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Variations in benthic foraminiferal assemblages in the Tagus mud belt during the last 5700 years: Implications for Tagus River discharge

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1 Abstract

We analyzed a 10-m sediment core retrieved at 82 m water depth off the coast of the 2 Tagus River (Western Iberian Margin, Portugal) to investigate a linkage between variations in 3 benthic foraminiferal assemblages and Tagus River discharge over the last 5700 years. Benthic 4 for a miniferal assemblages were studied at high resolution in combination with the stable carbon 5 6 and oxygen isotopic composition of fossil shells of Nonion scaphum, bulk and molecular organic matter properties (TOC, TN, C/N ratio, $\delta^{13}C_{TOC}$, $\delta^{15}N_{bulk}$, and BIT index), magnetic 7 susceptibility, and XRF analyses. Three periods of environmental changes were identified: 1) 8 high Tagus River discharge in 5750-2200 calendar year before present (cal yr BP), 2) lower 9 discharge characterized by intense upwelling conditions (2250-1250 cal yr BP), and 3) both 10 intense upwelling and Tagus River discharge (1250 cal yr BP-present). The data reveal 11 alternating intense upwelling periods, as shown by the dominance of *Cassidulina carinata*, 12 13 Valvulineria bradyana, or Bulimina marginata, whereas periods of increased river discharge are indicated by increase of N. scaphum, Ammonia beccarii, and Planorbulina mediterranensis. 14 The Tagus River discharge was the strongest during the first period, transporting riverine 15 16 material further offshore and preventing the establishment of a mud belt on the mid-shelf (around 100 m depth). During the second period, a decrease in Tagus River discharge favored 17 the formation of the Tagus mud belt and strongly influenced the benthic environment by 18 creating an organic matter stock. During the third period, intense upwelling and increased Tagus 19 20 River discharge were recorded by benthic foraminiferal distribution, with an increase of 21 terrestrial elements present in the mud belt. Furthermore, our results showed that variations in benthic foraminiferal assemblages corresponded to the well-known climatic periods in the study 22 area, such as the Roman Period, the Dark Ages, the Medieval Warm Period, and the Little Ice 23 24 Age. Our study strongly suggests that benthic foraminiferal assemblages can be used as a bioindicator to trace the influence of past river discharge. 25

Key words: Holocene, Portuguese Margin, Paleo-reconstruction, North Atlantic Oscillation

27

28 **1. Introduction**

In the last decades, benthic foraminiferal assemblages have often been used in 29 paleoceanography to investigate biostratigraphy, paleobathymetry, and abiotic conditions, such 30 31 as temperature, salinity, and pH (Jorissen et al., 2007 and references therein). The TROX model (Jorissen et al., 1995) established that oxygen concentration and organic matter content in the 32 33 sediment are the major environmental controls on the distribution of benthic foraminifera in marine sediments. Subsequent studies emphasized the importance of the quality of the organic 34 35 matter for the composition of living benthic foraminifera faunas in various marine environments 36 (e.g., Goldstein and Corliss, 1994; Suhr et al., 2003; Fontanier et al., 2005, Goineau et al., 2011, 37 Dessandier et al., 2015). Benthic foraminifera are known to bloom following high marine algal production (e.g., Kitazato et al., 2000). After a phytoplankton bloom, benthic foraminifera 38 39 respond rapidly to the increased influx of fresh organic matter (Fontanier et al., 2003). Terrestrial input may also impact the organic matter supply and its quality and thereby may 40 partly control the benthic foraminiferal distribution in continental shelf sediments. This is 41 particularly true for river-dominated shelves where living benthic foraminifera have been 42 identified to respond to river inputs (e.g., Mendes et al., 2004; Mojtahid et al., 2009; Goineau 43 44 et al., 2011).

Dessandier et al. (2016, 2018) have shown through studies on living and dead faunas that some species could be used as bio-indicators of river discharge and upwelling intensity in the Portuguese Margin. During the last millennia, these environmental conditions were influenced by rapid climatic changes (Abrantes et al., 2005, Lebreiro et al., 2006). Within the Quaternary, the Holocene is characterized by relatively stable climatic conditions with millennial-scale variability (Bond et al., 1997). The solar variability superimposed on long-term

changes in insolation seems to be one of the most likely important forcing mechanisms for the 51 52 rapid climate changes over the Holocene (Mayewski et al., 2004). The North Atlantic Oscillation (NAO) was also described as a major factor for the variability of climate in Europe 53 (Wanner et al., 2001), which played an important role in the control of the upwelling intensity 54 and humidity of the Iberian Peninsula (Abrantes et al., 2005; Lebreiro et al., 2006; Fig. 1). The 55 NAO is an atmospheric process characterized by a seesaw between the Icelandic low and the 56 Azores high pressures. The positive mode (NAO+) is characterized by well-developed 57 Icelandic low and Azores high pressures, associated with stronger westerlies over the eastern 58 North Atlantic and the European continent. The negative mode (NAO-) is characterized by a 59 60 rather weak pressure seesaw and reduced westerlies (Wanner et al., 2001). The last millennia were marked by short-scale climatic changes, mainly controlled by solar activity and NAO-like 61 processes. Several such periods of short-scale changes were identified, among them the Roman 62 63 Period (RP; e.g., Lamb, 1985), the Dark Ages (DA; e.g., Keigwin and Pickart, 1999), the Medieval Warm Period (MWP), the Little Ice Age (LIA; deMenocal et al., 2000; Trouet et al., 64 65 2009), and a warming during the twentieth century determined by instrumental temperature measurements of the last two centuries. The major climatic changes, MWP and LIA, have been 66 described as controlled by NAO+ and NAO-, respectively (Lebreiro et al., 2006; Trouet et al., 67 68 2009). The nature and chronology of these events are still debated (Desprat et al., 2003; Ortega et al., 2015; Swingedouw et al., 2015), but the influence of these abrupt changes on the rainfall 69 and upwelling intensity has been observed in the Iberian Margin (Abrantes et al., 2005; Bartels-70 Jónsdóttir et al., 2006). During the NAO- mode, the Iberian Margin is marked by increased 71 72 humidity, which is responsible for strengthened riverine discharge (Bernárdez et al., 2008). The 73 NAO+ mode is characterized by an increase in Iberian coastal upwelling (Lebreiro et al., 2006). Another oscillation, called the Atlantic Multidecadal Oscillation (Kerr, 2000), was also 74

described as a controlling factor of rainfall during the last century that had a greater influenceon the American continent, Sahel, and northern Europe (Knight al., 2006).

We tested the applicability of these benthic foraminifera as bio-indicators of past 77 upwelling intensity and changes in river discharge on a late Holocene sediment sequence from 78 79 the Tagus prodelta. We used a multiproxy approach based on organic parameters (total organic carbon [TOC], total nitrogen [TN], C/N ratio, $\delta^{15}N_{bulk}$, and $\delta^{13}C_{TOC}$), XRF data, benthic 80 81 foraminiferal assemblages, and carbon and oxygen stable isotopes of benthic foraminifera to compare climatic changes of the late Holocene with river regime change. The branched 82 isoprenoid tetraethers (BIT) index was measured in the core that was previously published in 83 84 Warden et al. (2016) and compared with our reconstruction. We applied our reconstruction on 85 a paleoclimatological record never studied at this water depth, under the influence of the Tagus River, and provided a new dataset to improve understanding of the effect of NAO-like processes 86 during the Holocene. 87

88

89 **2. Study area**

Our study area was located at the mid-shelf off the Tagus River mouth, on the western 90 Iberian Margin (Fig. 1) which is a narrow (20–34 km) shelf (Dias et al., 2002) that extends from 91 37°N to 42°N latitude. The shelf break is located at 140 m depth, on the outer continental shelf 92 and slope, and three main submarine troughs (the Cascais Canyon, Lisbon Canyon, and Setùbal 93 94 Canyon) are related to geological features (Jouanneau et al., 1998). The regional hydrodynamic 95 regime is driven by the influence of wave action, tidal and rip currents, and storm surges, which plays a role in determining the particle size distribution on shelf sediments (Oliveira et al., 96 97 2007). However, the Tagus area is protected against swell from the northwest (Jouanneau et al., 98 1998). Sandy deposits occur on the inner shelf, where there are high-energy littoral currents. The dominant regional surface current (the Portugal Coastal Counter Current) flows southward 99

and transports material from the shelf to the deep ocean during winter storms (Vitorino et al., 100 2002b). The outer shelf is affected by internal waves, especially during periods when water 101 masses are stratified, resulting in the deposition of large bodies of sandy and gravelly sands 102 103 (Jouanneau et al., 1998). The export of terrestrial sediment and nutrients along the shelf is predominantly influenced by sediment supplied by three main rivers: the Douro, Tagus, and 104 Sado (Jouanneau et al., 1998; Dias et al., 2002). At around 100 m water depth, the finest 105 particles are deposited in accumulation areas, called the mud belt (or mud patch). The mud belt 106 107 lies beyond the line where seasonal thermal stratification of surface waters occurs, especially off the mouths of the Douro and Tagus rivers (Jouanneau et al., 1998; Dias et al., 2002). These 108 109 accumulation zones are composed of mixed sources of marine, estuarine, and terrestrial organic matter (Schmidt et al., 2010). Winter storms can remobilize the sediment and transport it 110 northward by the action of bottom currents (Dias et al., 2002; Vitorino et al., 2002a), eventually 111 112 depositing it on the mid-shelf mud belt between 50 and 130 m water depth (Vitorino et al., 2002b). 113

The Eastern North Atlantic Central Water (ENACW) is a slope current characterized by 114 a decrease of salinity under the surface currents, with a minimum of 35.6 at 450-500 m water 115 depth (Oliveira et al., 2007). The ENACW is upwelled during summer. Between May and 116 September, the Azores high-pressure system is driven closer to the coast. Together with the 117 associated northerly winds, this atmospheric system makes the colder, less salty, and nutrient-118 enriched subsurface water (60–120 m water depth) rise to the surface along the Iberian Margin 119 (Fiúza, 1983). This upwelling leads to an increased productivity in the summer along a 50-km-120 wide zone. The main upwelling front is oriented along the bathymetry off the 100 m isobath in 121 the northern part of the Iberian Peninsula; it then turns slightly offshore and southward (Peliz 122 et al., 2002). Most of the primary producers, especially cyanobacteria and diatoms, that are 123 dominant in the Iberian Margin increase during upwelling events (Tilstone et al., 2003). Active 124

upwelling periods have a large impact on marine trophic conditions in this area (Fiúza, 1983) 125 and are characterized by maximal organic carbon exports to the seafloor (Jouanneau et al., 126 1998). Periods of strong Iberian river discharge that occur during phases when upwelling is 127 reduced are characterized by a substantially increased export of continental nutrients, which 128 triggers phytoplankton production (Prieto et al., 2009; Rodrigues et al., 2009). To a lesser 129 extent, phytoplanktonic blooms occur in November and between April and May (Ferreira and 130 Duarte, 1994). In winter, the Azores high-pressure system moves south, which results in 131 southerly winds and downwelling conditions that lead to the deposition of sediments on the 132 shelf (Frouin et al., 1990). Upwelling activity and fluvial discharge are consequently the two 133 134 major parameters controlling the marine biology on the Iberian Shelf (Lebreiro et al., 2006) due to their impact on the deposition of organic matter, which is important for benthic organisms. 135

136 The Tagus is the longest (1,008 km) Iberian river in the central part of the Peninsula and has a large mesotidal estuary with an area of 340 km² (Vale and Sundby, 1987). The Tagus 137 watershed is about 80,600 km² and has an annual mean water discharge of 360 m³ s⁻¹ (Jouanneau 138 et al.,1998), with strong seasonal changes in discharge from 1 to 2,200 m³ s⁻¹ (Atlas Nacional 139 de España), controlled by maximal rainfall in winter (Aguiar and Ferreira, 2005; Azevedo et 140 al., 2008). The Tagus River flows were largely modulated by the NAO during the last century 141 (Trigo et al., 2004) and the last millennia (e.g., Abrantes et al., 2005; Bartels-Jónsdóttir et al., 142 2006; Lebreiro et al., 2006). The river discharge also controls the input of chlorophyll 143 associated with phytodetritic material in the marine environment and the water column 144 stratification (Relvas et al., 2007). 145

146

147 **3. Material and methods**

148 Core 64PE332-30-2 was obtained in March 2011 during the Pacemaker 64PE332 cruise
149 on board the R/V *Pelagia* (38°39'04''N, 9°28'13''W). This 978-cm Kullenberg piston core

was retrieved from the Tagus mud belt at 82 m water depth (Fig. 1). Sediment slices 1 or 2 cm 150 thick were sampled every 10 cm, dried, weighted, and washed through 63 and 150 µm sieves. 151 For this study, 101 samples of >150 µm benthic foraminifera were handpicked and placed in 152 153 Chapman cells before taxonomic identification under a stereomicroscope. After splitting using an Otto microsplitter, a minimum of 250 specimens were counted. Diversity indices (Shannon 154 [S] and Evenness indices) were calculated using the PAleontological STatistics (PAST) 155 156 software (Version 2.14; Hammer et al., 2001). The benthic foraminiferal number (BFN), which represents the number of individuals per analyzed dry sediment mass, was calculated for all 157 samples. 158

159 Isotopic analyses were performed on monospecific samples of the benthic foraminiferal species Nonion scaphum, which is present all along the core and is typical of the Iberian mud 160 belt (Dessandier et al., 2015; 2016), at the EPOC laboratory, University of Bordeaux. This 161 162 species was observed alive below the oxygen penetration, between 1 and 2 cm depth in the mud belt area (Dessandier et al., 2016), suggesting that it may reflect subsurface sediment pore water 163 rather than bottom water conditions. However, we compared only the data measured on the 164 same species in every sample, avoiding any bias from the early diagenesis effect for 165 environmental reconstruction. For each sample, three or four specimens were handpicked and 166 167 dissolved in acid via the Micromass Multiprep autosampler system. The resulting carbon dioxide gas was analyzed against the international reference standard NBS 19 ($\delta^{13}C = +1.96$ 168 % / PDB and $\delta^{18}O = -2.20 \%$ / PDB) using an Optima Micromass mass spectrometer. 169 Measurements were taken for each depth horizon (1 cm) in triplicate to reduce uncertainties. 170 The analytical precision was better than 0.05 ‰ for δ^{18} O and 0.03 ‰ for δ^{13} C. 171

The age model for the sediment core, modified according to Warden et al. (2016), was based on the magnetic susceptibility (MS) and seven accelerated mass spectrometry (AMS) ¹⁴C radiocarbon dates (Table 1). The MS record of Core 64PE332-30-2 was compared with that of

Core GeoB 8903 (Abrantes et al., 2008), which was also retrieved in the Tagus prodelta at ~100 175 m water depth (Fig. 1). Table 1 summarizes the ¹⁴C AMS dating points of the two cores 176 considered in this study. Because of the low number of dating points in the upper part of Core 177 64PE332-30-2, we compared our record of MS and age model with Core GeoB 8903, which 178 had a substantial number of ¹⁴C AMS dating points in the upper section (Figs. 2 and 3). The 179 MS was measured on board at 5-cm intervals using a Bartington MS meter with a 12-cm 180 diameter loop. The final age model was achieved by a linear interpolation between each AMS 181 ¹⁴C age. The ¹⁴C data calibration was made via the program CALIB V0.6 with the Marine13 182 calibration curve (Stuiver and Reimer, 1993), using the common reservoir age of 400 years 183 because no regional effect on reservoir age is known in our sampling area (Abrantes et al., 184 2005). AMS ¹⁴C ages and the dating points of GeoB8903 were converted to cal yr BP. 185

Core 64PE332-30-2 was scanned with an Avaatech XRF core scanner at NIOZ at 1-cm 186 187 resolution. Detailed bulk-chemical composition records acquired by XRF core scanning allow accurate determination of stratigraphical changes and assessment of the contribution of the 188 various components in lithogenic and marine sediments (Stuut et al., 2014). The XRF core 189 scanner uses energy dispersive fluorescence radiation to measure the chemical composition of 190 the sediment as element intensities in total counts or counts per second (Tjallingii et al., 2007). 191 192 After cleaning and preparation of the archive-halve core surface and covering with SPEX Certi Ultralene[®] foil, the core was measured at both 10 kV and 30 kV. Element intensities are 193 presented as log ratios that are normally distributed and linearly related to log ratios of element 194 concentration (Weltje and Tjallingii, 2008). Terrestrial exports, such as metals or contaminants, 195 are indicated mainly by Fe/Ti and Pb/Ti ratios. The Zr/Rb ratio serves as a grain size indicator 196 (Taylor, 1965), and the Br/Cl ratio indicates organic sediment (Ziegler et al., 2008). 197

198 Sediments were freeze-dried and ground before the geochemical analyses. TN and δ^{15} N 199 were measured with a Thermo-Scientific Flash 2000 Elemental Analyzer interfaced at NIOZ. The analyses were determined at least in duplicate and the analytical error was, on average, smaller than 0.1 wt. % for the TN content. The TOC content, the stable carbon isotopic composition of TOC ($\delta^{13}C_{TOC}$), and BIT data were previously published by Warden et al. (2016). The C/N ratio was calculated as the division of TOC/TN.

A principal component analysis (PCA) was performed on 22 samples of normalized environmental and faunal data, using PRIMER version 6.0 software (Clarke and Warwick, 1994) to compare the response of the faunal and environmental parameters. The two major PCA scores were plotted to define the different phases of the reconstruction.

208

209 **4. Results**

210 <u>4.1. Sedimentological features and age model</u>

The final age model of Core 64PE332-30-2 (Fig. 3) revealed that the sedimentation rates 211 were increasing over time, with a first phase of ~ 0.06 cm/yr⁻¹ from 5750 to 2200 cal yr BP. In 212 a second phase, 2200-present, the sedimentation rate was ~ 0.52 cm yr¹. The MS record and 213 214 grain size distribution of the Core GeoB 8903 were plotted as a function of age, and the MS and Ca/Ti record of Core 64PE332-30-2 were plotted as a function of core depth (Fig. 2). 215 Similar trend signals in the MS records were identified in the two cores, and the Ca/Ti record 216 measured by XRF showed a similar trend to the grain size record of core GeoB 8903. The MS 217 record showed three phases, with the first characterized by an increase of MS at 770 cm 218 sediment core depth, the second a larger increase at around 400 cm, and the third a more stable 219 trend until the end. This trend was opposite that of the Ca/Ti content, which revealed two 220 successive decreases of values at the same depths. 221

222

223 <u>4.2. Environmental change phases</u>

XRF, organic matter, and benthic foraminiferal isotopes data are plotted in Fig. 4. Three 224 225 phases appeared following the environmental changes, mainly determined by the XRF ratios. Phase I (575-2250 cal yr BP) was characterized by high Zr/Rb and Ca/Ti counts, while the 226 Br/Cl and Fe/Ti counts were low. The BIT index was low and stable (below ~ 0.04), the TOC 227 content was also stable at around 0.9 wt. %, $\delta^{13}C_{TOC}$ was about – 24.3 ‰, and $\delta^{15}N_{bulk}$ was 228 about + 3.9 ‰ (Warden et al., 2016). The TN was the only organic parameter that slightly 229 230 increased during this phase, from 0.04 to 0.08 wt. % (Warden et al., 2016). The C/N ratio 231 showed an opposite trend, with a clear decrease from 18 to 11. Both the carbon and oxygen stable isotope composition of N. scaphum slightly decreased during this interval, $\delta^{18}O_{bf}$ from + 232 1.7 to + 1.4 and $\delta^{13}C_{bf} \text{ from} - 1.1 \text{ to} - 1.2 \text{ }$. 233

Phase II (2250-1250 cal yr BP) was characterized by a rapid decrease in Ca/Ti and Zr/Rb 234 counts, a strong increase in Br/Cl counts, and a slight increase in Fe/Ti. The BIT index increased 235 slightly, to 0.06. The TOC, $\delta^{13}C_{TOC}$, and C/N ratio reached high values at the end of the period 236 (around 1.5 wt. %, – 23.0 ‰, and 16, respectively) after a large increase. TN and $\delta^{15}N_{bulk}$ 237 showed a smaller increase; TN moved from 0.9 to 0.13 wt. % and $\delta^{15}N_{bulk}$ from + 3.6 to + 3.9 238 239 ‰. Isotopes measured on benthic foraminifera became more variable at the beginning of the period, and this trend continued until the end of the record. The decrease in δ^{13}_{bf} observed in 240 phase I was stronger, and $\delta^{18}O_{bf}$ remained fairly constant. 241

Phase III (1250 cal yr BP-present) was characterized by a continued decrease of Ca/Ti counts and a large decrease of Zr/Rb counts. Br/Cl counts were also unstable and lacking any clear trend. The Fe/Ti and Pb/Ti counts from Phase III showed an increase until the end of the record, especially during the last 500 years for Pb/Ti, after a stable trend during the two previous phases. They both peaked at 250 cal yr BP, when Br/Cl decreased. The TOC content, $\delta^{13}C_{TOC}$, and C/N ratio decreased, to 1.2 wt. %, – 24 ‰, and 8, respectively. The $\delta^{13}C_{TOC}$ and TOC increased during the last 200 years, reaching – 28.8 ‰ and 1.2 wt. %, respectively. TN and $\delta^{15}N_{\text{bulk}}$ were roughly constant before a large increase at the end of the period, reaching 0.17 % and + 4.7 ‰, respectively. The $\delta^{18}O_{\text{bf}}$ increased at the beginning from + 1.3 to + 1.8 ‰ and decreased until + 1.5 ‰ at the end; $\delta^{13}C_{\text{bf}}$ showed the opposite trend, with a large decrease from -1.5 to -3.5 ‰. The C/N ratio was slightly decreasing from the start of this period until the present. Conversely, the BIT index was progressively increasing, reaching 0.12, but dropped in the most recent sediment horizon analyzed.

255

256 <u>4.3. Benthic foraminiferal distribution over the last 5750 years</u>

257 Fig. 5 shows percentages of the major species (> 5 %). Based on the distribution of these dominant taxa, three different phases could be identified. The first phase does not map to the 258 environmental phases and ends at 2500 cal yr BP. During this phase (5750-2500 cal yr BP), N. 259 scaphum and Ammonia beccarii dominated, making up 30 and 18 % of the total species, 260 respectively. Planorbulina mediterranensis and Bolivina spathulata were relatively abundant 261 262 as well, with each ~10 % of the population. A. beccarii was particularly dominant (10-20 %) between 5750 and 4750 cal yr BP and then quickly decreased to ~ 5 %. The epibenthic species 263 264 *Cibicides lobatulus* was only > 5 %, while *Hyalinea balthica* and *Uvigerina bifurcata* increased 265 until the end of the period. This first period was marked by a progressive increase of specific richness; the S index increased from 28 to 36, and H' from 2.3 to 2.8. The foraminiferal density 266 (BFN) was relatively low during this period. 267

Between 2500 and 1250 cal yr BP, *Cassidulina carinata* increased sharply, while *N*. *scaphum* clearly decreased in relative abundance. *Valvulineria bradyana* was nearly absent during the first phase but became abundant from 2250 cal yr BP and reached 10 % of the fossil assemblage during the latter period of this second phase. Smaller variation in the relative abundances of the other dominant species, such as *P. mediterranea*, *A. beccarii*, *H. balthica*, and *U. bifurcata*, was observed without a clear trend. *V. bradyana* and *Bulimina marginata* increased, and *C. lobatulus* and *B. spathulata* diminished. The highest percentages of *C. carinata* (40 %) were observed between 1750 and 1500 BP, whereas *Bulimina marginata*, *P. mediterranensis*, and *B. spathulata* decreased. The BFN increased as both specific richness and the S index began an initial decline; specific richness was decreasing until 30 taxa, and H' until 2.5.

The last phase (1250 cal yr BP-present, Phase III) was characterized by higher relative 279 abundance of V. bradyana, dominance of C. carinata, and a progressive decrease of P. 280 mediterranensis, A. beccarii, and B. spathulata. The relative abundance of N. scaphum slightly 281 increased from 1250 cal yr BP, compared to 2600-1250 cal yr BP, but decreased again from 282 283 500 cal yr BP. After 1000 cal yr BP, a strong increase was recorded for C. carinata, to more than 40 % at 500 cal yr BP and in the modern period. Deep infaunal species (i.e., Chilostomella 284 oolina and Globobulimina affinis), B. marginata, and Eggerelloides scaber also increased 285 286 during this phase. The last 200 years showed strong abundances of C. carinata (44 %) and a large loss of both V. bradyana and N. scaphum, which decreased to ~ 10 %. Bolivina spathulata 287 and *H. balthica* totally disappeared, while *U. bifurcata*, *A. beccarii*, and *B. marginata* declined. 288 With C. carinata, only B. aculeata and E. scaber are increasing during this modern period. The 289 last 750 years showed a large decline of BFN, specific richness (~ 20), and S index (~ 2). 290

291

292 <u>4.4. Multiproxy approach</u>

A PCA was performed on the major environmental parameters (TOC, TN, δ¹³C_{TOC},
δ¹⁵N_{bulk}, BIT index, Ca/Ti, Fe/Ti, Br/Cl, Zr/Rb, and benthic foraminiferal stable isotopes) and
on the relative abundances of the major benthic foraminiferal species (*C. carinata*, *N. scaphum*, *V. bradyana*, *A. beccarii*, *P. mediterranensis*, *B. spathulata*, *E. scaber*, *H. balthica*, *U. bifurcata*, *B. marginata*, and deep infaunas) (Fig. 6A). PC1 and PC2 explained 64 % (52 and
12 %, respectively) of the total variance observed in the dataset. The relative abundance of *N*.

scaphum, A. beccarii, and P. mediterranensis loaded positively on PC1 and negatively on PC2, together with $\delta^{13}C_{bf}$, Ca/Ti, and Zr/Rb. Most organic compounds, such as TOC, TN, $\delta^{13}C_{TOC}$, and $\delta^{15}N_{bulk}$, loaded negatively on PC1 and PC2 with *C. carinata*, Br/Cl, and Pb/Ti. The deep infaunas and *B. marginata*, *V. bradyana*, and *E. scaber* loaded negatively on PC1 and positively on PC2, together with Fe/Ti and the BIT index. *Uvigerina bifurcata*, *H. balthica*, and *B. spatulata* loaded positively on PC1 and PC2 with $\delta^{18}O_{bf}$.

The scores of the different samples on PC1 and PC2 were plotted as a function of age 305 in Fig. 6B. The score of PC1 was stable during the first phase and then showed a slight increase 306 307 from 2500 to 2250 cal yr BP. At the start of Phase II (2250 cal yr BP), the score of PC1 decreased until 1500 cal yr BP, then increased again. At the start of Phase III (1250 cal yr BP), 308 a second progressive decrease started and characterized this phase until the present. The plot of 309 PC2 showed an increase toward positive loading until the end of phase I. The second phase 310 (2250-1250 cal yr BP) was characterized by a sharp decrease, before an increase at the 311 beginning of Phase III that continued until 750 cal yr BP. PC2 decreased, reaching 0 at 500 cal 312 yr BP, showed a slight increase between 500 and 250 cal yr BP, and decreased again during the 313 314 last period of Phase III.

315

316 **5. Discussion**

317 5.1. Benthic foraminiferal response to environmental and climatic changes

Climatic changes during the late Holocene have been well studied in the Portuguese Margin, especially for the last 3000 years (e.g., Desprat et al., 2003; Abrantes et al., 2005; Bartels-Jónsdóttir et al., 2006; Alt-Epping et al., 2009). This rendered the Holocene a suitable period to test benthic foraminifera as bio-indicators for past Tagus River discharge and environmental changes based on the living foraminiferal calibration developed by Dessandier et al. (2016, 2018) in the same location. Previous sediment cores in the area extended back to 2000 (Abrantes et al., 2005; Bartels-Jónsdóttir et al., 2006) or 3000 cal yr BP (Alt-Epping et
al., 2009). The 10-m core retrieved in our study provided a record that dated to 5750 cal yr BP.
Next, we present the three phases defined by environmental changes related to sediment
supplies, as shown by the XRF ratios and visible in the scores of the PCA (Fig. 6B).

The first phase was characterized by positive values of PC1 and a continuous increase 328 329 in PC2, starting from negative values. The Zr/Rb ratio indicates a bigger grain size, which had a strong positive loading on PC1 together with Ca/Ti. Ca was previously linked to grain size in 330 the Portuguese Margin and thought to be associated with coarse reworked shells of 331 macrobenthic organisms (Martins et al., 2007; Abrantes et al., 2008; Alt-Epping et al., 2009). 332 333 The two different increases of Zr/Rb and Ca/Ti were two successive phases in grain size decrease from Phase I to Phase III (Fig. 4). During Phase I, the TOC content of the sediment 334 and the Br/Cl were very low; both indicated low amounts of organic compounds, likely because 335 336 of this coarse grain size. This phase was characterized by river-influenced species, such as N. scaphum, A. beccarii, and P. mediterranensis. This first group of species consequently 337 338 represents the river discharge bio-indicator in this study. Among them, N. scaphum was interpreted as an indicator of active upwelling conditions in previous studies of the Portuguese 339 Margin (Bartels-Jónsdóttir et al., 2006). However, the results from late winter showed that this 340 species was clearly dominant in the living community of the Portuguese inner shelf during 341 winter. By contrast, during active upwelling context, it was present only in the dead community, 342 reflecting lower relative abundances (Dessandier et al., 2016; 2018). The large occurrences of 343 344 A. beccarii and P. mediterranensis during this first phase indicated strong bottom currents and coarse sediments (e.g., Murray, 2006; Schönfeld, 2002), whereas C. lobatulus was previously 345 believed not to be endemic to this study area (Dessandier et al., 2018). The presence of the latter 346 was probably an indicator of transport from the estuary to the shelf, as this species is typically 347 found in Portuguese estuaries (Martins et al., 2015). The dominance of N. scaphum and A. 348

beccarii further suggested inputs from the estuary of phytoplankton or nutrients that may boost
local marine productivity, such as coccolithophores, as has been previously observed where the
Douro River flows into the ocean, where these species dominated the living fauna in late winter
(Dessandier et al., 2015). This may indicate an influx of relatively labile organic matter,
consistent with the progressive increase of faunal diversity during this period.

The second phase began with the Tagus mud belt establishment at around 2250 cal yr 354 355 BP, which was represented by the plot of PC1, when the Ca/Ti and Zr/Rb counts clearly 356 decreased and terrestrial elements (Fe/Ti, Pb/Ti, and BIT index, Fig. 4) increased. This suggests a deposition of finer sediments of terrestrial origin in the Tagus prodelta during this period. This 357 358 important change in sedimentary conditions toward muddier and organic-rich sediment (Martins et al., 2006) was observed in the Portuguese Margin earlier, at around 2000 cal yr BP 359 (Alt-Epping et al., 2009; Martins et al., 2007), and indicated the onset of the Tagus mud belt, 360 361 which was mainly composed of sediment exported from the Tagus estuary (Jouanneau et al., 1998). This increase of organic compounds was also highlighted by the large increase in the 362 363 Br/Cl ratio during this phase. The reduction of bottom water currents driven by decreasing wind and more humid conditions have been postulated as the physical processes that triggered a 364 strong Tagus River export of sediment and led to the formation of the mud belt around this time 365 (Alt-Epping et al., 2009). The mud belt formation was responsible for the increased 366 accumulation of organic matter, especially during Phase II, corresponding to the increased 367 levels of Br/Cl, TOC, and TN content (Fig. 4). The strong negative loading of both $\delta^{13}C_{TOC}$ and 368 TOC on PC2 might suggest that the major supply of organic matter was driven by marine 369 production, particularly during the strong upwelling conditions that characterized Phase II. The 370 $\delta^{18}O_{bf}$ in this area was primarily controlled by salinity, with a reduced temperature effect, 371 showing an increase in salinity during upwelling events (Lebreiro et al., 2006). This phase was 372 characterized by a large increase of C. carinata, V. bradyana, and B. marginata abundances, 373

indicating that higher trophic levels occurred in the sediments, caused by high organic matter 374 375 content in the mud belt. In this area, the active upwelling period corresponds to the most eutrophic conditions, mainly highlighted by the abundance of C. carinata, associated with 376 marine organic matter (Br/Cl and $\delta^{13}C_{TOC}$; Fig. 6). Cassidulina carinata was already interpreted 377 as highly dominant in an active upwelling context and adapted to cold, nutrient-rich waters 378 379 (Bartels-Jónsdóttir et al., 2006; Martins et al., 2006). Together with this species, V. bradyana, B. marginata, H. balthica, and U. bifurcate have essentially been found only in the dead 380 381 community and with almost no occurrence in the late winter (Dessandier et al., 2016). The 382 increase in V. bradyana abundance correlated with the onset of Phase II and the mud belt. This result corroborated this species' need for rich trophic conditions and suggested that the second 383 phase stabilized with the onset of strong upwelling conditions. Nonion scaphum and V. 384 bradyana were highly dominant during this interval, although N. scaphum was less abundant 385 than in Phase I. These two species live under organic-rich conditions and can tolerate anoxic 386 387 sediments (Fontanier et al., 2002; Barras et al., 2014), which may suggest that large terrestrial inputs in this area led to periodic anoxia. The upwelling events create ideal environmental 388 conditions for diatom blooms that resulted in the presence of other species during summer 389 390 periods (Dessandier et al., 2016). This seasonal production indicated a preference of these opportunistic species for summer periods, when the influence of upwelling is at its maximum. 391 During upwelling periods, diatoms are the major phytoplankton group responding to cold and 392 nutrient-rich ENACW. However, the preservation of this group as fossils is dependent on high 393 fluxes of individuals to the seabed (Abrantes et al., 1988), which constrains the use of this proxy 394 395 for environmental changes. Conversely, winter periods characterized by maximum continental runoff had evidence of coccolithophore blooms, which responded to stratified waters (Abrantes 396 and Moita, 1999). Since phytoplanktonic groups favor certain environmental conditions, these 397 phytodetrital sources for benthic foraminifera may partially explain the seasonal variation in 398

species composition. This result may be the consequence of preferences for the specific
component of seasonally deposited phytodetritus, as has been observed in the Antarctic (Suhr
et al., 2003).

During Phases II and III, $\delta^{13}C_{bf}$, representing the exported phytodetritus to the seafloor 402 (Curry et al., 1988), indicated higher primary production, whereas the increase of $\delta^{15}N_{bulk}$ may 403 illustrate nutrient degradation or a stronger influence of estuarine sources of organic matter 404 (Owens, 1985). Phase III was characterized by a change in organic matter sources from 405 predominantly marine origin (negative loads on PC2) to predominantly terrestrial origin 406 (positive loads on PC2). This period showed a drop in foraminiferal diversity corresponding to 407 the dominance of C. carinata (up to 43 %). The increase in abundance of the deep infaunal 408 species at around 1000 cal yr BP suggested this is when the enrichment in organic matter of the 409 410 prodelta occurred and might indicate episodic periods of dysoxia or even anoxia related to 411 potential eutrophication, such as the occurrence of G. affinis and C. oolina, well known to live in highly eutrophic conditions, often below the oxic sediments (Jorissen et al., 1998; Mojtahid 412 et al., 2010a). The faunal evolution, in terms of assemblages and diversity within Phases II and 413 III, revealed other controlling factors independent of the presence of the Tagus mud belt, 414 represented by the PC2 (Fig. 6B). An increase in pollutants during this period was thought to 415 416 occur concomitantly with the disappearance of *H. balthica* and appearance of *E. scaber*, as has been observed off Iberian rivers (Diz et al., 2002; Bartels-Jónsdóttir et al., 2006). The increase 417 418 of pollutants indicated by the appearance of *E. scaber* also fits the results of Alve (1995), who 419 described *E. scaber* as a pollution-tolerant species under aquaculture influence.

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421 <u>5.2. Reconstruction of environmental evolution in the Tagus prodelta during the last 5750 years</u>

422 <u>5.2.1. First phase (5750-2250 cal yr BP): High Tagus River discharge</u>

The modern sediment cover in the Tagus prodelta is mainly supplied by terrestrial silts 423 424 and clays exported by Tagus River discharge (Jouanneau et al., 1998). The influence of sea level changes on the Iberian Margin is no longer significant on the sedimentation after ~ 7000 425 426 yr BP (Vis et al., 2008). The sedimentary evolution of the Tagus prodelta was mainly controlled by Tagus River discharge and impacted by climatic changes after this period. This area was 427 particularly characterized by very humid conditions between 6500 and 5500 yr BP, 428 corresponding to the African Humid Period (deMenocal et al., 2000; Renssen et al., 2006; Vis 429 et al., 2010). The coarse sediments observed during Phase I, highlighted by both environmental 430 and faunal evidence, are in good agreement with conditions described by Rodrigues et al. 431 432 (2009), who interpreted intense deforestation and soil destabilization as factors responsible for the increased current velocity of the Tagus River. These coarse sediments with low organic 433 content and of marine origin were similar to sediments of modern inner shelf conditions 434 435 observed down to 50 m water depth in the Portuguese Margin (Schmidt et al., 2010). These conditions are synthesized in Fig. 7A and demonstrate that when the Tagus River discharge was 436 437 large, it prevented any deposition of fine material on the mud belt, resulting in low sedimentation rates. The fine sediment deposits of terrestrial origin were probably transported 438 further offshore during this phase. The position of Core 64PE332-30-2 might have been a zone 439 440 of fine sediment bypass similar to what is currently observed on the inner shelf. This explains the low organic content, the low BIT index, and the $\delta^{13}C_{TOC}$ that signaled the marine origin of 441 the sediments. Similar conditions were observed for this period in the Galicia mud deposit 442 (Martins et al., 2007; Bernárdez et al., 2008) and off the Guadiana River (Mendes et al., 2010) 443 and were believed to indicate a strong hydrodynamic regime. Tagus River floods have been 444 reconstructed in the lower Tagus River valley between 4900 and 3500 yr BP and were 445 associated with strong deforestation during this period (Vis et al., 2010). 446

The transition between Phases I and II was marked by a shift between the faunal and 447 448 environmental signals, between 2600 and 2250 cal yr BP (Fig. 5). The species that first reacted to this, C. carinata, started to increase in abundance, resulting in a decrease in the relative 449 abundances of the other species, including the taxa indicators of river discharge. This reflected 450 the very opportunistic nature of C. carinata and provided a more accurate signal of the 451 environmental change than the geochemical parameters, which merely recorded the mud belt 452 453 conditions. A progressive decrease of fluvial influence was visible following the strong decline of A. beccarii and N. scaphum, which has also been observed during the last 3000 years within 454 the Ría de Vigo (Diz et al., 2002). This may be the transition between colder and wetter 455 456 Subboreal conditions and warmer, dryer Sub-Atlantic conditions at 3000 yr BP (Alt-Epping et al., 2009; Bernárdez et al., 2008). The Sub-Atlantic period was associated with the reduced 457 influence of winds on the Iberian Margin (Martins et al., 2007), a reduction in hydrodynamic 458 459 marine currents, and the collection of fine sediments in the Galicia mud deposit. These conditions were responsible for the lateral movement of the mud deposit on the shelf, with the 460 construction of the mud belt at the beginning of Phase II (2250 cal yr BP). The accumulation 461 of muddy sediments on the shelf off the Tagus River at ~ 2000 years BP was synchronous with 462 the establishment of the mud belt off the Douro River (Drago et al., 1998). This coordination 463 464 suggests a response to regional rather than local change in the climate, responsible for a dryer period and an increase of clay accumulation closer to the Tagus River mouth, due to the 465 decrease in Tagus discharge. 466

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468 <u>5.2.2. Second phase (2250-1250 cal yr BP): High upwelling intensity</u>

After the onset of the Tagus mud belt at ~ 2250 cal yr BP, two alternative regimes prevailed, one characterized by intense upwelling and the other by strong river discharge, as summarized in Fig. 7B. During this phase, TN, $\delta^{13}C_{bf}$, and $\delta^{15}N_{bulk}$ showed an increase in

productivity, probably due to a change in Tagus River flux conditions. However, this was not 472 473 recorded by all environmental parameters. The faunal results could indicate that the organic matter supply during Phase II was related to upwelling events more than Tagus River inputs. 474 The decrease in the C/N ratio was not in agreement with $\delta^{13}C_{TOC}$ data, making it difficult to 475 evaluate the organic matter source. The $\delta^{13}C_{TOC}$ and the C/N ratio are often used to determine 476 the sources of organic matter (e.g., Hedges and Parker, 1976; Peters et al., 1978; Alt-Epping et 477 al., 2007). However, the C/N ratio is affected by the preferential remineralization of nitrogen in 478 479 marine sediments or nitrogen sorption onto clay minerals (Schubert and Calvert, 2001), and the $\delta^{13}C_{TOC}$ signal could be from a mixture of C3 and C4 plants, mimicking the isotopic signal of 480 marine algae (e.g., Goñi et al., 1998). Both of these indicators in the paleorecords might be 481 482 affected by organic matter degradation. Additionally, $\delta 15N_{bulk}$ could be affected in the study area by a higher influence of agriculture and pollution (Alt-Epping et al., 2009). Nevertheless, 483 $\delta^{13}C_{TOC}$ and the C/N ratio were useful to discriminate the marine and terrestrial sources of 484 organic matter in another study on the Portuguese Margin (Schmidt et al., 2010). In the present 485 study, only the C/N ratio seemed to be clearly affected by the early diagenesis. These organic 486 parameters were essential to compare the environmental signal with benthic foraminifera, and 487 488 their disagreement confirms the importance of using a multiproxy approach that included bio-489 indicators that are less affected by organic matter degradation.

Phase II began at 2250 cal yr BP with a low amount of terrestrial input (indicated by the negative loading of PC2; Fig. 6) and lasted until 1800 cal yr BP, which corresponded to the RP (Lamb, 1985; Bernárdez et al., 2008). Despite the increase in eutrophy-tolerant species, the faunal diversity and BFN were low at the beginning of the RP. The deposition of contaminants and increased sediment accumulation rate may have limited faunal production during this period. Lebreiro et al. (2006) also observed a large export of terrestrial particles into the Tagus prodelta during the RP. This export was interpreted as a consequence of anthropogenic

activities, such as Roman gold mining, along the Tagus River. This export of terrestrial material 497 498 may have been enhanced by the NAO- phase that occurred during that period and led to intensified rainfall in the Portuguese Margin (Abrantes et al., 2005). Periods of NAO- have 499 500 strongly influenced Iberian river discharge, especially on the Tagus River (Trigo et al., 2004). The northern part of the Iberian Margin is marked by varying rainfall responses. This zone, 501 which is very close to the limit of the NAO influence, is known to alternate between positive 502 503 and negative correlation with the NAO and humid conditions (Alvarez and Gomez-Gesteira, 504 2006). Desprat et al. (2003) also identified warm and relatively humid conditions during the RP in the Ría de Vigo. A strong river regime such as this one was not observed off the Capbreton 505 506 in the bay of Biscay (Mojtahid et al., 2009), suggesting an anti-correlation of the NAO phases between the two study areas. 507

508 The end of Phase II showed a decrease in the Tagus River influence that corresponded 509 with an increase in positive loading of PC2, characterized by high marine organic matter content likely brought by active upwelling conditions. The increased abundance of C. carinata 510 511 corroborated this context in the middle of Phase II, corresponding to a period of intense upwelling activity, which has also been described in the same area in a study using benthic 512 foraminifera (Bartels-Jónsdóttir et al., 2006). The dominance of the opportunistic species C. 513 514 *carinata* constrained faunal diversity, as observed in other studies (Fontanier et al., 2003; Dessandier et al., 2018). However, C. carinata did not increase until the end of Phase II, 515 suggesting that the intensity of upwelling slowed down at the end of this phase. By contrast, 516 the Tagus mud belt built up, allowing an increased deposition of marine organic matter until 517 518 the end of Phase II. This difference could be the result of biased organic parameters (e.g., caused by early diagenesis). In addition, this study area was characterized by organic matter from 519 different sources, which were recorded as a mixture of marine and terrestrial compounds by the 520 organic parameters. What was revealed by the organic parameters as a record of upwelling 521

activity could instead result from high productivity throughout the entire year. Therefore,benthic foraminiferal species recorded the organic matter sources more accurately.

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525 <u>5.2.3. Third phase (1250 cal yr BP-present): Alternating upwelling and Tagus discharge</u> 526 <u>influence</u>

The beginning of Phase III (Fig. 7C), which corresponds to the DA, was followed by 527 the MWP, which occurred between 1100 and 600 yr BP, as reported in several studies 528 529 performed on the Portuguese Margin (e.g., Desprat et al., 2003; Rosa et al., 2007). The DA was described as a period characterized by strong upwelling activity off the Douro River (Rosa et 530 al., 2007). The MWP is also well known in the Portuguese Margin as a period characterized by 531 active upwelling conditions triggered by NAO+ conditions (Abrantes et al., 2005; Bartels-532 Jónsdóttir et al., 2006; Rosa et al., 2007; Rodrigues et al., 2009). The high primary productivity 533 534 was linked to upwelling events and created eutrophic conditions in sediments that allowed for less competition among B. spathulata, P. mediterranensis, Cribroelphidium gerthi, and H. 535 536 balthica. This period, characterized by a warm and dry climate, was influenced by anthropogenic activities and soil erosion (Rodrigues et al., 2009) that caused Fe and Pb to be 537 massively transported via Tagus River runoff to the prodelta. Dry soils restrained infiltration 538 and possibly triggered large flooding of the Tagus River (Benito et al., 2003). The increased 539 BIT index (Warden et al., 2016) and Fe/Ti and Pb/Ti ratios during the beginning of the MWP 540 suggested increased terrestrial input, corresponding with the increased loading of PC2 in the 541 first part of Phase III and the accumulation of finer sediments. 542

The LIA, between 600 and 100 yr BP, was characterized by the increase in terrestrial material, as shown by the abrupt increase in the BIT index and Fe/Ti and Pb/Ti ratios at 250 cal yr BP. This increase was also highlighted by the sharp decrease in PC2 (Fig. 6B), which could be linked to the Lisbon earthquake in 1755 AD (200 cal yr BP; Abrantes et al., 2008) or to

massive exports of fine sediments from the Tagus River (low Zr/Rb). This period has been 547 548 described as characterized by abrupt cooling and wet conditions (Bradley, 2000). The LIA was affected by high-frequency episodic Tagus River paleo-floods (Benito et al., 2003) and has been 549 550 associated with the transport of fine sediments from the continent via discharge from the Tagus River under NAO- conditions (Abrantes et al., 2005; Bartels-Jónsdóttir et al., 2006). During the 551 LIA, an abrupt decrease in the abundance of C. carinata may indicate that a reduction in 552 upwelling conditions occurred. V. bradyana, E. scaber, A. beccarii, and deep infaunas 553 554 increased, possibly as a consequence of episodic anoxia and the presence of contaminants. The presence of the deep infaunas that can tolerate refractory organic matter (Murray, 2006) 555 556 suggests a decrease in organic matter quality, as was also observed in the Galicia mud deposit during this period (Martins et al., 2006). This decrease in quality may be the major cause of the 557 decreased infaunal diversity that occurred between 1000 cal yr BP and the present. 558 559 Nevertheless, the faunal results suggested that intense upwelling periods also occurred during the LIA, as demonstrated by the increased abundance of C. carinata. This highlighted that this 560 561 period was unstable and characterized by several environmental changes. The results from this study were in good agreement with previous studies in the same area for the last 2000 years 562 (Abrantes et al., 2005; Bartels-Jónsdóttir et al., 2006; Alt-Epping et al., 2009), except for small 563 time shifts in the upwelling versus Tagus River discharge periods corresponding to the 564 transition between Phases I and II. These time shifts could be the consequence of different age 565 models. Bartels-Jónsdóttir et al. (2006) demonstrated that an "intense upwelling period" 566 occurred, followed by a "very intense upwelling period" during the MWP, and finally a large 567 Tagus River discharge during the LIA, all determined through the analysis of benthic 568 foraminiferal distribution. 569

570 There were some differences during the MWP and the LIA between the reconstructions 571 from this study and the results from Bartels-Jónsdóttir et al. (2006). These variations could be due to a different interpretation of certain species, specifically *N. scaphum*, which was determined to be controlled by upwelling in previous studies; by contrast, it was used as a river discharge proxy in this study, based on the ecological results from the study area (Dessandier et al., 2016). The use of environmental data combined with the composition of the major species in this study may allow a better interpretation of the mud belt onset than merely using benthic foraminiferal data. Our results were also in good agreement with the reconstruction of the Tagus River discharge and the upwelling strength reported in Abrantes et al. (2009).

579 Finally, during the last century, this area was contaminated by anthropogenic pollution, including numerous trace metals that were measured in excess in the Tagus estuary, such as 580 581 AS, Pb, Zn, Cu, and Cd (Caçador et al., 1996; Jouanneau et al., 1998). The catchment area was also influenced by anthropogenic contaminants, such as domestic sewage and industrial wastes 582 (e.g., petrochemistry, fertilizers, smelters; Carvalho, 1997). Our analyses were not performed 583 584 to identify the sources of anthropogenic pollution; however, the XRF signal shows a clear increase of several contaminants (such as Pb) in the most recent interval. We do not have 585 586 enough faunal resolution and environmental data to investigate the anthropogenic influence on benthic ecosystems, but we assume that the faunal assemblages respond to this anthropogenic 587 activity, as shown in other environments (e.g., Alve, 1995). The faunal distribution shows 588 589 increased abundances of deep infaunal species and of E. scaber but the disappearance of other species, such as *H. balthica*, as has already been observed as a consequence of anthropogenic 590 pollution in the Tagus prodelta (Bartels-Jónsdóttir et al., 2006). The last century was also 591 characterized by the construction of dams, which have likely influenced the sequestration of 592 organic matter in the estuary and changed the influence of the Tagus River discharge on the 593 shelf through increased input of finer material (Jouanneau et al., 1998) and increased correlation 594 with NAO phases (Trigo et al., 2004), all of which could have significant environmental 595 consequences that will be crucial to understand in the future. 596

598 **6.** Conclusions

The results of this study demonstrate the validity of using benthic foraminifera as bio-599 600 indicators of past river discharge and upwelling intensity in the Tagus prodelta during the last 5750 years. Major environmental changes, linked with late Holocene climatic variations, are 601 602 summarized by key benthic foraminiferal taxa data in Fig. 8. This study showed an additional 603 phase, between 5750 and 2250 cal yr BP, that has not yet been investigated in the Portuguese 604 shelf. This phase was characterized by coarser sediment cover and dominated mostly by N. scaphum and A. beccarii, suggesting a very dynamic Tagus River system that facilitated the 605 606 dispersion of coarse terrigenous particles onto the prodelta.

The progressive decrease of the Tagus River flow, resulting in the establishment of the modern Tagus mud belt, was the major process explaining the environmental changes that occurred before Phase II. The presence of a transition period (2500-2250 cal yr BP) at the end of Phase I was only supported by the composition of the benthic foraminifera; geochemical data did not reveal any environmental changes. This might be due to the extremely poor preservation of organic matter in coarse sediments (in which benthic foraminifera are fairly well preserved).

Phase II (2250-1250 cal yr BP) was characterized by fine continental deposits, responsible for a better food stock for benthic organisms throughout the record. This phase was marked by the increase of *C. carinata*, *V. bradyana*, and *B. marginata*, opportunistic species that mark the increase in upwelling intensity in this area, revealing the strongest upwelling activity during the DA and the MWP. The high organic matter stocks in the Tagus prodelta during this phase created more refractory organic matter, responsible for a faunal diversity decrease from 2250 cal yr BP to present.

Phase III (1250 cal yr BP-present) showed the disappearance of *B. spathulata*, *P. mediterranensis*, and *H. balthica* and the decrease of *A. beccarii* and *N. scaphum*, which may

be a consequence of a decrease in the organic matter quality and/or the occurrence of anthropogenic pollution, as indicated by benthic foraminiferal assemblages and the XRF data. Conversely, *E. scaber* increased, probably because of its pollution tolerance, and *C. carinata* remained dominant, likely due to the strong upwelling conditions. Benthic foraminifera responded accurately to record environmental and climate changes in the North Atlantic continental shelf and therefore could be used as a bio-indicator of environmental changes, such as changes in upwelling activity and river discharge, that were directly linked with the NAO.

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Figures

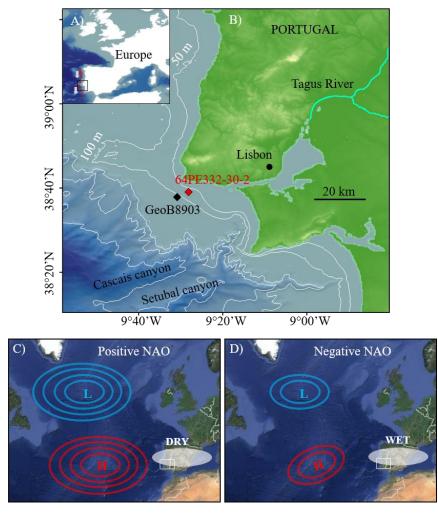


Fig. 1. A) General location map of the study area, with a schematic representation of the Portugal Coastal Counter Current (red arrow) and the ENACW (grey arrows). B) Detailed map of the core locations considered in this study. Red diamond: piston cores 64PE332-30-2, this study; black diamond: GeoB8903 (Abrantes et al., 2008). C) Positive and D) Negative phase of NAO with L = Iceland low-pressure system and H = Azores high-pressure system.

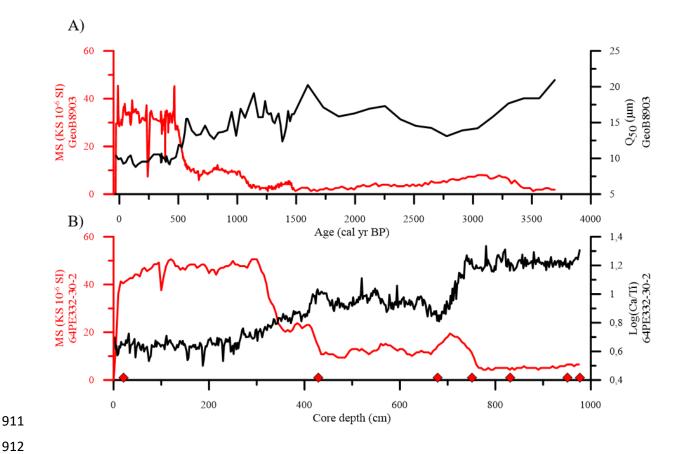


Fig. 2. Comparison of Ca/Ti and magnetic susceptibility (MS) of Core 64PE332-30-2 with grain

size (Q₅₀) and MS from Core GeoB 8903 (Abrantes et al., 2008; Alt-Epping et al., 2009). Red diamonds represent ¹⁴C data points.

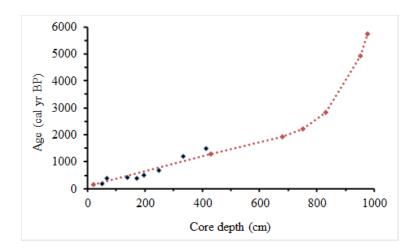
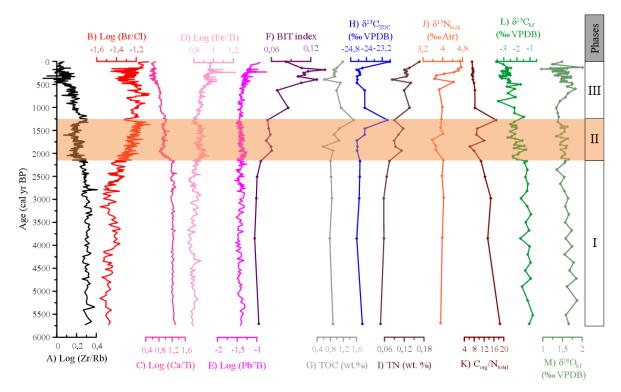


Fig. 3. Age model of Core 64PE332-30-2 based on AMS ¹⁴C data (Table 1). Blue diamonds represent the ¹⁴C data points of GeoB 8903.





922 Fig. 4. Distribution of environmental parameters for the last 5750 years. XRF data: A) Zr/Rb, B) Br/Cl, C) Ca/Ti, D) Fe/Ti and E) Pb/Ti. Organic measures: F) TN, G) TOC, H) C/N ratio, I)

923 $\delta^{15}N_{bulk}$, J) $\delta^{13}C_{TOC}$, and K) BIT index. Carbonate isotopes L) $\delta^{13}C_{bf}$ and M) $\delta^{18}O_{bf}$ analyzed

- 924
- for the species *N. scaphum*. The orange square indicates the limits of the three phases. 925
- 926

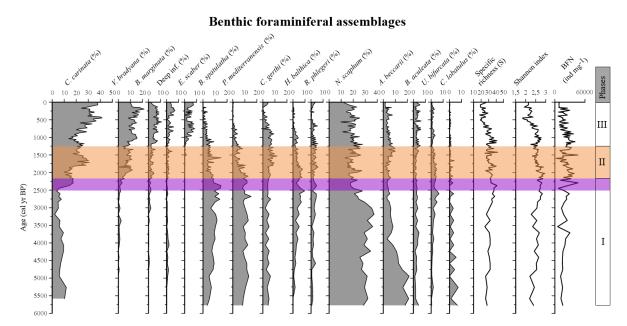


Fig. 5. Distribution of major species of benthic foraminifera (>5%) and specific richness from 928 the 101 samples of Core 64PE332-30-2 for the last 5750 years. Deep inf. = G. affinis + C. 929 oolina. The orange square indicates the limits of the three phases, and the purple square follows 930 931 the main change of the faunal distribution.

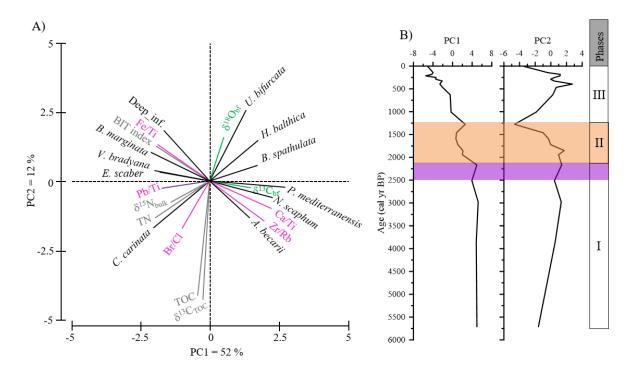




Fig. 6.A) PCA based on the major species percentages (in black), XRF data (in pink), organic
compounds (in grey), and benthic foraminiferal isotopes (in green). B) Score profiles of PC1
and PC2 as a function of time. The orange square indicates the limits of the three phases, and
the purple square follows the main change of the faunal distribution.

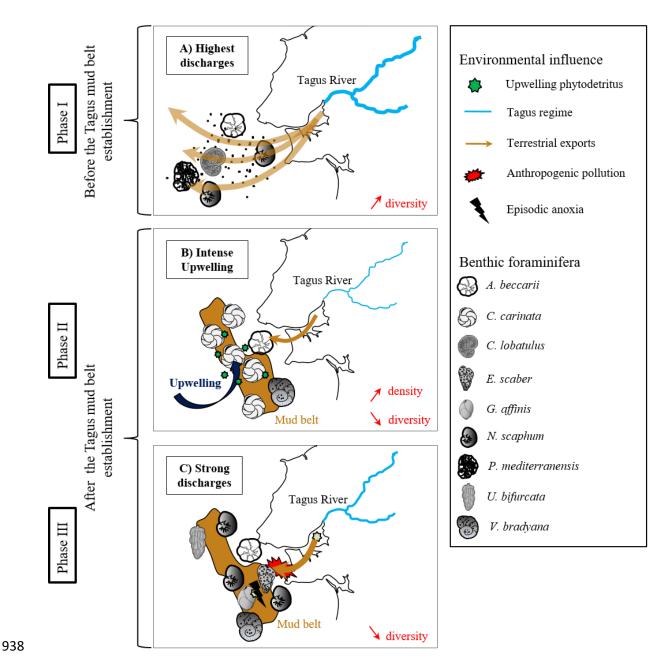


Fig. 7. Schematic representation of the three modes associated with the Tagus mud belt
establishment, Tagus River regime, and upwelling intensity. A) First phase, characterized by
very high Tagus River discharge. B) Second phase: mud belt establishment and very intense
upwelling activity. C) Alternating periods of strong upwelling activity and high Tagus River
discharge.

Key taxa	Faunal diversity	Benthic Productivity	Tagus vs. Upwelling	Climatic Period	Phase	
C. carinata, E. scabra, S. karreriana	Contaminant- tolerant species		High upwelling influence	PWP		
V. bradyana, A. beccarii, G. affinis, C. oolina	Contaminant- tolerant species		🕈 Tagus Discharges	LIA	III	
C. carinata, B. marginata	لم ل diversity		Upwelling and Tagus alternances	MWP		
A. beccarii, N. scaphum, C. carinata	 diversity diversity 		High upwelling influence	DA		
N. scaphum, V. bradyana	🖌 🖌 diversity		Mud Tagus exports	RP		
C. carinata	Stable diversity		¥ Tagus dynamic	SA		
A. beccarii, P. mediterranensis, N. scaphum			High Tagus regime	SB	I	

Fig. 8. Summary of the paleoenvironmental evolution of the Tagus River during the last 5750
years. RP = Roman Period, DA = Dark Ages, MWP = Medieval Warm Period, LIA = Little Ice
Age, PWP = Present Warm Period, SA = Sub-Atlantic Period, SB = Subboreal Period. The
orange square indicates the limits of the three phases, and the purple square follows the main
change of the faunal distribution.

Sediment	Lab no.	Core depth	Mean depth	Uncorrected	Analytical	Ages	Ages	Analyzed	Reference
core		interval	in core	AMS ¹⁴ C ages	error (±1σ)	$(\Delta \mathbf{R} = 0$ yr)($\pm 2\sigma$)		material	
		[cm]	[cm]	[cal yr BP]	[yrs]	[cal yr BP]	[cal yr BP]		
GeoB 8903	KIA30888	52-53	51	210	35	138-223	182	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	KIA30890	65-70	69	335	55	300-501	394	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	139-141	140	360	25	349-456	425	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	171-173	172	285	30	314-408	381	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	198-199	198	360	45	423-498	487	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	248-249	248	735	30	657-726	679	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	333-334	333	1260	35	1121-1282	1210	Foraminifera	Alt-Epping et al. (2009)
GeoB 8903	-	413-414	413	1600	40	1390-1567	1478	Foraminifera	Alt-Epping et al. (2009)
64PE332-30-2	BETA 348791	20-22	21	500	30	41 - 235	138	Gastropod	Warden et al. (2016)
64PE332-30-2	BETA 348792	428-430	429	1730	30	1219 - 1350	1284.5	Foraminifera	Warden et al. (2016)
64PE332-30-2	BETA 348793	678-680	679	2320	30	1848 - 2033	1940.5	Gastropod	Warden et al. (2016)
64PE332-30-2	VERA-51394	750-752	751	2530	70	2122-2293	2207.5	Gastropod	This study
64PE332-30-2	VERA-51395	830-832	831	3060	70	2749-2902	2825.5	Gastropod	This study
64PE332-30-2	VERA-51396	950-952	951	4690	70	4831-5007	4919	Gastropod	This study
64PE332-30-2	BETA 317911	976-978	977	5370	30	5644 - 5850	5747	Shell fragments	Warden et al. (2016)

953 Table 1. 14 C AMS dates of cores GeoB 8903 and 64PE332-30-2.

Note that ¹⁴C data from GeoB 8903 were from Alt-Epping et al. (2009) and reconverted using the CALIB V0.6 with the Marine13 calibration
 curve (Stuiver and Reimer, 1993).