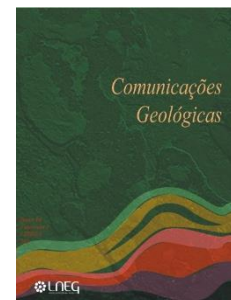


19th century environmental transformation of an industrialized estuary: the Avilés sedimentary record (Asturias, N Spain)

Transformação ambiental de um estuário industrializado no século XIX: o registo sedimentar de Avilés (Asturias, N Espanha)



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Abstract: The Avilés estuary is one of the most important industrial ports in northern Spain, whose natural characteristics have been largely altered since the 1830s. Here, the environmental transformation of this estuary during the 19th century is explored using a multi-proxy approach including benthic foraminiferal content, sedimentology (grain-size), trace metals and short-lived radionuclides (²¹⁰Pb and ¹³⁷Cs) analysed from a 50-cm-long sediment core and a surface sample collected from the middle part of the estuary in the Pedro Menéndez Channel. The obtained results provide evidence that indicate an evolution from a naturally-driven environment with high a marine influence towards a more restricted setting under brackish conditions. The observed environmental change is interpreted as being mostly derived from physical modifications linked to the marsh reclamation and channelling starting in 1833 and intensified since 1860. This study provides a preindustrial environmental reference framework for future studies in coastal areas of the NW Atlantic Iberian margin.

Keywords: Foraminifera, sedimentology, geochemistry, reclamation, channeling.

Resumo: O estuário de Avilés é um dos portos industriais mais importantes do norte de Espanha, cujas características naturais foram bastante alteradas desde 1830. A transformação ambiental deste estuário durante o século XIX é estudada usando uma abordagem *multi-proxy* incluindo foraminíferos bentónicos, sedimentologia (dimensão do grão), metais traço e radionuclídeos de vida curta (²¹⁰Pb e ¹³⁷Cs) analisados a partir de sedimento de um testemunho de sondagem com 50 cm de comprimento e uma amostra recolhida à superfície na parte média do estuário do Canal de Pedro Menéndez. Os resultados obtidos fornecem evidências que indicam evolução de um ambiente com elevada influência marinha para um ambiente mais restrito em condições salobras. A alteração ambiental observada é interpretada como resultado de modificações físicas ligadas à recuperação e canalização de pântanos a partir de 1833, e intensificada desde 1860. Este estudo fornece um exemplo de referência ambiental pré-industrial para estudos futuros em áreas costeiras da margem NW Ibérica do Atlântico.

Palavras-chave: Foraminíferos, sedimentologia, geoquímica, recuperação, canalização.

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1. Introduction

Cantabrian intertidal flats have been witnessing different hydrological and environmental changes derived from the anthropogenic pressure since the beginning of the regional Industrial Revolution in the late 19th century and, more intensively, since 1950 (*e.g.* Cearreta *et al.*, 2002; Irabien *et al.*, 2020). While natural processes including tides, floods and both continental and marine sediment supplies have controlled the sedimentary and morphological dynamics of estuarine environments, these ecosystems have increasingly become more vulnerable to human-driven disturbances due to industrial and urban development (Kennish, 2002). In fact, anthropogenic activities have been reported to be leading to profound ecological disruptions linked to several biological, chemical and physical imprints (Zalasiewicz *et al.*, 2019). As benthic foraminiferal assemblages have very specific environmental requirements, as well as a wide distribution and a good preservation potential in coastal areas (Murray, 2006), such changes can be reconstructed through the study of this important group of shelly microfossils present in the estuarine sedimentary records.

The number of environmental reconstructions based on benthic foraminifera assemblages has considerably increased over the last decades in the southern Bay of Biscay (*e.g.* Cearreta *et al.*, 2002; García-Artola *et al.*, 2016; Irabien *et al.*, 2018), although important gaps still exist concerning the understanding of their diversity composition, abundance and ecological dynamics in the northern Atlantic Iberian margin, particularly on its western sector. Here, we present the first microfaunal study of the benthic foraminifera recorded in the Avilés estuary (Asturias), in terms of species diversity and relative and absolute abundances. Foraminiferal data have been combined with sedimentological (grain size), geochemical (trace elements) and radioisotopic (²¹⁰Pb and ¹³⁷Cs) proxies to further disentangle the full complexity inherent to the recent environmental transformation history of this estuary.

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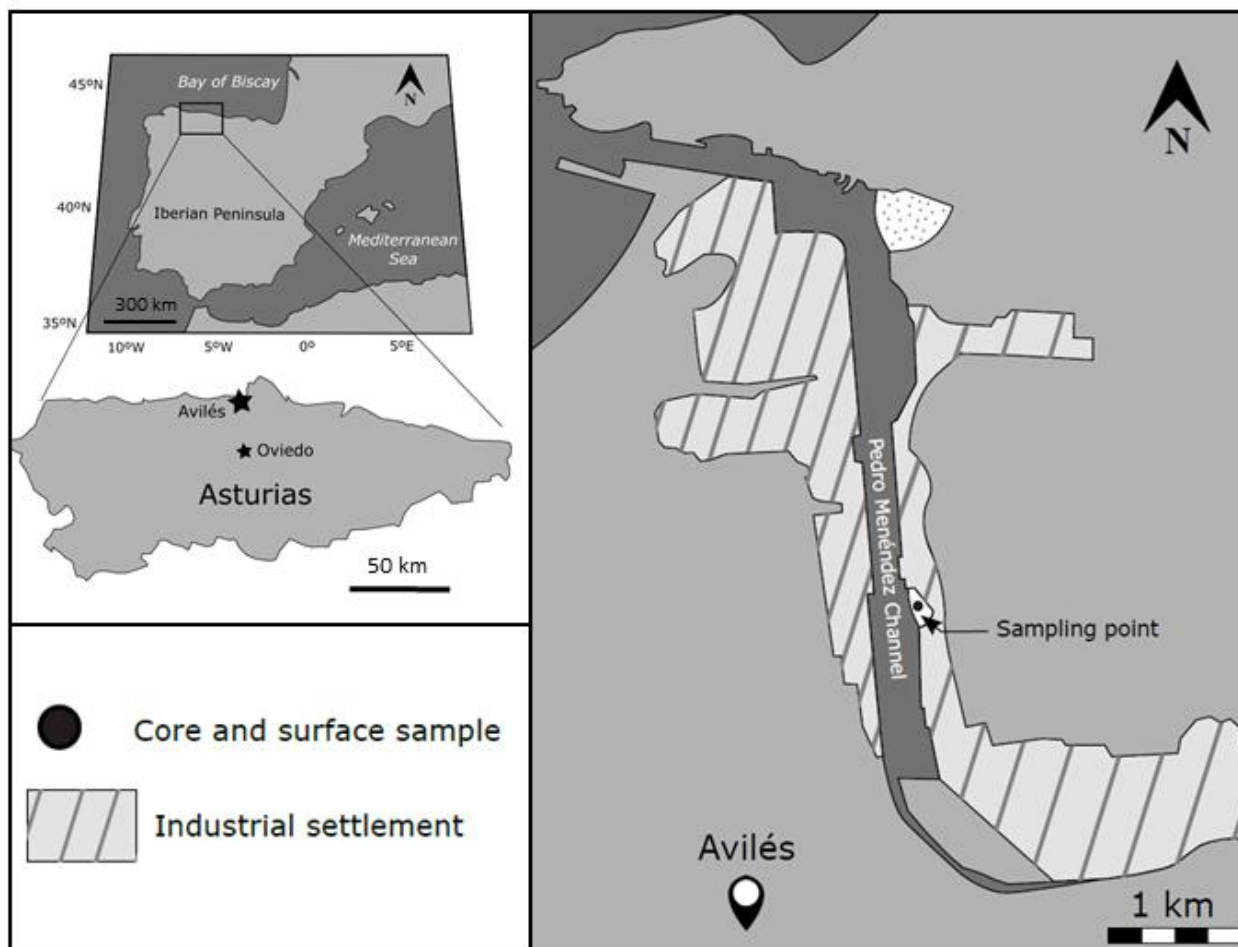


Figure 1. Geographic location of the Avilés estuary (N Spain) showing the sampling point.

Figura 1. Localização geográfica do estuário de Avilés (N Espanha), com indicação dos pontos de amostragem.

The Avilés estuary (43° 34' N, 5° 55' W) is located in the midwestern part of Asturias, NW Iberia, at 24 km distance of Oviedo, Spain (Fig. 1), near the Avilés industrial city. This mesotidal estuary is drained by various coastal rivers located on its left bank (López Peláez and Flor, 2008) and it is considered one of the main ports in the North of Spain. The original estuarine surface area was rapidly reduced for industrial and urbanisation purposes during the late 19th and 20th centuries, being about 1.8 km² nowadays.

2. Materials and Methods

Sediment cores were drilled in October 2020 from the intertidal eastern middle part of the Avilés estuary, in the Pedro Menéndez Channel. Two 50-cm-long core replicates were extracted with PVC tubes. Microfaunal and geochemical studies were performed on one replicate, whereas the second replicate was used for radiometric analysis. In order to study the modern assemblage of foraminifera, the top 0-1 cm sediment interval (80 cm³) was placed in a plastic jar containing ethanol, to prevent decay of the protoplasm (Murray, 2006).

The benthic foraminiferal analysis was performed at a resolution of approximately 5-cm intervals, from 0-1 cm to 48-49

cm depth, with 11 samples. Samples were sieved using a 2 mm mesh, to remove the larger clasts, and further wet sieved with 63 µm mesh to remove clay and silt fractions. Subsequently, samples were dried at 40 °C and the foraminiferal tests were concentrated by flotation in trichloroethylene. After being washed, a solution of 1 g/L of Rose Bengal (to differentiate stained alive individuals from uncoloured dead tests) was added to the content of the surface sample and left for an hour, following Walton (1952). The sediment sample was sieved and washed again to eliminate the stain solution, dried at 40°C and concentrated in trichloroethylene as described by Murray (2006). Identification mainly followed Murray (1979), Loeblich and Tappan (1988) and more recent taxonomic nomenclature updates provided in the World Register of Marine Species (WoRMS). A minimum of 300 tests was obtained and studied under a stereoscopic binocular microscope using reflected light to identify each assemblage (buried, surface dead and alive). Results were expressed as relative percentages of the total number of foraminiferal tests counted in every sample. Absolute abundances (foraminiferal density) in the core were expressed as the number of tests per 15 g of bulk dry sediment and grouped (very low, low, moderate, high, very high) according to the quantification of absolute abundances of foraminiferal tests for the estuaries of the eastern Cantabrian coast presented in García-

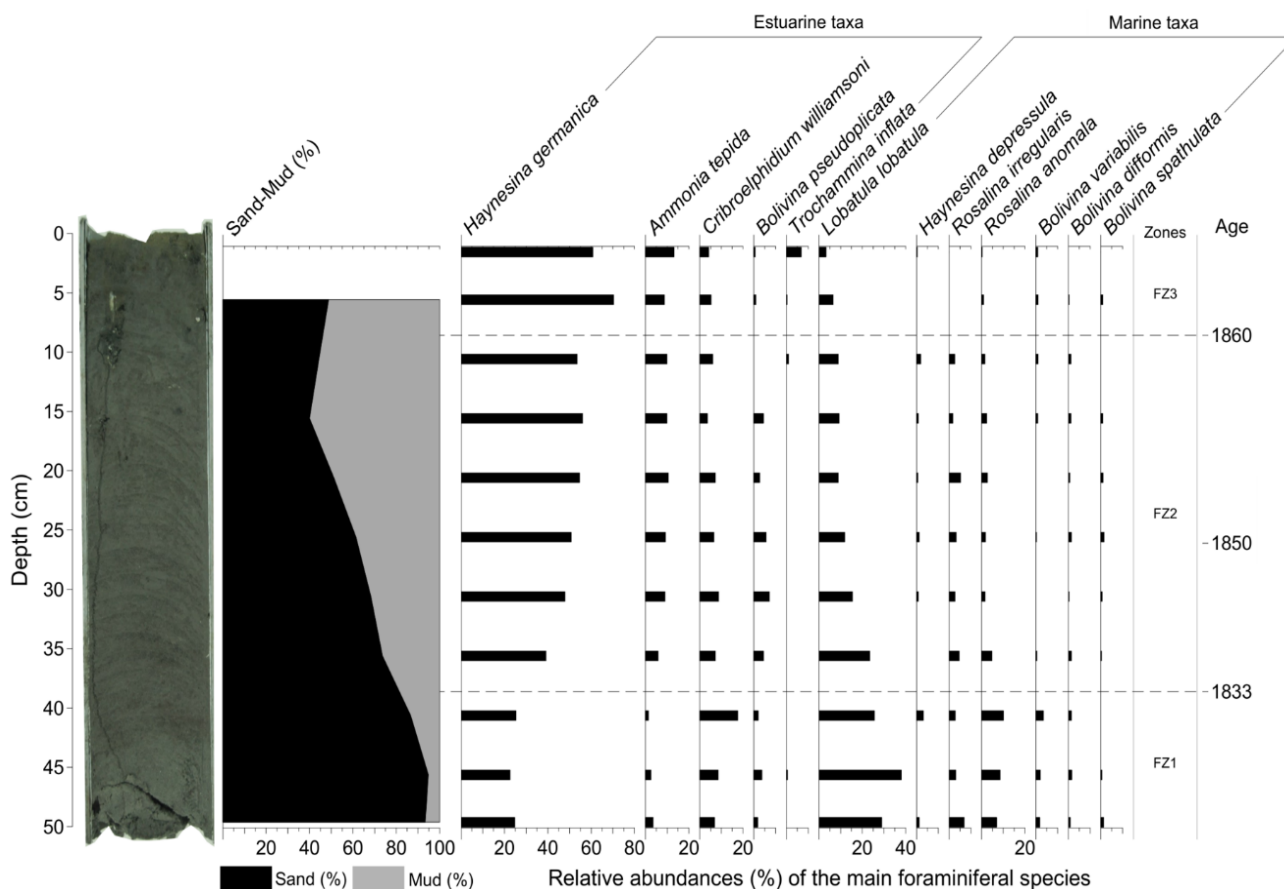


Figure 2. Sediment core photograph, sand (black) and mud (grey) content (%), foraminiferal stratigraphy diagram of selected taxa (> 2%) and Foraminiferal Zones (FZ).

Figura 2. Fotografia do testemunho de sondagem, proporção de areia (preto) e lama (cinzento), diagrama estratigráfico das espécies de foraminíferos (> 2%) e Zonas de Foraminíferos (FZ).

Artola *et al.* (2016). For the modern assemblages, results are referred to a total volume of 80 cm³. The Species Richness (S), Shannon-Wiener (H') and Fisher's alpha (α) diversity indexes were calculated using the software PAST version 3.26 (Hammer *et al.*, 2001). For the palaeoenvironmental reconstruction, only the species recording > 2% in at least one sample were considered.

An amount of 0.5 g of sediment sampled from the core and from the modern sediment samples was mechanically homogenised using an agate mortar and pestle. Trace metals concentrations were analysed in Activation Laboratories Ltd. (Actlabs, Ontario, Canada). The samples were digested with aqua regia for two hours at 95 °C, after which the samples were cooled, diluted with deionized water and analysed by Inductively Coupled Plasma-Optical Emission Spectrometry (ICP-OES). Isotopic activities analysis of both natural (²¹⁰Pb) and anthropogenic (¹³⁷Cs) radionuclides was performed by gamma spectrometry at the University of Cantabria, using a low-background high purity HPGe detector. The isotopic activities of the natural and artificial short-lived radiotracers were further used to assign an age to each sedimentary interval. The chronological model established by ²¹⁰Pb decay was validated with the activity of ¹³⁷Cs. This anthropogenic nuclide was formed as a result of the atmospheric testing of nuclear weapons, reaching a globally recorded peak

concentration in 1963-1964 CE, when the maximum of thermonuclear weapons' testing occurred (Hancock *et al.*, 2014).

3. Results

Gamma spectrometry analysis did not detect the presence of ¹³⁷Cs artificial radionuclide concentrations nor an excess of ²¹⁰Pb in the core samples and, therefore, establishing an accurate chronology is complex.

In total, > 3800 tests were counted and taxonomically classified. A total of 46 taxa were identified, although only 14 species were > 2% in at least one sample. The sediment core sequence was further divided into three different zones (Fig. 2, Table 1), based on its microfossil contents, named *Foraminiferal Zones* (FZ):

Foraminiferal Zone 1 (FZ1, 50-37 cm): Very high number of foraminiferal tests, varying from ca. 3200 to 9400 test/15 g, and high sand contents (range 86-95%). More than 94% of the foraminiferal tests are hyaline and up to 53% are allochthonous (marine) taxa. *Lobatula lobatula* (Walker and Jacob) (25.6-38.1%), *Haynesina germanica* (Ehrenberg) (22.5-25.3%), *Cribroelphidium williamsoni* (Haynes) (7-17.5%) and *Rosalina anomala* Terquem (7-10%) are the main species. *Rosalina*

Table 1. Summary of the microfaunal, sedimentological and geochemical characteristics of the Avilés estuary sediment core. The single value represents the median while the range is represented by values in parentheses. Metal concentration values are reported in $\text{mg}\cdot\text{kg}^{-1}$. Species are expressed as absolute numbers, marine and main taxa abundances and sand-mud data as percentages, and foraminiferal density as the number of tests/15 g of dry sediment.

Tabela 1. Resumo das características da microfauna, sedimentologia e geoquímica do testemunho de sondagem do estuário de Avilés. O valor único representa a mediana enquanto que o intervalo é representado por valores em parênteses. Os valores de concentração de metais estão representados em $\text{mg}\cdot\text{kg}^{-1}$. As espécies estão representadas em valores absolutos, as abundâncias principais e marinhas e os dados areia-lama em percentagens, e a densidade de foraminíferos como número de testes/15 g de sedimento seco.

Foraminiferal Zones (FZ – cm)	Foraminiferal data	Sedimentology	Geochemistry	Environment and driving processes	Chronology
FZ3 (7-0)	Species 18 (17-19)	Sand 51	Pb 18 (11-25)	High intertidal flat to low vegetated marsh - General channelling of the estuary	Post-1860s
	Marine taxa 10.7 (9-12.5)	Mud 49	Zn 73.5 (46-101)		
	Foraminiferal density (< 2000)	Frequent SCP	Cr 14.5 (12-17)		
	<i>H. germanica</i> 65.6 (61-70)				
	<i>A. tepida</i> 10.9 (8.7-13)				
	<i>T. inflata</i> 3.6 (0.3-7)				
FZ2 (37-7)	Species 22 (18-25)	Sand 56.4 (44.3-73.6)	Pb 10.1 (6-13)	Low to high intertidal flat - Marsh reclamation and canalization of the estuary	1830s-1860s
	Marine taxa 30.5 (24.1-45)	Mud 43.4 (26.4-55.7)	Zn 40 (25-53)		
	Foraminiferal density 11,619 (3724-18,659)	25-24 cm first appearance of SCP	Cr 16.1 (11-20)		
	<i>H. germanica</i> 42.3 (39-56)				
	<i>L. lobatula</i> 13 (9-23.5)				
	<i>A. tepida</i> 9 (5.8-10.6)				
FZ1 (50-37)	Species 25 (23-28)	Sand 91.5 (86-95)	Pb 4.6 (3-8)	Low intertidal flat - Natural conditions	Pre-1830s
	Marine taxa 58.7 (53.7-63.7)	Mud 8.4 (5.2-13.4)	Zn 18.3 (13-29)		
	Foraminiferal density 6490 (3197-9444)	No SCP	Cr 7.3 (5-12)		
	<i>L. lobatula</i> 30.9 (25.6-38.1)				
	<i>H. germanica</i> 24.2 (22.5-25.3)				
	<i>C. williamsoni</i> 11 (7-17.5)				
	<i>R. anomala</i> 8.5 (7-10)				

irregularis (Rhumbler) (2.8-7%) is a secondary taxon, while species with accessory abundances include *Ammonia tepida* Cushman (1.4-3.6%), *Haynesina depressula* (Walker and Jacob) (0-3.16%) and *Bolivina* spp. (6-8.3%) among others. Diversity indexes are relatively high ($S = 23-28$, $H' = 2-2.3$, $\alpha = 5.5-6.3$). Pb (3-8 $\text{mg}\cdot\text{kg}^{-1}$), Zn (13-29 $\text{mg}\cdot\text{kg}^{-1}$) and Cr (5-12 $\text{mg}\cdot\text{kg}^{-1}$) profiles exhibit very low values along the studied interval.

Foraminiferal Zone 2 (FZ2, 37-7 cm): The foraminiferal density and sand contents decrease towards the top from ca. 18,700 to 3700 test/15 g and from 73.6% to 44.3% respectively, whereas the mud content increases (from 26.4 to 55.7%). Hyaline tests are highly dominant (almost 100%) while the allochthonous taxa abundances vary from 45 to 24% at the top. *Haynesina germanica* (39-56%), *L. lobatula* (9-23.5%) and *A. tepida* (5.8-10.6%) are the dominant species in this interval. *Rosalina* spp. (4-9.5%) and *C. williamsoni* (3.6-8.7%) are secondary taxa. Diversity decreases ($S = 18-25$, $H' = 1.7-1.9$, $\alpha = 4.1-5.9$) while trace metals present similar trends with respect to FZ1 (Pb = 6-13 $\text{mg}\cdot\text{kg}^{-1}$, Zn = 25-53

$\text{mg}\cdot\text{kg}^{-1}$, Cr = 11-18 $\text{mg}\cdot\text{kg}^{-1}$). The 25-24 cm interval marks the first appearance of some spheroidal carbonaceous particles (SCP).

Foraminiferal Zone 3 (FZ3, 7-0 cm): It is formed by sandy muds (average mud content is 51%) with a lower abundance of benthic foraminifera (ca. 2000 test/15 g). Hyaline tests are dominant (90-100%), but the abundances of agglutinated foraminifera increase to more than 10% in the uppermost sample. Micropalaeontological content is mainly composed of *H. germanica* (61-70%) and *A. tepida* (8.7-13%). The most remarkable secondary species are *Trochammina inflata* (Montagu) (0.3-7%) and *L. lobatula* (3.3-6.5%). Diversity decreases with respect to the previous zones ($S = 17-19$, $H' = 1.2-1.5$, $\alpha = 3.8-4.4$) as well as the abundance of marine taxa (9-12.5%). Geochemical features of this last interval stand out because of higher concentrations of Zn (46-101 $\text{mg}\cdot\text{kg}^{-1}$), while SCP are frequent in this interval.

The living assemblage is characterized by low abundance (108 individuals/80 cm^3) and very low diversity ($S = 4$, $H' = 0.7$, $\alpha =$

0.8). The main taxa are *H. germanica* (80.6%) and *A. tepida* (10.2%), whereas *C. williamsoni* (6.5%) and *Quinqueloculina seminula* (Linnaeus) (2.8%) are present as secondary species.

4. Discussion

The micropalaeontological content of *FZ1*, mainly formed by a substantial percentage of sandy sediments (median 91.5%) and marked by a very high foraminiferal density dominated by marine taxa (e.g. *L. lobatula*, *Rosalina* spp. and *Bolivina* spp.) and a scarce number of estuarine tests indicates a highly marine-influenced intertidal environment where tidal currents favoured the transport of sediments from the outer estuary into its inner zone. According to Murray and Hawkins (1976), the high proportion of marine foraminifera (30-70%) indicates that *FZ1* was deposited in a low intertidal flat environment. Moreover, trace metal concentrations in this zone are below the regional geochemical background established for coastal areas (Cearreta *et al.*, 2002), indicating a non-polluted depositional environment, characteristic of preindustrial contexts. The *FZ2* features, characterized by higher abundances of brackish taxa (*H. germanica* and *A. tepida*), a high abundance of marine species and an increase in fine-grained sediments, indicate a transition from a low to a high intertidal flat environment (Murray and Hawkins, 1976), while still under important marine influence. Finally, the *FZ3* is mainly formed by *H. germanica* and *A. tepida*, both typically found in great abundances within high intertidal flats and low vegetated marshes, indicating an evolution towards a more confined environment. Furthermore, the presence of *T. inflata* (ca. 7%), a dominant taxon in vegetated inter- and supratidal marshes together with an important percentage of fine-grained sediments, suggest more marginal conditions. The living assemblage presents low diversity and abundance, being exclusively made of typical estuarine taxa (e.g. *H. germanica*, *A. tepida*, *C. williamsoni* and *Q. seminula*), indicating more brackish and highly confined environmental modern conditions compared to previous recorded environmental stages.

Former geochemical studies have shown current anomalous concentrations of potentially toxic elements in the Avilés industrial estuary, mainly Zn, due to the anthropogenic pollution derived from metallurgical and mining discharges (Sierra *et al.*, 2014). Nevertheless, the features of the analysed sediment record (an overall very high foraminiferal density and diversity, and low metal concentrations) are characteristic of preindustrial stages and also of environmentally recovered estuaries (Cearreta *et al.*, 2002; García-Artola *et al.*, 2016). Adding to the micropalaeontological and geochemical features of the analysed record, its total absence of short-lived radionuclides suggests an interval prior to the final 19th century. Thereby, we propose that our observations can be chronologically framed in a preindustrial stage. More precisely, since the SCP have been generated by the industrial combustion of coal at high temperature (Swindles *et al.*, 2015), their first appearance at 25-24 cm depth suggest that this interval corresponds approximately to the 1850s, when the *Real Compañía Asturiana de Minas* started metallurgical activities near the estuary (López-Peláez and Flor, 2008).

Despite not finding a pollution imprint in this record, there is a probable underlying anthropogenic process driving the observed micropalaeontological and sedimentological changes. While historical information indicates that the Avilés estuary has undergone different human-driven modifications since the Middle Ages (López Peláez, 2017), this estuary is considered to have been mainly naturally-driven until the 1830s, when major

anthropogenic geomorphological shifts started (López Peláez and Flor, 2008). In 1833 small factory walls began to be built upstream and in the inner part of the intertidal flats, in order to channelling the estuary and desiccating large marshes for agricultural purposes (López Peláez, 2017). Later, in 1858, the Spanish engineer Pedro Pérez de la Sala designed a plan consisting of the general channelling of the estuary by building factory walls located at the mouth of the estuary (Ruiz Seisdedos and Navarro Bidegain, 2002). This modification process started in 1860 and was completed by 1900, determining the final morphology of the main channel (López Peláez and Flor, 2008; López Peláez, 2017). The evolution of the Avilés estuary towards a more brackish and muddier environment since the beginning of *FZ2* is likely to be derived from these initial land reclamations and physical modifications (Table 1), resulting in impacts on the microfaunal assemblages and causing the decline of the sandy sediments. In fact, it has been previously observed in other estuaries from the Cantabrian coast that land reclamation of marshes reduces significantly the amount of sand in recent sedimentary records (Cearreta *et al.*, 2013; García-Artola *et al.*, 2016). Finally, the tendency towards a more brackish environment intensified since 1860, when the marine influence was reduced in response to the general estuary channelling process, possibly coinciding with the beginning of *FZ3*.

5. Conclusions

In this work, we have reconstructed the preindustrial environmental transformation of the Avilés estuary during the 19th century using a multiproxy approach (benthic foraminifera, sedimentology, geochemistry, radionuclides) to investigate a sedimentary core collected in the region. Results from the foraminiferal analysis indicate an evolution towards a more brackish and confined depositional environment driven by desiccation and morphological modifications that affected the marsh in response to agriculture-related activities undertaken in the region since 1833 and intensified since 1860, including the construction of factory walls leading to the channelling and the reduction of the marine influence inside the estuary. This work contributes to establish environmental reference conditions previous to the anthropogenic impacts that occurred since the regional Industrial Revolution in the late 19th century. This is crucial for future regional studies on the western sector of the North Atlantic Iberian margin, an area historically characterized by intense mining and industrial activities.

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