast aperture was employed for maximum transmission together with a 60 µm entrance slit, a 2001 µm field aperture restricting the field of view on the sample to an area of ca. 12 x 12 µm at the transfer magnification of ca. 160x, and a 45 eV wide energy window; all of which combined to yield adequately flat topped peaks on the species of interest. Each analysis comprised of 16 cycles of ^{44}Ca (1s integration/cycle) and ^{55}Mn (2s). Each analysis lasted approximately 9 minutes. Secondary ions were measured using a low noise (<0.01 cps) ion counting electron multiplier. Multiple analysis points were undertaken upon each individual test of *E. exilis* (ca. 6-10 targets per individual specimen) starting from the aperture

and taking measurements alternating between the outer wall and internal walls. For the best targets, programming was performed manually to ensure that widest chamber walls and "t" junctions were targeted since they provide a wider test wall for the analyses (Fig. 2). Furthermore, at such high spatial resolution and precision, it is easy to visually observe and avoid encrusting prior to selecting analysis targets via the connected screen and avoid measuring secondary calcite or authigenic clays which would otherwise affect measurements. With cautious positioning of the primary beam on the test walls and observations of the element distributions during measurements, such detrital material and potential contaminants were avoided and annulled. Therefore only the elements actually incorporated into the calcitic tests were measured. As an additional prerequisite to this, analyses with Ca values > 500 kcps were classified as being reliable. Mn/Ca was first normalised to those determined in the OKA standard and subsequently converted to the true value in the OKA based on the Glock et al. (2012) value for Mn/Ca of 4920 µmol mol^{-1}.

The advantage of SIMS is that it is non-destructive and as the foraminiferal cross sections are preserved within the mounts, they can be stored for further analyses. Mounts are archived at the NORDSIM laboratory.

2.2 FT-ICP-OES analysis

For FT-ICP-OES, 20-50 specimens per sample depth of *E. exilis* from the GeoB7926-2 record were selected from samples corresponding to H1, BA and the YD for comparisons with the SIMS data. These three climatic intervals encompassed the only samples where a sufficient number of pristine *E. exilis* individuals were present. The tests were gently crushed in a 0.5 ml vial and fragments were transferred into a PTFA filter with 0.45 µm mesh.

For analysis, the filters were connected to a Flow-Through – Automated Cleaning Device (Klinkhammer et al., 2004; Haarman et al., 2011). Automatic cleaning prevents the loss of material which occurs with traditional cleaning allowing the analysis of very small samples (~20 µg). The Flow-Through was then connected to an ICP-OES (Agilent Technologies, 700 Series with autosampler (ASX-520 Cetac) and micro-nebulizer). Time Resolved Analysis (TRA) was used to analyze the samples at MARUM, University of Bremen, Germany. After an initial rinse (5 minutes) with buffered Seralpur water to remove clays, the samples were slowly dissolved using an acid ramp formed by mixing of Seralpur with 0.3 M QD HNO_3 (no additional oxidative

and/or reductive cleaning was performed). Starting with pure Seralpur the acid contribution was stepwise increased every minute to 100% acid after 30 minutes. The flow speed of the solution was 250 µl/l. Mn/Ca of the foraminiferal calcite was determined by identifying the TRA interval which showed a consistent linear relationship between Mn and Ca counts. Potential diagenetic phases like Mn(oxy)hydroxides are avoided this way, as they would have a different slope, i.e. a significant Mn-signal along with the absence of a Ca- signal. Mn/Ca was then calibrated using the characteristic slope of this linear relationship of a known in-house standard solution analyzed on the same acid ramp. The average standard error on the determination of the slope for Mn/Ca was 0.75%. We analysed an international limestone standard (ECRM752-1) for Mg/Ca to validate the results following the same FT-protocol. The average Mg/Ca of the ECRM752-1 (n = 4) was 3.76 mmol mol^{-1}, which compares well with average published value of 3.75 mmol mol^{-1} (Greaves et al., 2008).

2.3 Mn bulk measurements

For geochemical bulk analyses, samples were dried and ground in an agate mortar and homogenised. Total dissolution of samples was undertaken using HF and HNO_3 following the standard procedures of Gallego-Torres et al., (2007). Mn and Al content for samples corresponding to 35-10 ka was determined by Atomic Absorption Spectrometry, using Re and Rh as internal standards at the Analytical Facilities at the University of Granada, Spain. Mn being redox sensitive was normalised to Al content in order to correct for detrital variations (Van Der Weijden, 2002). Mn/Al data corresponding to 25-10 ka has previously been published (Gallego-Torres et al., 2014) and here we extend the Mn/Al record to 35 ka.

3. Results

3.1 SIMS and FT-ICP-OES data

Overall, Mn/Ca determined by SIMS varied between 2 and 750 µmol mol^{-1}. Average values ranged from 6.5 to 260 µmol mol^{-1} throughout the record and displayed a decreasing trend down core (Fig. 3-4, Table 1).

Mn/Ca was lowest in foraminiferal tests from Marine Isotope Stage 3 (MIS3) and the Last Glacial Maximum (LGM) with values ranging from 25-68 µmol mol^{-1} (average 50 µmol mol^{-1})

and 2-225 µmol mol^{-1} (average 70 µmol mol^{-1}) respectively. Foraminifera from samples derived from the Heinrich Event 1 (H1) and the Bølling Allerød (BA) exhibited a slightly elevated range of Mn/Ca at 50-380 µmol mol^{-1} (average 117 µmol mol^{-1}) and 27 to 280 µmol mol^{-1} (average 133 µmol mol^{-1}) respectively. Highest Mn/Ca occurred in foraminiferal tests from the Younger Dryas (YD) (average 175 µmol mol^{-1}) period ranging from 23 to 750 µmol mol^{-1} (Fig. 3, Table 1).

In order to determine statistically significant differences between samples from different productivity regimes, Kruskal-Wallis tests were run and showed a statistically significant difference between the mean Mn/Ca values (per individual foraminifera specimen) between all five climatic intervals ($p = 0.003$). By testing the mean Mn/Ca of each foraminifera between each climatic interval individually in turn, using Post hoc (Mann Whitney U) tests, significant differences lay between climatic intervals with high and low export productivity regimes (based on diatom accumulation rate). Namely, significant differences in Mn/Ca were evident between the YD interval and the LGM and MIS3 (Table 2).

Mean SIMS derived Mn/Ca per sample depth compare well with FT-ICP-OES results (Table 1), agreeing with maximum differences of 15 - 24 µmol mol^{-1} (Fig. 4). Mn/Ca from the FT-ICP-OES on bulk foraminiferal samples tended to be slightly higher compared to the mean ratios obtained from the SIMS microanalyses for the same sediment depth (for example 140 µmol mol^{-1} compared to 116 µmol mol^{-1} during the YD).

3.2 Mn bulk data

Bulk sedimentary Mn/Al showed highest values during MIS3 at 30-24 ka as well as during 19-17.5 ka and the YD. The YD was characterised by a sharp Mn/Al increase at 12.3 ka, coinciding with maximum diatom productivity (Fig. 4). Relatively low Mn values occurred during 35-32 ka and during the LGM Mn was below the level of detection (<0.06%). Between 32 ka and the onset of the LGM, a progressive increased was observed.

4. Discussion

4.1.1 Comparison of analytical methods

Our results indicate that Mn/Ca in benthic foraminifera might prove to be a valuable proxy for oxygen in the bottom and pore waters. The downcore variability in foraminiferal Mn/Ca at site GeoB7926-2 displays a consistent agreement between the mean SIMS determined Mn/Ca of each sample depth and the bulk foraminifera Mn/Ca measured by FT-ICP-OES. In general, the agreement in values suggests that the SIMS determined Mn/Ca is likely to be a true signal within our reconstruction. The slightly higher Mn/Ca determined by FT-ICP-OES in comparison to SIMS derived Mn/Ca highlights the issue of comparing bulk foraminiferal samples with individual tests comprising of only 6-10 analytical targets. Overall, when a sufficient number (minimum weight of 0.1 mg) of benthic foraminiferal specimens are not available in sediment samples for solution-based analyses (in this case from 35-18 ka), SIMS has the potential to provide reliable results from a few individuals to compensate for this.

Recent culturing experiments on benthic foraminifera demonstrate that calcification can occur even under anoxic conditions (Nardelli et al., 2014). This is key for the discussion of trace elemental data derived from the foraminiferal tests, as not only does the timing of the calcification determine the geochemical signature, it also shows that the signature is recorded in a wide range of oxygen conditions.

When comparing foraminiferal Mn/Ca to previously published sediment measurements of Mn/Al (Fig. 4) for site GeoB7926-2 (Gallego-Torres et al., 2014), in general we do not find a continuous relationship in trends throughout the record, but overall they largely agree on the former oxygen conditions. This is most likely due to diagenetic processes and migration of redox fronts through the sediment which redistributes the bulk Mn after deposition, whereas the foraminiferal tests record the Mn concentration at the time of calcification. In fact, bulk sediment Mn concentrations are often interpreted as being related to diagenetic (post-production) oxidation fronts and less often to the syn-sedimentary environment (e.g. Thomson et al., 1995; de Lange et al., 2008). Thus, two different processes govern Mn fixation in sediment and foraminifera tests respectively and consequently we can expect an offset between the two signals.

4.2 Reconstruction of bottom water oxygen conditions: Late MIS3 to the Younger Dryas (35-11.5 ka)

4.2.1 Late MIS3 - late LGM (35 - 19 ka)

Foraminiferal Mn/Ca derived from SIMS measurements were comparably low and exhibited less variability within individual tests (Fig. 3) during episodes of low diatom export between 35 and 19 ka. The lower Mn/Ca indicates that the pore waters and water column immediately above were oxygenated (Fig. 5a). These low Mn/Ca results are in agreement with the benthic foraminiferal response of a low abundance (ca. 2 specimens cm^{-3}) of low oxygen tolerant *E. exilis* (McKay et al., 2014) to the extent that not enough well preserved specimens were available for FT-ICP-OES analysis. Such low Mn/Ca and lack of low oxygen dwellers is to be expected since benthic foraminifera living in an environment where productivity export does not fluctuate at great magnitudes are potentially less exposed to a wide range of oxygen conditions and according Mn concentrations in the pore waters during their growth. This stable and relatively low export productivity is not only exhibited by diatom accumulation but also carbonate ($CaCO_3$ = 30-45%) during late MIS3 to late LGM (Romero et al., 2008; McKay et al., 2014).

However, by late Heinrich Event 3 (H3: 32.5-29 ka), whilst foraminiferal Mn/Ca is low, there is an increase in bulk sediment Mn/Al, suggesting penetration of oxygen-rich bottom waters within the upper centimetres of the sediment column. Dissolved Mn available for the precipitation of Mn (oxyhydr)oxides would most likely be sourced by diffusion from underlying sediments where anoxic conditions prevail (Burdige, 1983). Based on this increase in sedimentary Mn/Al coinciding with relatively low foraminiferal Mn/Ca, we therefore infer that the Mn/Al enrichment occurred immediately below the oxygen-rich pore waters during late H3 and throughout the period 30-25 ka, delimiting the oxygen penetration front and the upward diffusion of Mn (Fig. 5a). Therefore we suggest that the oxygen penetration depth is deeper, and thus the precipitation of Mn too, than the living depth of *E. exilis*, which has been documented to live at 1-2 cm sediment depth (Caulle et al., 2014). As both foraminiferal and sedimentary Mn results correspond to low relative abundances (<5%) of the low oxygen indicator *E. exilis* and low diatom input (Romero et al., 2008; McKay et al., 2014), this reinforces our interpretation of more oxygenated conditions at the sea floor during H3. It associates with the scenario that during low primary productivity export, oxygen levels are not lowered by the decomposition of large amounts of fresh phytodetritus.

During the LGM, Mn/Ca show a greater range between specimens (three specimens exhibit consistent Mn/Ca in the order of 4 - 16 µmol mol^{-1} whilst others suggest a greater intra-test variability of 16 – 230 µmol mol^{-1}) than samples from MIS3 (Fig. 3) despite diatom input being relatively low and stable during this interval. We interpret the greater range in Mn/Ca as more

variable oxygen levels and a relative decrease within the pore water overall, in comparison to earlier times within the record (Fig. 5b), but not to the extent of hypoxia since *E. exilis* abundance remains low whilst species diversity is high (McKay et al., 2014). Simultaneously, Mn/Al remains low (Mn content <0.06%) which could relate to bottom water masses since Mn can be transported to deep waters via scavenging (Glasby, 2006). In particular, Gallego-Torres et al., (2014) suggest that site GeoB7926-2 was under the influence of Antarctic Bottom Water (AABW) during the LGM. AABW contains lower Mn concentrations relative to North Atlantic water masses (Statham et al., 1998; Idrus, 2013). Mn is scavenged from AABW as it flows north and thus when reaching site GeoB7926-2 is Mn-impoverished. However, we propose that low Mn fixation in the sediment during the LGM is due to low export production leading to relatively more Mn being located in the water column.

4.2.2 Heinrich Event 1 – Younger Dryas (18 - 11.5 ka)

During H1, the increase in foraminiferal Mn/Ca indicates lower oxygen conditions in the pore waters (Fig. 5c). Lower oxygen conditions are evidenced in terms of both the greater variability within the individual tests (Fig. 3) and the higher average Mn/Ca per sample depth (Fig. 4). The comparably lower oxygen conditions are consistent with our hypothesis which stemmed from the benthic foraminiferal faunal assemblage composition (Filipsson et al., 2011; McKay et al., 2014) whereby the dominance of *E. exilis* indicated low oxygen conditions at the seafloor as a result of high primary productivity and export flux.

Bulk Mn/Al is moderately low (below average of 3.4×10^{-2} at the corresponding sample depth of the benthic foraminifera analysed by SIMS) when compared to the rest of the record, despite high diatom accumulation rate. The combined interpretation of Mn, Mo and U to Al ratios presented in Gallego-Torres et al., (2014) evidenced that the H1 period experienced suboxic (<0.1 ml l^{-1} O_2 as defined by Morrison et al., 1998) to anoxic bottom water conditions. Under these conditions, the redoxcline would have been very shallow or even located at the sediment-water interface, so that Mn was not preferentially fixed in the sediment. Furthermore, the sedimentary layer might have acted as a source of reduced dissolved Mn to the water column and/or the uppermost pore waters, allowing for higher Mn availability for incorporation into benthic foraminiferal tests.

During the BA, foraminiferal Mn/Ca determined by SIMS exhibit a similar range of values and intra-test variability as during H1 (Fig. 3). We relate this similarity in oxygen conditions to comparable diatom accumulation rates during both climatic intervals. Previously, from the relative decrease in *E. exilis* abundance during the BA, the bottom water oxygen concentrations were interpreted to have increased in comparison to H1 and the YD (Filipsson et al., 2011). Despite this benthic faunal response to productivity export, the diatom input itself was relatively higher at the core depth sampled for SIMS analysis within the BA than the one in H1. Subsequently, our SIMS derived Mn/Ca results do not suggest vast redox shifts between the two climatic intervals generally the values follow the diatom input. This emphasizes that whilst the benthic foraminiferal community composition is a reliable indicator of past environmental conditions, the trace elemental composition of foraminiferal tests has potential to reveal a more detailed palaeoenvironmental interpretation. Furthermore, the foraminiferal Mn/Ca coincides with the lack of Mn enrichment in the bulk sediment and Mn/Al does not substantially differ from the previous climatic interval either. Mo and U suggest prevailing suboxic conditions during the BA (Gallego-Torres et al., 2014) and further, we infer that the redoxcline would be shallow within the sediment which is similar scenario to H1.

From both SIMS and FT-ICP-OES, the highest foraminiferal Mn/Ca and greatest Mn variability within individual tests are found during the YD (Fig. 3). This consistent pattern of Mn enrichment coincides with high primary productivity, high export flux and the dominance of low oxygen tolerant *E. exilis*. As MnO_2 is rapidly reduced to soluble Mn^{2+} in hypoxic pore waters (Burdige 1983; Limburg et al., 2011 and references therein) and since high phytodetritus export typically causes low bottom water oxygen concentrations or even anoxia within millimetres of the sediment-water interface, we can expect a high accumulation of Mn (Hunt, 1983; McKay et al., 2005) as represented in Fig 5d. Furthermore, the sediment accumulation rate during the interval is the highest of the record (350 cm ka^{-1}, Fig. 4) which can also intensify the low oxygen concentrations and promote Mn enrichment (McKay et al. 2005). This confirms our hypothesis that Mn/Ca values in *E. exilis* respond to the oxygen deficiency instigated by the large increase in diatom input. Furthermore, the redox front would also vary in position during this oxygen depleted period with diatom export fluctuating at such great magnitude. Therefore the strong intra-test variability (Fig. 3) may be representative of actual changes in oxygenation of the habitat during test growth, due to amplified seasonal fluctuations in diatom export. Moreover, since *E. exilis* is an infaunal species, it is influenced by the microhabitat of the pore waters. Therefore, the sediment depth at which this species resided and possibly migrated to during

calcification in such low oxygen conditions could further explain the greater Mn/Ca variability during the YD.

We emphasise that the benthic foraminiferal Mn/Ca seems to represent a more regional signal due to diatom input opposed to being generated by deep bottom water formation and poor ventilation. However, whilst we interpret that the sheer level of diatom input provoked O_2 deficiency in the bottom and pore waters, we acknowledge that even in the modern ocean, it is difficult to separate the effects of productivity and deep-water oxygen concentrations since they are inter-related. Gallego-Torres et al., (2014) interpret the YD as a phase of reduced ventilation coinciding with reduced Atlantic Meridional Overturning Circulation (AMOC), promoted by intense export productivity. Both effects provided dissolved Mn available for incorporation into the foraminiferal calcite. Therefore, whilst Mn/Ca has potential for oxygen level reconstruction, supplementary proxies are required in order to determine the precise factor driving the conditions.

5. Conclusion

Our study contributes to the development of Mn/Ca in benthic foraminiferal calcite as a proxy for reconstructing past oxygen conditions. The results based on data from the low latitude NE Atlantic Upwelling System indicate that shifts in oxygen levels occurred during different productivity regimes between 35 and 11.5 ka and thus foraminiferal Mn/Ca can assist our understanding of the past environment in the region studied. The foraminiferal Mn/Ca results are supported by benthic foraminiferal faunal data.

The highest foraminiferal Mn/Ca and greatest Mn variability within individual tests were obtained during the YD and indicate Mn enrichment which coincides with very high primary productivity and the dominance of low oxygen tolerant benthic foraminifera *Eubuliminella exilis*. The results confirm our hypothesis that Mn/Ca in *E. exilis* can be applied as a proxy for oxygen deficiency, in this case instigated by the increase in diatom input. Therefore, whilst the benthic faunal abundance data is indicative of such a scenario, foraminiferal Mn/Ca allows a more comprehensive interpretation. Our initial down-core dataset raises the implication of calibrations. Once developed, Mn/Ca determined by the SIMS method in particular may have the potential to be applied to other study regions and foraminiferal species for reconstructing former bottom water oxygen conditions.

Furthermore, we conclude that SIMS determined Mn/Ca upon individual tests is comparable with bulk foraminiferal Mn/Ca measured by FT-ICP-OES. However, due to the processing time required to program and target delicately thin foraminiferal test walls, SIMS may not be practical for Mn/Ca studies where large numbers of samples must be processed. Nevertheless, we emphasize that SIMS has great potential to provide reliable Mn/Ca results from just a few individual foraminifera. Therefore, SIMS is a robust alternative method to FT-ICP-OES; ideal for employing on samples that lack a sufficient abundance of individual benthic foraminiferal specimens for solution-based bulk analyses. Furthermore, SIMS is also non-destructive and thus foraminiferal test cross-sections can even be re-measured.

In contrast, foraminiferal Mn/Ca data does not continuously exhibit a consistent trend with Mn/Al determined from bulk sediment measurements. The reason for this discrepancy is that Mn related to redox fronts within the sediment provides a diagenetic signal and thus it continues to react and shift after deposition, whereas foraminiferal tests record the Mn concentration at the time of calcification.

Author contribution:

H.L. Filipsson initiated the project; C.L. McKay designed the sampling plan and carried out sample selection and preparation with assistance from T. Toyofuku. J. Groeneveld performed FT-ICP-OES analyses, C.L. McKay and M.J. Whitehouse performed SIMS analyses. D. Gallego-Torres and O.E. Romero provided Mn/Al and diatom data. C.L. McKay prepared and wrote the manuscript with contributions from all co-authors.

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Van der Weijden, C. H.: Pitfalls of normalization of marine geochemical data using a common divisor, Marine Geology, 184, 167-187, 2002.

725 Table 1. Sample list: climatic intervals, export productivity according to diatom export, average
726 foraminiferal Mn/Ca for different samples determined by SIMS and FT-ICP-OES.

727

Sample no.	Depth (cm)	Age (ka)	Climatic interval	Export productivity	SIMS: average Mn/Ca (µmol mol^{-1})	1 sd (µmol mol^{-1})	SIMS: average Mn/Ca (µmol mol^{-1}) per sample depth	FT-ICP-OES : average Mn/Ca (µmol mol^{-1})
YD A	170	12.2	YD	High	138	62	198	220
YD B	170	12.2	YD	High	191	116		
YD C	170	12.2	YD	High	321	251		
YD D	170	12.2	YD	High	141	56		
YD E	270	12.7	YD	High	178	82	164	160
YD F	270	12.7	YD	High	96	53		
YD G	270	12.7	YD	High	117	48		
YD H	270	12.7	YD	High	164	120		
YD I	270	12.7	YD	High	183	51		
YD J	270	12.7	YD	High	111	27		
YD K	270	12.7	YD	High	261	275		
YD L	270	12.7	YD	High	204	78		
BA A	365	13.5	BA	Moderate-high	155	37	Only 1 specimen available	280
BA B	430	15.1	BA	Moderate-high	242	42	130	110
BA C	430	15.1	BA	Moderate-high	63	51		
BA D	430	15.1	BA	Moderate-high	217	51		
BA E	430	15.1	BA	Moderate-high	49	18		
BA F	430	15.1	BA	Moderate-high	179	88		
BA G	430	15.1	BA	Moderate-high	175	87		
BA H	430	15.1	BA	Moderate-high	67	149		
BA I	430	15.1	BA	Moderate-high	47	27		
H1 A	500	16.7	H1	High	57	17	117	140
H1 B	500	16.7	H1	High	154	36		
H1 C	500	16.7	H1	High	186	81		
H1 D	500	16.7	H1	High	191	142		
H1 E	500	16.7	H1	High	76	57		
H1 F	500	16.7	H1	High	92	37		
H1 G	500	16.7	H1	High	62	19		
H1 H	500	16.7	H1	High	116	36		
LGM A	723	20.7	LGM	Low	27	14	74	
LGM B	723	20.7	LGM	Low	146	28		
LGM C	723	20.7	LGM	Low	115	71		
LGM D	723	20.7	LGM	Low	7	4		Insufficient no.
LGM E	773	22.7	LGM	Low	7	3	65	of specimens
LGM F	773	22.7	LGM	Low	10	3		
LGM G	773	22.7	LGM	Low	70	49		

LGM H	773	22.7	LGM	Low	175	91		
MIS3 A	928	29.9	MIS3	Low	38	13	43	
MIS3 B	928	29.9	MIS3	Low	48	38		Insufficient no.
MIS3 C	928	29.9	MIS3	Low	45	24		of specimens
MIS3 D	1058	34.0	MIS3	Low	67	23	61	
MIS3 E	1058	34.0	MIS3	Low	55	62		

Table 2. Post hoc test (Mann Whitney U) *z* values of the mean foraminiferal Mn/Ca (per individual) between climatic intervals. Significant differences are highlighted in bold.

	YD	**BA**	**H1**	**LGM**	**MIS3**
YD					
BA	-1.14				
H1	-1.93	-0.19			
LGM	**-2.70**	-1.83	-1.68		
MIS3	**-3.16**	**-2.07**	0	0	

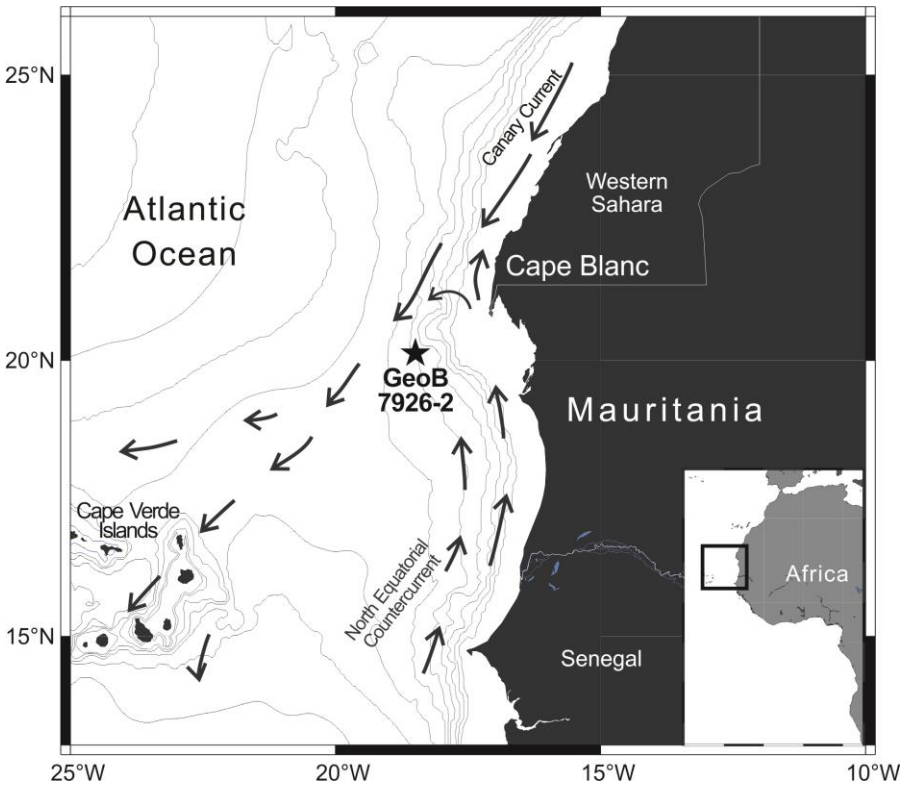

Figure 1. Locality of gravity core GeoB7926-2 (black star) in the low latitude NE Atlantic upwelling area. Arrows indicate the major oceanic currents in the study area. Inset: Location of the study area off coastal NW Africa. Modified after Romero et al. (2008).

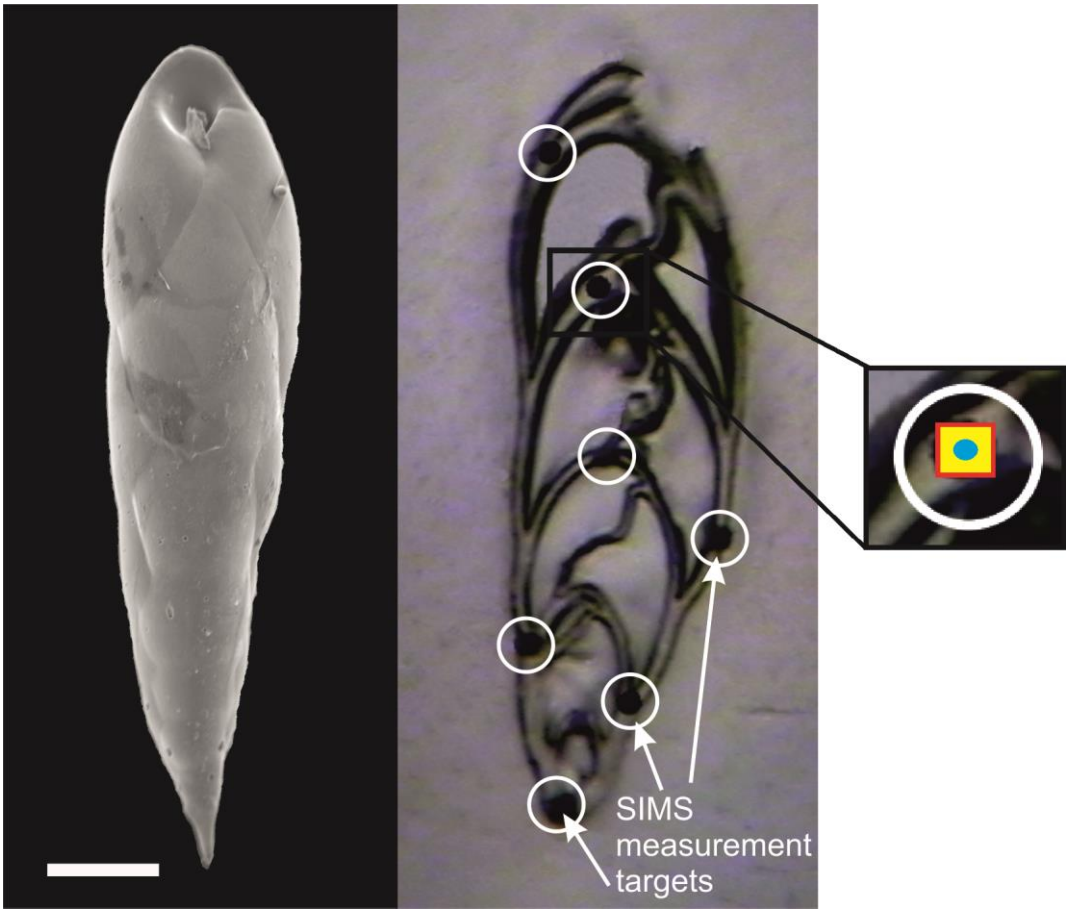

Figure 2. SEM image (scale bar: 100 µm) and cross section image during SIMS analysis of a single *Eubuliminella exilis* specimen. The white circles highlight the selected spots for SIMS analyses, measuring 5 µm in diameter. Note that the black areas of the SIMS measurement targets visible in this image are actually the 5 µm spots plus the 10 µm pre-sputters. Inset is a close-up of the SIMS targets: the red square is the approximate pre-sputter area (15 x 15 µm, i.e. 5 µm spot + 10 µm raster), the yellow area is the field of view admitted to the mass spectrometer (controlled by magnification and field aperture) and the blue ellipse is the nominal 5 µm spot.

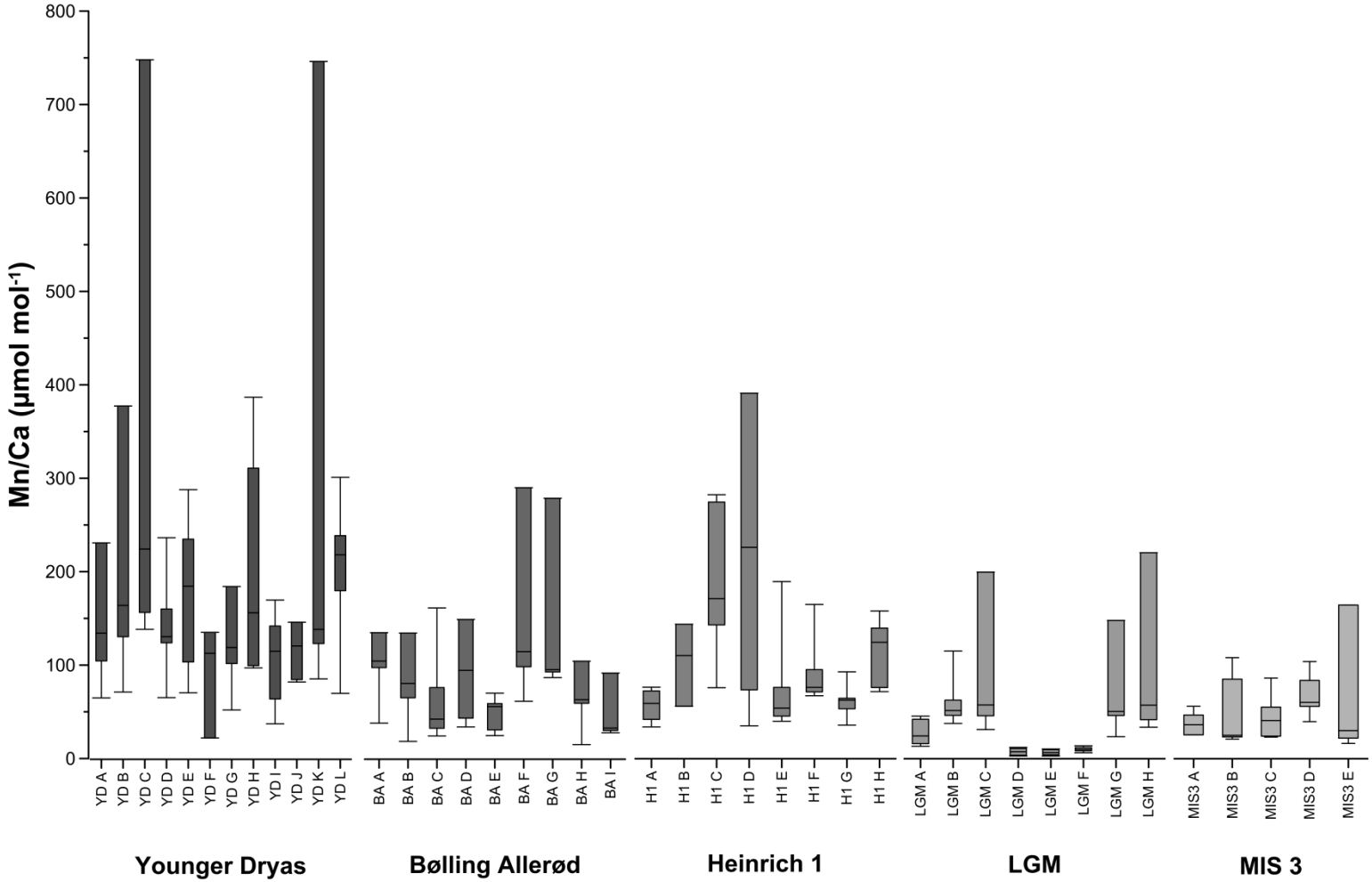

Figure 3. The Mn/Ca (µmol mol^{-1}) variability within each individual *Eubuliminella exilis* specimen for each climatic interval (labelled on the x-axis), determined by SIMS.

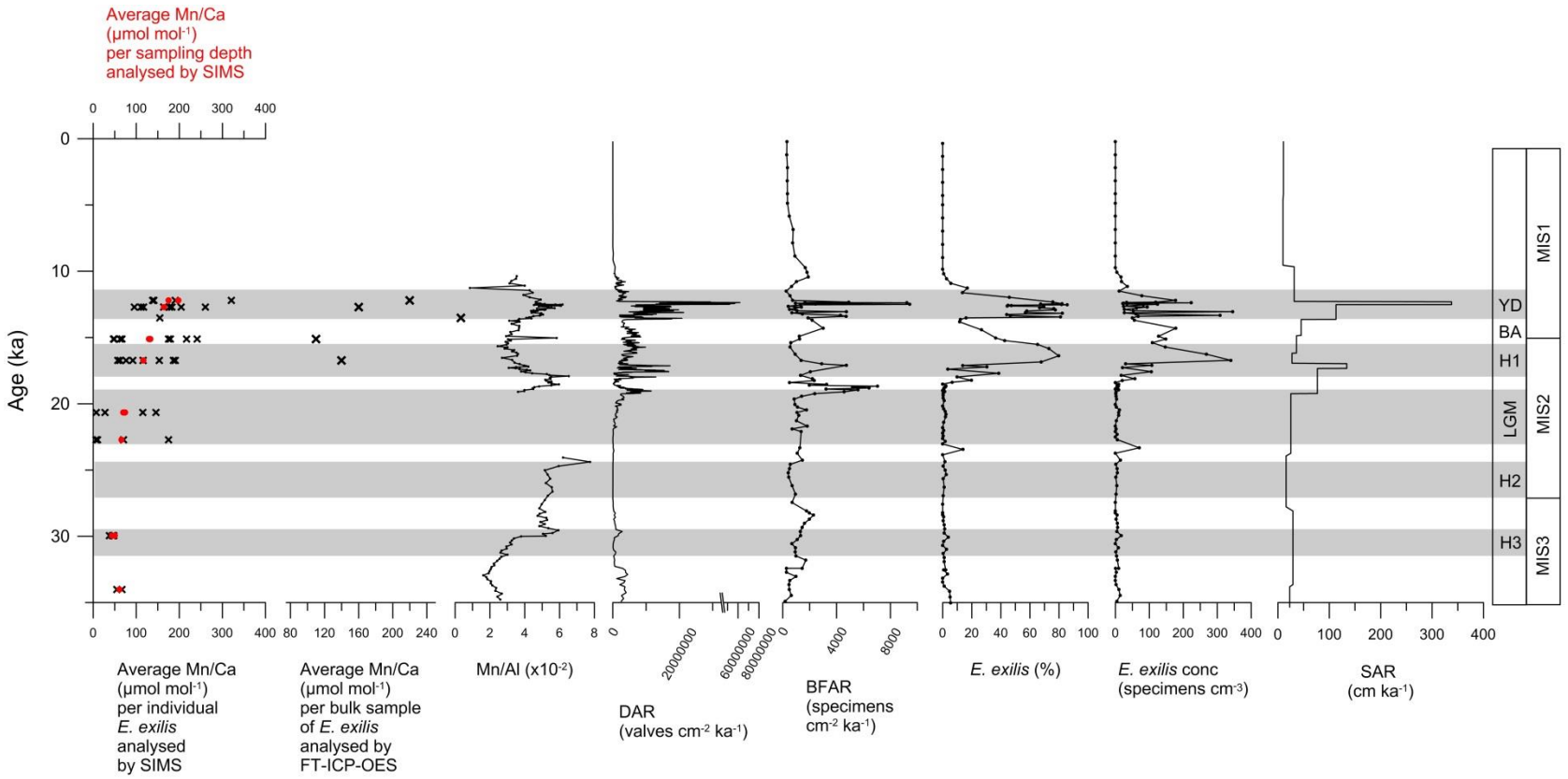

Figure 4. Foraminiferal Mn/Ca (µmol mol^{-1}) determined by SIMS (black crosses: average per specimen, red circles: average per depth) and FT-ICP-OES, bulk sediment Mn/Al (Gallego-Torres et al., 2014), diatom accumulation rate (DAR; Romero et al., 2008), Benthic foraminiferal accumulation rate (BFAR; McKay et al., 2014), relative abundance and concentration of low oxygen indicating *Eubuliminella exilis* and sediment accumulation rate (SAR) of core GeoB7926-2.

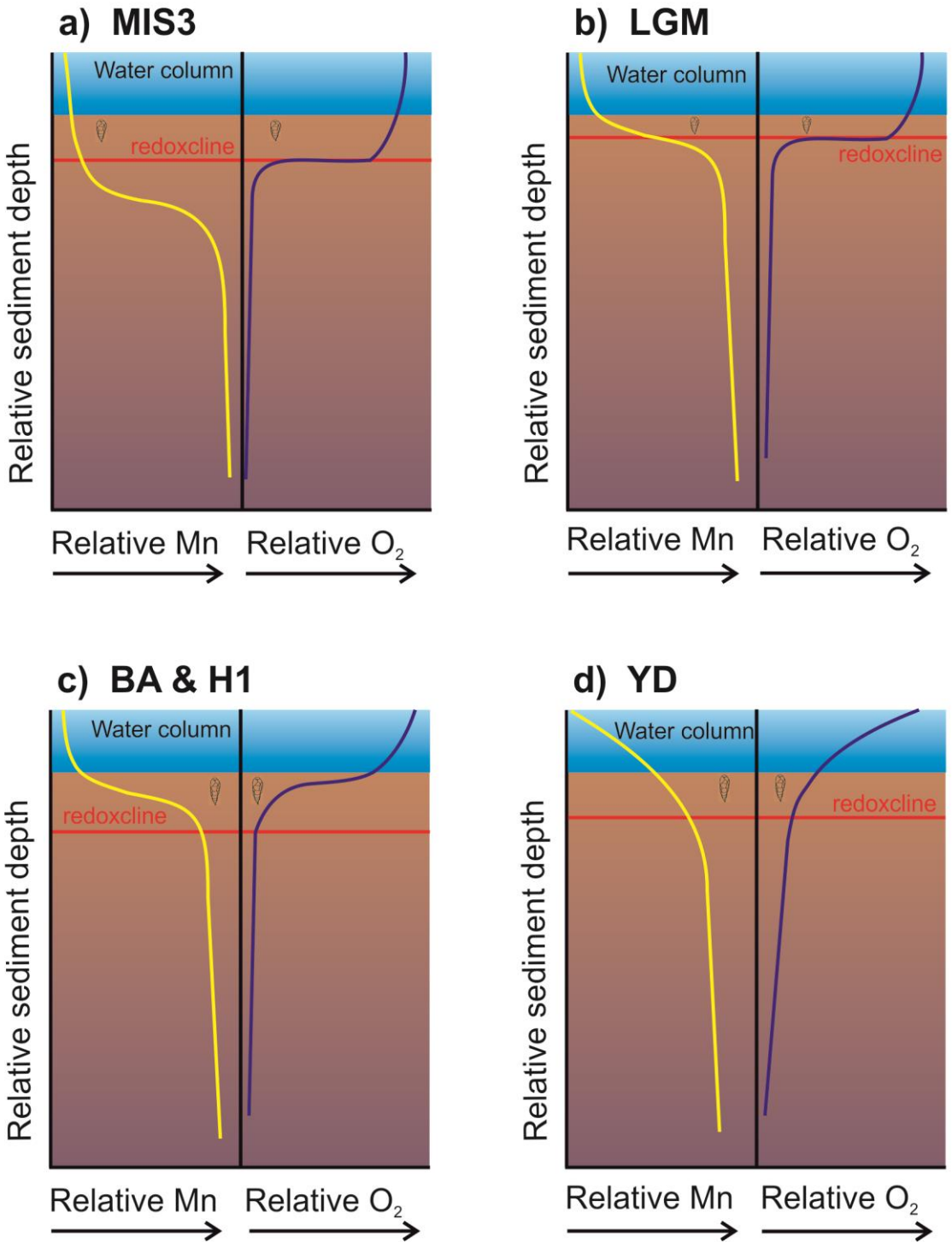

Figure 5. Idealised schematic pore water Mn (yellow) and oxygen (blue) profiles for the climatic intervals studied from the GeoB7926-2 record. The red line represents the redoxcline.

a) MIS3: Low foraminiferal Mn/Ca and therefore the sedimentary Mn was precipitating below the living depth of *E. exilis* and more oxygenated conditions prevailed in the bottom and pore

815 waters. b) LGM: foraminiferal Mn/Ca increases and therefore a relative decrease in pore water
816 oxygen is apparent and the redoxcline is shallower. c) BA & H1: foraminiferal Mn/Ca increases
817 further and the moderate-low sedimentary Mn/Al indicates low bottom water conditions. d) YD:
818 highest accumulation of Mn and therefore oxygen deficiency in the bottom and pore waters.

819