



Recent environmental and morphosedimentary evolution of the mining-impacted Nalón Estuary (Asturias, N Spain): Disentangling natural and anthropogenic processes

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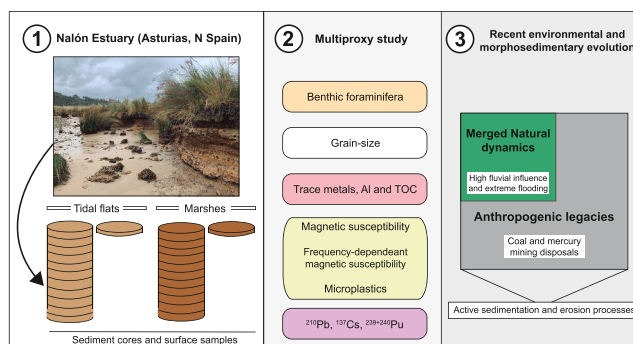
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HIGHLIGHTS

- Natural and human driving forces interacted influencing sedimentation-erosion processes.
- High sedimentation was induced by natural turbidity and mining disposals.
- Extreme flooding affected sedimentary deposits impacted by mining.
- Fluvial erosion caused the remobilization of mercury-contaminated tidal sediments.
- Erosion of marshes containing high levels of Hg, coal microparticles and abundant microplastics should be monitored.

GRAPHICAL ABSTRACT



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ABSTRACT

Distinguishing between natural and anthropogenic processes in sedimentary records from estuaries with legacy pollutants is an essential task, as it provides baselines to predict future environmental trajectories of coastal areas. Here, we have addressed the recent transformation history of the mining-impacted Nalón Estuary (Asturias, N Spain). Surface and core sediment records from marshes and tidal flats were examined through a broad multidisciplinary approach, involving micropaleontological (benthic foraminifera), sedimentological (grain-size), geochemical (trace metals, major element Al and total organic carbon), physical (magnetic susceptibility, frequency-dependent magnetic susceptibility and large microplastics) and radioisotopic (^{210}Pb , ^{137}Cs and $^{239+240}\text{Pu}$) proxies. Results suggest that the interplay between natural (high fluvial influence and extreme hydrological events) and anthropogenic (coal and mercury mining disposals) factors induced strong sedimentation-erosion processes, further shaping the recent evolution of the estuary. Short-time scale and intense

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sedimentation processes were revealed by overall high sediment accumulation rates, the dilution of some geological signatures and the rapid formation of a marsh in the lower estuary bay. The increasing mining fingerprints during the 20th century were shortly interrupted by the catastrophic riverine flooding of 1938. Conversely, current erosional processes by fluvial influence led to the remobilization of contaminated sedimentary materials and exposure of mining-legacy Hg levels in tidal flats from the middle sector. Fluvial activity, floodings and taphonomic biases exerted a major control on benthic foraminifera since the 19th century, although Hg ecotoxicological effects on modern assemblages at certain areas within the estuary cannot be discarded. These findings, along with the documented enhanced erosion of marshes with ‘trapped’ pollutants (Hg, coal micro-particles and microplastics), highlight the importance of monitoring the environmental and geomorphic processes taking place in historically-contaminated estuaries.

1. Introduction

Estuaries hold a profound importance and their distinctive shape emerges from the combined relationship between natural agents and, more recently, human societies. Anthropogenic pressures are having detrimental effects on these coastal environments, involving uncontrolled discharges, fragmentation of habitats, ecosystems disruption, loss of biodiversity, natural landscape alteration, and biological invasions (e.g., Davis et al., 2000; Kennish, 2002; Lotze et al., 2006). Likewise, humans have also exerted a pronounced influence in sediment production, transport and dynamics, leading to coastal retreats, among other phenomena (Cendrero et al., 2022; Syvitski et al., 2005). These disturbances, amidst the ongoing global climate change, are severely compromising the environmental and ecological integrity of estuaries,

further deviating these ecosystems from their previous natural equilibrium (Borja and Dauer, 2008). Coastal sedimentary deposits, as natural archives, encapsulate a complex environmental history resulting from natural processes and anthropogenic legacies. In fact, the study of these deposits through surface samples and sediment cores has considerably increased in recent decades, providing crucial insights into estuarine dynamics and responses to pollution (Birch et al., 2013; Janssen et al., 2021), extreme disturbing events (Gallagher and Ross, 2018) and sea-level rise (Delgado et al., 2012; Lamb et al., 2007).

In the Asturian region of Spain anthropogenic activities, primarily derived from mining and industry, have compromised the quality state of several estuaries along its coastline (e.g., Flor-Blanco and Flor, 2019; Sierra et al., 2014). The region encompasses mountainous Paleozoic outcrops crossed by drainage systems with the largest coal mining

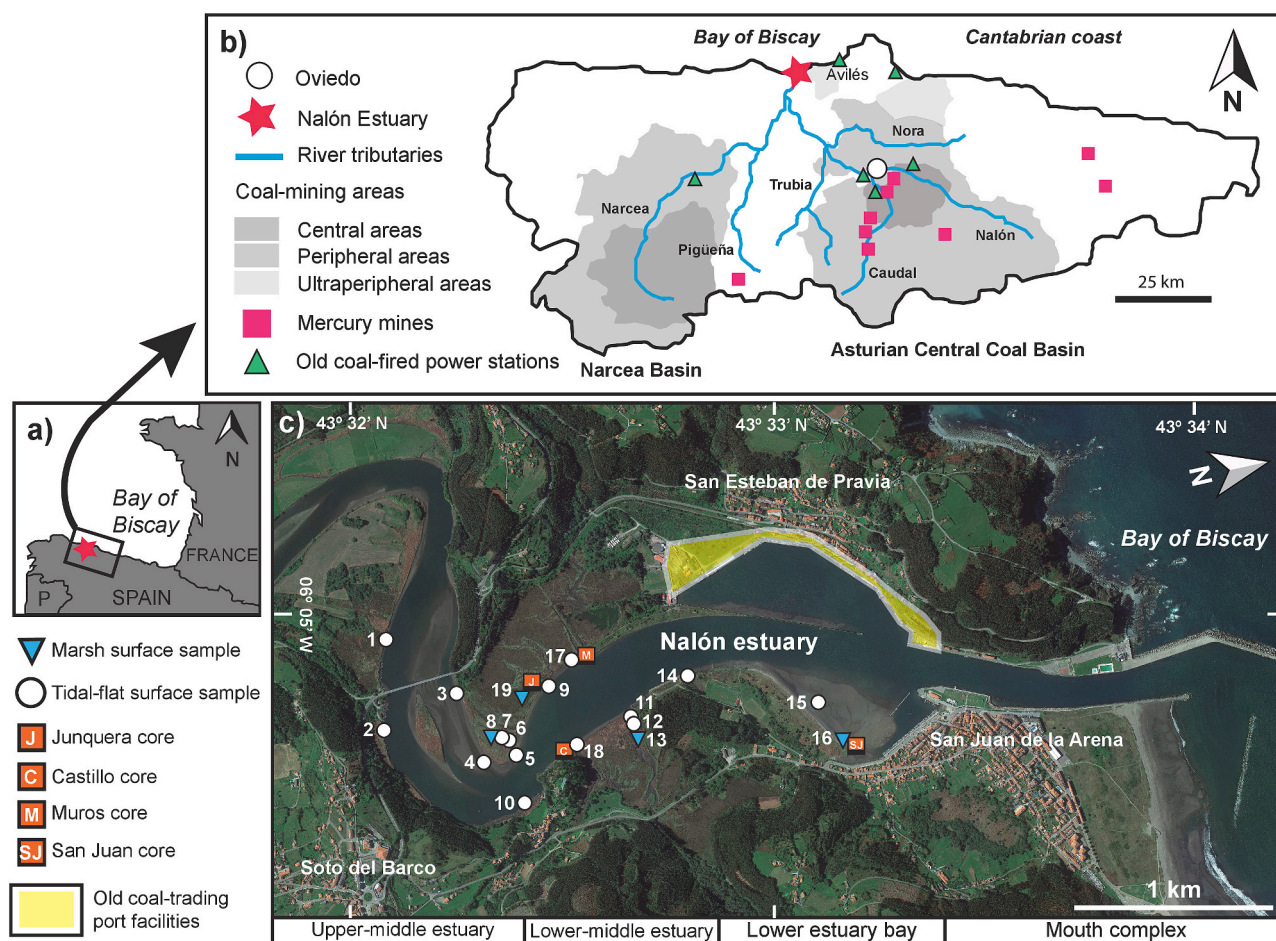


Fig. 1. Geographic location of the Nalón Estuary in N Spain (a). The Asturian region with the location of the Nalón Estuary (red star), its main tributaries, coal mining areas, mercury mines and coal-burning power stations (b). The localities mentioned in the text and position of marsh (inverted blue triangles) and tidal flat (white circles) surface samples and cores (orange squares) (c). For the interpretation of the references to color in the legend of this figure, the reader is referred to the web version of this article.

complexes of northern Spain: the Asturian Central Coal Basin and the Narcea Basin (Colmenero et al., 2008) (Fig. 1). Despite coal mining in the region dates back to before the 18th century, the intensive exploitation did not start until the second half of the 19th century (IGME, 2006), reaching its maximum rate in the 1960s (IGME, 2024). It is estimated that in the last two centuries, nearly 70 % of the Spanish coal production derived from the Asturian Central Coal Basin (Álvarez et al., 2016). Additionally, the Nalón valley hosts metallogenic deposits of mercury, with a paragenesis formed by native Hg, cinnabar and other sulphides, like As-enriched pyrite and pyrrhotite, among others (Lorredo et al., 2003). The exploited mines of this area (Fig. 1) were the second largest producer in Spain during the 20th century, and during the 1960s–1970s period they were considered the third global producer (Ordóñez et al., 2013). Despite closure of mercury exploitations in 1973–1974, coal mining in Asturias continued with a markedly decreasing production until 2018, when the Spanish coal mines were closed because of the measures enforced by the European Commission.

Under this scenario, the Nalón Estuary (Asturias, Spain), draining the waters of such deposits in the catchment area (Fig. 1), provides an excellent example of a coastal ecosystem impacted by coal and mercury mining. Previous studies in the area revealed inputs of trace metals due to mining exploitation (García-Ordiales et al., 2019, 2020). However, these historical anthropogenic fingerprints have the potential to mask the inherently diverse environmental variability arising from natural hydrological phenomena, such as floodings. There remains a significant knowledge gap regarding the distinction of these processes in sedimentary records, particularly in estuaries with anthropogenic legacies. In the context of global climate change, where the frequency of extreme hydrological processes is expected to increase (Hirabayashi et al., 2013), the task of assessing the evolution and drivers of human-impacted coastal areas through a comprehensive perspective is of paramount importance.

This work tackles the recent environmental transformation of the Nalón Estuary through a comprehensive multidisciplinary approach to its sedimentary record, involving micropaleontological (benthic foraminifera), sedimentological (grain size), geochemical (Pb, Zn, Cu, Ni, Hg, Al and TOC), physical (magnetic susceptibility, frequency-dependent magnetic susceptibility and large microplastics) and radiometric (²¹⁰Pb, ¹³⁷Cs and ²³⁹⁺²⁴⁰Pu) proxies. By examining intertidal surface and core samples, for the first time in the area, this study aims to gain insights and to reconstruct the primary anthropogenic activities and the merged natural dynamics conditioning the environmental and morphosedimentary evolution of this polluted estuary. This pivotal information provides a knowledge framework to assist local and regional authorities in predicting future environmental responses, developing science-based adaptive measures, and formulating coastal management strategies for historically-contaminated areas.

2. Study area

The Nalón is a relatively small (~6 km in length) and shallow (0–2 m deep) mesotidal estuary located at the Asturian region (43° 32' N, 6° 04' W), in the northern coast of Spain (Bay of Biscay). The estuary is classified as partially stratified, and its sedimentary dynamics is highly influenced by fluvial processes (Flor et al., 1998; Flor-Blanco and Flor, 2019). It is characterized by a sinuous and relatively narrow profile divided in four major zones (Fig. 1): upper channel, intertidal flats and marshes in the middle estuary, a small bay in the lower estuary, and the mouth complex (Flor et al., 2018). The upper sector presents a narrow meandering channel, dominated by fluvial inputs. The middle estuary consists of marshes, with palustrine *Phragmites australis* and *Juncus maritimus* as dominant vegetation (García-Ordiales et al., 2019), and intertidal flats. The lower estuary bay on its eastern sector is formed by a sandy intertidal flat and the San Juan de la Arena marsh, whereas on its western part the small touristic port of San Esteban de Pravia is located. The mouth complex is restricted by a group of factory dams, sand

barriers, a beach and a dune complex. The estuary is excavated into siliciclastic (sandstones, slates and quartzites) and carbonatic (limestones) Paleozoic materials. Its drainage network presents a large basin (~4890 km²) and receives the highest water discharge of the entire Cantabrian coast with an annual average flow of 109 m³ s⁻¹ (Prego et al., 2008). The regional wet climate, conditioning abundant rainfalls, shows an average annual precipitation of ~1000 mm (AEMET, 2012). Based on these features, the Nalón contributes with an annual load of 379,900 t of suspended solids, accounting >30 % of the total discharge of the Cantabrian rivers to the Bay of Biscay (Prego et al., 2008).

Human occupation on the margins of the Nalón valley date back to the Late Pleistocene (López-Tascón et al., 2020). Nevertheless, human impact in the estuarine surroundings did not become relevant until late 19th-early 20th centuries. In 1904, the estuary turned into the most important coal trading port of northern Spain due to the construction of the Basque-Asturian Railway, which transported coal from upstream mines to the San Esteban de Pravia's port (Fig. 1). On the left bank of the estuarine bay, three large cranes were installed in 1929–1930, accompanied by coal loading docks that were constructed to charge the mineral into the ships (Fig. 1). It is estimated that by the mid-20th century, the Nalón was the second most important coal trading port in Spain (Vera González, 1958). Despite the middle and upper estuarine banks have remained physically unaltered in general terms, the lower estuary has suffered significant modifications since late 19th century. In fact, it is inferred that >12 % of the estuarine surface was lost due to anthropogenic reclamation (Cabrera Ceñal and Flor, 1993). In 1910, the San Esteban de Pravia port's dock was built to protect coastal activities from the strong hydrodynamic regime and sediment infilling. During the period of 1928–1944, this mouth was channelled for navigation purposes and a large factory dam was built in the western sector, accompanied by another dam in the eastern part (Flor et al., 1998, 2018; Vera González, 1958). Finally, between 2004 and 2006 a small dock for leisure vessels was constructed in San Juan de la Arena.

3. Materials and methods

Nineteen surface samples were collected along the Nalón Estuary from intertidal flat (15) and marsh (4) environments in October 2020 and October 2021 during low tide (Fig. 1). To study modern foraminiferal assemblages and minimize patchiness at microscale distribution, two pseudo-replicates of 40 cm² (80 cm³ = 80 cm² × 1 cm) were taken (Murray and Alve, 2000) by pressing a hard-plastic ring into the sediment. The sediment was placed in a plastic jar containing ethanol to prevent the decay of foraminiferal protoplasm (Murray, 2006). At each sampling station, surface sediments were scrapped with a plastic spatula for grain-size, geochemical (trace metals, major element Al and TOC) and environmental magnetism. Two intertidal flat (Castillo and Muros) and two marsh (Junquera and San Juan) short sediment cores were drilled in October 2020 and October 2022 from the lower-middle estuary and the bay area (Fig. 1). Sediment cores were retrieved by inserting three 50-cm-long PVC tubes (11.5 cm internal diameter) into the sediment at each location. Micropaleontological (benthic foraminifera), grain size and geochemical (trace metals, Al and TOC) studies were performed on one replicate, whereas the second and the third replicates were used for radiometric and environmental magnetism analyses. Additionally, to study the content of large microplastics (>1 mm) two more replicates were extracted at Muros intertidal flat and Junquera marsh coring points, respectively. At the laboratory, all core replicates were split lengthwise, described, photographed and divided into 1-cm increments. The analyses of benthic foraminifera, grain size, trace metals and major element Al, and microplastic content were performed on alternate samples from 0 to 1 cm to 49–50 cm depth, except where changes were detected that the intermediate sample was studied to provide a better resolution. Radiometric and environmental magnetism studies were performed every 1 cm.

For benthic foraminifera and grain-size characterization, surface and

core samples were wet sieved through 2 mm and 63 μm meshes. After washing, to distinguish the living individuals from the dead tests, a solution of Rose Bengal (1 g L^{-1}) was added to the surface samples and left for an hour (Walton, 1952). Subsequently, the samples were washed again to remove the excess of stain. The retained sand-size material was dried at $50\text{ }^{\circ}\text{C}$ and concentrated by flotation in trichloroethylene (Murray, 2006). The residue was poured through a splitter and analyzed under a stereoscopic binocular microscope Nikon SMZ645 using reflected light. For each sample, a minimum number of 300 tests was extracted for correct statistical representation. Otherwise, when fewer foraminifera were present, all the available tests were picked and studied. Foraminiferal abundances were stated as relative percentages considering the total number of tests picked in each sample. The standing crops or densities of the stained-living assemblages were expressed as the total number of individuals per 80 cm^3 . Foraminiferal densities from core samples were explicated as the number of tests per 15 g of bulk dry sediment and grouped according to García-Artola et al. (2016). The sand content was determined by weighing the dry residues retained in the 63- μm sieve, and expressed as relative percentages of bulk dried weight sediment. In total, 153 samples and >34,200 benthic foraminifera were studied at the University of the Basque Country UPV/EHU (Spain). Classification followed Loeblich and Tappan (1988) and later taxonomic updates based on the World Register of Marine Species (WoRMS). Taxa identified in this study have been listed in Table A.1. For the taphonomic characterization of foraminiferal tests, scanning electron microscope images were obtained at the Analytical and High-resolution Microscopy in Biomedicine Service (SGIker, UPV/EHU/ERDT, EU). Cearreta (1988, 1989), Murray (2006) and Scott et al. (2007) and other similar works were used as a base for the ecological and environmental requirements of the foraminiferal species. To compare the homogeneity between living and dead assemblages, the Bray-Curtis similarity index (Bray and Curtis, 1957) was calculated using PAST version 4.15. This index varies from 0 to 1, considering as a good similarity those values above 0.7. In the calculation, only samples with >100 tests in both assemblages were considered. Based on the abundances and modern distribution of the species found in surface and core samples, they were divided into autochthonous, those living and reproducing in the estuary, and allochthonous, transported into the estuary from outer marine and shelf areas.

For the analysis of Al and trace metal concentrations (Pb, Zn, Cu, Ni, Hg) an amount of 0.5 g of sediment (<2 mm fraction) was oven-dried and mechanically homogenised using an agate mortar and pestle. Elemental concentrations were analyzed in Activation Laboratories Ltd. (Actlabs, Canada) by inductively coupled plasma–optical emission spectrometry after digestion of each sample in aqua regia (a mixture of concentrated nitric and hydrochloric acids) for 2 h at $95\text{ }^{\circ}\text{C}$. The detection limits were 0.01 % for Al, 2 mg kg^{-1} for Pb and Zn, and 1 mg kg^{-1} for Cu, Ni, and Hg. Concentrations of Hg in some selected samples were measured by inductively coupled plasma–mass spectrometry at the same laboratory (detection limit 0.1 mg kg^{-1}). Trace metal concentrations were normalised to Al values to account for mineralogical and granulometric variability (i.e., Pb/Al). The analysis of total organic carbon (TOC) in the Junquera marsh core was performed on 100 mg of dry sediment samples using a LECO CNS 928 analyser at the Scientific and Technique Soil Analysis Service of the Pyrenean Ecology Institute (IPE-CSIC, Spain), see Gardoki et al. (2023) for further details.

Dating recent sedimentary deposits by ^{210}Pb vertical activity distribution was performed in order to estimate sedimentation rates and to establish absolute chronologies. The method is widely used in recent sedimentary records, and relies on the $^{210}\text{Pb}_{\text{xs}}$ accumulated by atmospheric deposition, which is the excess activity resulting from the difference between the total measured concentration of ^{210}Pb and the ^{210}Pb in equilibrium with the ^{226}Ra present in the sediment. Isotopic activities of natural (^{210}Pb and ^{226}Ra) and artificial (^{137}Cs) short-lived radionuclides were analyzed by gamma spectrometry in core samples using a HPGe detector at the University of Cantabria (Spain), see Cearreta et al.

(2013) and Irabien et al. (2008) for complete procedural details. Likewise, after sequential chromatographic separations, measurements of artificial $^{239+240}\text{Pu}$ radiotracers were carried out in the Laboratory of Environmental Radioactivity at the Polytechnic University of Valencia (Spain) by alpha spectrometry, as described in Sáez-Muñoz et al. (2020).

Low-frequency (LF, 0.465 kHz) mass-normalised bulk magnetic susceptibility (χ , $\text{m}^3\text{ kg}^{-1}$) was measured ten times for each sample with a Bartington MS3/MS2B dual frequency equipment, after being dried at $40\text{ }^{\circ}\text{C}$, at the Material and Environmental Magnetism Laboratory (National Geographic Institute, Spain). Average values and standard deviations were calculated. High-frequency (HF, 4.65 kHz) magnetic susceptibility was also measured ten times per sample, computing afterwards the frequency-dependent susceptibility ($\chi_{\text{fd}} = 100 \cdot (\chi_{\text{LF}} - \chi_{\text{HF}}) / \chi_{\text{LF}}$) as a relative estimator of the proportion of superparamagnetic (very fine) grains in the samples. These fine superparamagnetic grains often have their origin in soil-forming processes on land (Evans and Heller, 2003).

The extraction of microplastics (only particles with a total length of >1000 μm total length were considered in this study) from sediments was conducted in a controlled laboratory environment to prevent airborne fiber contamination, following the method described in Gardoki et al. (2023) and adapted from Bessa et al. (2019). In brief, sediment samples were dried at $40\text{ }^{\circ}\text{C}$ for 48 h, then treated with 20 % hydrogen peroxide to reduce organic matter (24 h at room temperature). Afterward, each sample was sieved through a 1000 μm steel sieve using filtered distilled water. The solution was then placed in a glass beaker with supersaturated saline solution (NaCl , 1.2 g cm^{-3}) to allow potential microplastics to float and settle for 2 h. The supernatant was vacuum filtered through 1.2 μm Whatman GF/C microfiber filters, sealed in Petri dishes for analysis under a stereomicroscope. Suspected particles were extracted using tweezers and characterized by shape and color, measured at their largest cross-section (mm) using a dissecting microscope with image analysis software. Additionally, to avoid the inclusion of natural particles, a hot-needle test was conducted by heating a thin needle and poking suspected microplastics under the stereomicroscope (De Witte et al., 2014). In total, 365 large microplastics were identified from microscopic observation of sediment samples at the University of Coimbra, Portugal.

To obtain historical graphic documents and aerial photographs, several sources have been consulted, involving the National Geographic Institute (Spain) and the Territorial Information System and Spatial Data Infrastructure of Asturias (Spain) virtual servers.

4. Results and discussion

4.1. Surface samples

Marshes located in the middle sector on both banks of the estuary contained a low sand abundance (average 12 %) and their foraminiferal contents are almost exclusively constituted by agglutinated forms both in the living (92 %) and dead (100 %) assemblages (Table 1). These environments present moderate standing crop values (586 individuals/ 80 cm^3). In the living and dead assemblages, *Trochammina inflata*, *Miliammina fusca* and *Entzia macrescens* stand out as principal species, whilst *Haplophragmoides wilberti* appears as a secondary taxon (Table 1). The similarity index (0.79) indicates a high correspondence between the biocoenosis and the thanatocoenosis, and further suggests barely no taphonomic and environmental controls biased their modern assemblages. Based on conducted field surveys, these middle estuary marshes present highly developed eroding fronts, characterized by notable and pronounced edge cliffs (Fig. S1a, b). Likewise, the San Juan de la Arena marsh in the lower estuary bay (station 16; Fig. 1) presents a marked eroding, stepped profile (Fig. S1c), and its living and dead assemblages are also almost entirely constituted by agglutinated forms. Although the living assemblage presented <100 tests, the thanatocoenosis is also characterized by *E. macrescens*, *T. inflata* and *H. wilberti* as dominant

Table 1

Summary of sedimentological and micropaleontological (foraminifera) data from surface samples collected along the Nalón Estuary (Asturias, N Spain). The single value represents the average and those in parentheses the range.

	Living assemblages	Dead assemblages
Middle estuary		
Marshes (n = 3)		
Mud 88 (79–93) %		
Sand 12 (7–21) %		
Standing crop 80 cm ³	586 (172–1358)	
Similarity Index 0.79 (0.6–0.94)		
Marine tests %	0	0
Hyaline %	8 (1–12)	1 (0–1)
Porcellaneous %	0	0
Agglutinated %	92 (90–99)	99 (99–100)
<i>T. inflata</i> %	61 (54–70)	51 (36–66)
<i>E. macrescens</i> %	18 (0–22)	20 (7–30)
<i>M. fusca</i> %	9 (0–14)	22 (0–49)
<i>H. wilberti</i> %	8 (3–15)	5 (3–6)
<i>A. morphogroup tepida</i> %	6 (1–9)	0
Tidal flats (n = 14)		
Stations 1, 2, 3, 4, 5, 6, 7, 9, 10, 11, 12, 14, 17, 18		
Mud 60 (1–86) %		
Sand 40 (14–99) %		
Standing crop 80 cm ^{3a}	327 (63–663)	
Similarity Index 0.62 (0.37–0.82)		
Marine tests %	0 (0–1)	2 (0–7)
Hyaline %	83 (47–95)	59 (5–91)
Porcellaneous %	1 (0–6)	1 (0–4)
Agglutinated %	16 (3–53)	40 (9–95)
<i>T. inflata</i> %	4 (0–22)	21 (0–53)
<i>E. macrescens</i> %	1 (0–3)	12 (0–45)
<i>M. fusca</i> %	10 (2–27)	5 (1–15)
<i>H. wilberti</i> %	0.1 (0–1)	2 (0–9)
<i>A. morphogroup tepida</i> %	15 (2–32)	6 (0–19)
<i>H. germanica</i> %	60 (38–79)	49 (4–74)
<i>E. excavatum</i> %	5 (0–10)	3 (0–7)
<i>E. oceanense</i> %	3 (0–19)	0.2 (0–2)
<i>E. williamsoni</i> %	1 (0–6)	1 (0–2)
<i>Q. seminulum</i> %	1 (0–6)	0.4 (0–2)
Lower estuary bay		
Marsh (n = 1)		
Station 16	<100 tests	
Mud 36 %		
Sand 64 %		
Standing crop 80 cm ³	34	
Similarity Index -		
Marine tests %	–	0
Hyaline %	–	3
Porcellaneous %	–	0
Agglutinated %	–	97
<i>E. macrescens</i> %	8 tests	56
<i>T. inflata</i> %	20 tests	23
<i>H. wilberti</i> %	2 tests	15
Tidal flat (n = 1)		
Station 15	<100 tests	
Mud 14 %		
Sand 86 %		
Standing crop 80 cm ³	23	
Similarity Index -		
Marine tests %	–	23
Hyaline %	–	92
Porcellaneous %	–	7
Agglutinated %	–	1
<i>H. germanica</i> %	13 tests	41
<i>A. morphogroup tepida</i> %	8 tests	19
<i>L. lobatula</i> %		8
<i>Q. seminulum</i> %		6
<i>E. excavatum</i> %		6

^a Except sample 5: Sand (99 %), Mud (1 %). Standing crop: 2 individuals/80 cm³. Living assemblage: *H. germanica* (2 tests). Dead assemblage: *T. inflata* (5 tests), *A. morphogroup tepida* (4 tests) and *H. germanica* (1 test).

species. The documented main agglutinated species (*T. inflata*, *E. macrescens*, *H. wilberti* and *M. fusca*) typically inhabit vegetated marshes along the Cantabrian coast (Pascual et al., 2019; Rodríguez-Lázaro et al., 2013), and other temperate estuaries in the northern Atlantic realm (Fatela et al., 2009; Kemp et al., 2012).

On the other hand, tidal flats in the upper- and lower-middle estuary present a higher sand content (40 %), and their living foraminiferal assemblages are almost entirely constituted by hyaline forms (91 %). Their standing crops exhibit lower values (327 individuals/80 cm³), with the minimum observed at station 5 (2 individuals/80 cm³) likely due to its high sand content. The living assemblages are characterized by *Haynesina germanica*, *Ammonia* morphogroup *tepida*, and *M. fusca*, with secondary abundances of *Elphidium excavatum* (Table 1). The tidal flat in the lower estuary bay (station 15) presents a very low standing crop and high sand and allochthonous foraminiferal dead contents (Table 1). The biocoenosis only comprises some tests of *H. germanica* and *A. morphogroup tepida*, and the main species forming the dead assemblage are *H. germanica* and *A. morphogroup tepida*, with *Lobatula lobatula*, *Quinqueloculina seminulum* and *E. excavatum* as secondary taxa (Table 1). The dominance of these living euryhaline species in the intertidal flat domains of the Nalón Estuary indicates prevailing brackish conditions, with slightly higher marine influence toward the mouth of the estuary (Camacho et al., 2015; Debenay et al., 2006). Conversely, the increasing abundances of living agglutinated salty-tolerant taxa (*T. inflata* and *E. macrescens*) and the decrease of hyaline estuarine brackish individuals (*H. germanica*, *A. morphogroup tepida* and *E. excavatum*) (Fig. S2a) toward the marshes indicate that the topographic gradient, mostly controlled by tidal inundation, is a primary factor influencing the living microfauna. A similar topographic biozonation of foraminiferal assemblages was reported by Rodríguez-Lázaro et al. (2013) and Pascual et al. (2019) in nearby Cantabrian coast estuaries, also documented in other Atlantic transitional ecosystems (Horton and Murray, 2007; Leorri et al., 2008).

In contrast, dead assemblages from tidal flats comprise high percentages of both hyaline (59 %) and agglutinated (40 %) tests. *Haynesina germanica* stands out as the main taxon, accompanied by dead agglutinated taxa inhabiting these marshes, such as *T. inflata* and *E. macrescens* (Table 1). While living agglutinated foraminifera do not exhibit very high abundances, the tests of marsh agglutinated species constitute an important part of the thanatocoenosis, accounting for 77 % and 95 % of the assemblage in tidal flat stations 6 and 7, respectively. In this regard, according to the Bray-Curtis similarity index, tidal flat assemblages present moderate similarities (0.62), with particularly remarkable low resemblances in stations 6 (0.37), 1 (0.44), 4 (0.49) and 7 (0.51) (Fig. 1). The enrichment of agglutinated taxa in the surface sediments of the Nalón Estuary (Fig. S2b) due to early taphonomic factors could be discarded, as no signals of chemical dissolution were observed in calcareous tests. Dissimilarities between the biocoenosis and thanatocoenosis, in many cases associated with the dissolution of calcareous taxa during burial, are a recurrent phenomenon in transitional environments worldwide (e.g., Moreno et al., 2007; Murray and Alve, 1999), including the Cantabrian coast estuaries (Cearreta, 1988; Cearreta et al., 2002; Rodríguez-Lázaro et al., 2013). However, the remarkable abundances of agglutinated dead tests in tidal flat sediments from the middle sector of the Nalón Estuary are certainly triggered by the ongoing erosion of adjacent marshes (Fig. S1a, b), since the aforementioned taxa (e.g., *T. inflata* and *E. macrescens*) live solely in these vegetated marshes. The eroding marsh banks in the upper- and lower-middle estuary are causing collapsed metric blocks of sediments that supply abundant agglutinated microfauna to contiguous intertidal flats (Fig. S1a, b). A similar process was observed by Figueira and Hayward (2014) at Waikawa Harbour (New Zealand), where erosive processes led to substantial mixing of reworked agglutinated foraminiferal tests with modern in situ microfauna in sandy intertidal flats.

The documented foraminiferal standing crops in the Nalón Estuary (Table 1) exhibit low values compared to those identified in nearby tide-

Table 2

Range of concentrations in surface and core sediments from the Nalón Estuary (Asturias, N Spain), values for northern Iberian estuaries proposed by different authors, Sediment Quality Guidelines and Spanish classification of dredged materials. Trace metal values are expressed in mg kg^{-1} , TOC (%), magnetic susceptibility (χ) values in $10^{-7} \text{ m}^3 \text{ kg}^{-1}$ (standard deviation $<0.05 \cdot 10^{-7} \text{ m}^3 \text{ kg}^{-1}$) and frequency-dependent susceptibility (χ_{fd}) values in % (standard deviation <1 %). Large microplastic content is expressed as the number of particles per kg of dry weight sediment.

	Cu	Pb	Zn	Ni	Hg	TOC	χ	χ_{fd}	Large microplastics
Nalón estuary (this work)									
Marshes (n=4)	31–37	34–38	107–149	24–30	0.63–1.39	–	9.1–15.0	0.5–2.2	–
Tidal flats (n=15)	12–41	12–44	31–113	12–28	0.12–5.31	–	2.8–17.1 ^a	0.6–2.2	–
Jun-1 (n=5)	17–20	51–82	96–140	23–35	0.90–1.4	11–17	1.7–4.2	2.0–3.5	0
Jun-2 (n=6)	15–21	19–70	86–123	25–31	0.33–1.79	4–17	2.7–5.7	1.6–4.8	0
Jun-3 (n=18)	37–48	45–126	113–183	25–36	0.54–3.46	8–21	5.1–36.7	1.2–2.5	0–170
Cas-1 (n=20)	8–22	8–26	41–127	13–26	<1–13	–	1.5–9.8	4.7–13.9	–
Cas-2 (n=9)	13–22	14–34	58–122	16–29	2–10	–	3.6–9.4	2.2–12.9	–
Mur-1 (n=27)	33–55	38–50	159–516	26–32	<1	–	23.2–54.5	1.3–2.4	28–202
Mur-2 (n=8)	38–50	31–44	116–265	23–31	<1	–	14.1–40.6	1.2–1.6	9–193
SJuan-1 (n=10)	30–41	48–60	95–111	23–29	<1	–	39.4–64.6	0.7–1.6	–
SJuan-2 (n=17)	33–45	33–81	87–137	24–31	<1	–	25.7–64.9	1.4–2.5	–
Cantabrian coast estuaries									
Avilés (Sierra et al., 2014)	1–1,272	3–8,710	37–10,000	1–167	–	–	–	–	–
Bilbao (Cearreta et al., 2000)	41–1,946	75–2,566	167–7,687	20–129	–	–	–	–	–
Suances (Serrano, 2020)	26–104	544–2,700	2,070–10,000	11–28	<1–41	–	–	–	–
Pasaia (Irabien et al., 2020)	7–4,490	12–>5,000	65–>10,000	12–175	–	–	–	–	–
Reference values									
Background Asturian coastline (Sanz-Prada et al., 2022)	11	14	47	26	0.6	–	–	–	–
Background Basque estuaries (Rodríguez et al., 2006)	33	31	174	29	0.13	–	–	–	–
ERM (Long et al., 1995)	270	218	410	51.6	0.71	–	–	–	–
Action Level C	675	600	1640	234	2.84	–	–	–	–
Non-hazardous sediments (Buceta et al., 2015)	2,500	1,000	2,500	1,000	17	–	–	–	–

^a Except sample 16, that shows values of $48.7 \cdot 10^{-7} \text{ m}^3 \text{ kg}^{-1}$.

dominated estuaries (Cearreta, 1988, 1989; Leorri et al., 2008), as well as other transitional ecosystems within the Atlantic realm (e.g., Horton and Murray, 2007; Murray and Alve, 2000). Likewise, the scarcity of marine allochthonous and porcelaneous taxa in the middle sector (Table 1; Fig. S2a, b) underscores the prevalent fluvial dominance in the Nalón, recognized as the estuary with the highest river discharge and sediment load of the entire Cantabrian coast (Prego et al., 2008). Hence, fluvial influence and discharges conditioning the sedimentation of the estuary (Flor et al., 1998) could represent primary environmental factors affecting benthic foraminiferal distribution and, potentially, the erosion of marshes.

Surface concentrations of Pb, Zn, Cu and Ni are generally similar in both estuarine subenvironments (Table 2), with the lowest values corresponding to stations from the lower estuary bay due to the sandy nature of its sediments. The range values of these trace metals are comparable to the geochemical background established by Sanz-Prada et al. (2022) and Rodríguez et al. (2006) for the Asturian coastline and the Basque estuaries (Table 2), respectively, located along the same Cantabrian coast. However, Hg concentrations, in most cases, are comparable to or even higher than these geochemical backgrounds. The persistent presence of high concentrations of Hg in some surface samples (Fig. S2) attests to the mercury mining legacy and its potential environmental risk, even after the closure of upstream mines five decades ago (Ordóñez et al., 2013). Surface sediments also present a high coal microparticles content, associated to the influence of past coal mining activities (Cabrera Ceñal and Flor, 1993). Magnetic susceptibility in the tidal flats and marshes shows relatively similar and low to moderate values (Table 2), except for the lower estuary marsh (station 16) which presents the highest value of all the surface samples. These findings point out to a persistent source of anthropogenic ferromagnetic contamination in the area (Evans and Heller, 2003).

4.2. Junquera core

The $^{210}\text{Pb}_{\text{xs}}$ activity shows decreasing values downward until its final cancellation at the core base (Fig. 2). The chronology of this sedimentary record has been established by using the Constant Rate of Supply model (Appleby and Oldfield, 1978; Sanchez-Cabeza and Ruiz-Fernández, 2012). According to these results, it can be framed between the 1858 ± 54 and 2022 CE. The temporal margins of uncertainty are associated with the low activity of $^{210}\text{Pb}_{\text{xs}}$ at the lowermost section. However, the artificial radionuclides ^{137}Cs and $^{239+240}\text{Pu}$ exhibit a distinctive peak at 30–31 cm depth (Fig. 2) coinciding with 1954 ± 16 CE, broadly validating the absolute ages provided by the ^{210}Pb . The first appearance of large microplastics occurring at 31 cm depth, with upcore increasing abundances, corroborates the radiotracers chronology (Table 2). Their content is made of fibers (55 %), followed by filaments (14 %), fragments (13 %), films (13 %) and tangles of fibers (5 %) (Fig. S3).

According to the information provided by the different proxies, three different units have been distinguished in the Junquera core (Fig. 2; Table 3). The basal unit Jun-1 (50–40 cm) comprises black muddy sediments with a low sand content (5 %) and foraminiferal density (135 tests/15 g) (Table 3). The interval is primarily constituted by agglutinated foraminifera (100 %) typical of marsh environments, with the dominant species *E. macrescens* and *T. inflata* (Table 3). Sediment accumulation rate (SAR) exhibits low values, varying from 0.04 to 0.16 cm yr^{-1} . Al-normalised trace metals and TOC profiles display a slightly increasing trend upward, whereas magnetic susceptibility and very fine superparamagnetic particles contribution (χ_{fd}) are low. Hg concentrations above the geochemical background (Table 2), together with a high abundance of coal microparticles (Fig. S4a), support the deposition of this interval after the mid-19th century once the beginning of coal and mercury mining activities upstream had taken place (García-Ordiales et al., 2019, 2020).

In contrast, unit Jun-2 (40–34 cm) comprises light-brownish

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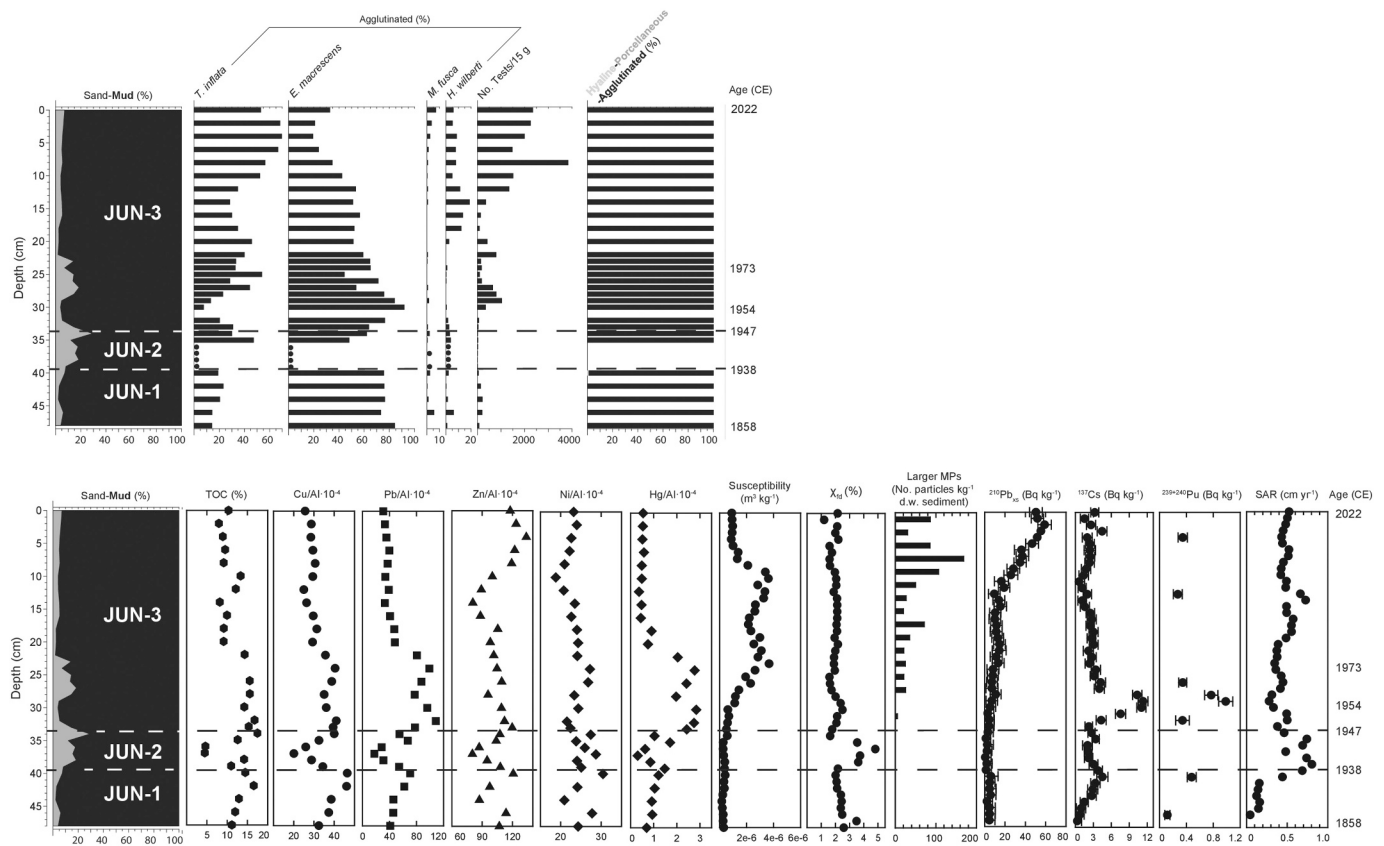


Fig. 2. Vertical distribution of grain-size contents (%), relative abundance of main foraminiferal species (%), TOC (%), Al-normalised trace metals, magnetic susceptibility ($\text{m}^3 \text{kg}^{-1}$) and frequency-dependent susceptibility (%), microplastics content (particles kg^{-1} d.w. sediment), $^{210}\text{Pb}_{\text{xs}}$, ^{137}Cs and $^{239+240}\text{Pu}$ activities (Bq kg^{-1}), and Sediment Accumulation Rate (cm yr^{-1}) in the Junquera core, Nalón Estuary. Black dots represent samples with <100 tests.

sediments and is characterized by the occurrence of substantial changes (Table 3). It exhibits an increase in the sand content (17 %), remarkably low foraminiferal densities (15 tests/15 g) and, interestingly, a very low content of coal microparticles (Fig. S4b). The same agglutinated species as in Jun-1 are present, but mostly in very low abundances. In this second interval, the SAR considerably increases to a maximum of 0.8 cm yr^{-1} (Fig. 2). Regarding trace metals and TOC, these parameters display a marked decreasing profile, with most of them presenting minimum concentrations. Magnetic susceptibility is also relatively low, but the content in very fine superparamagnetic particles, expressed by χ_{fd} , considerably increases (Fig. 2). Considering all these characteristics, this interval, whose basal limit is chronologically located at $1938 \pm 21 \text{ CE}$, is in very good agreement with the catastrophic flooding of September 1938 occurred in Asturias (Lastra et al., 2008; PLANINPA, 2010). The dilution of the micropaleontological, geochemical and magnetic signatures, and the notable increase in SAR and in very fine superparamagnetic particles of probable pedogenic origin, are highly likely derived from the input of “allochthonous” sediments from fluvial catchment areas. These features were also documented by Irabien et al. (2019) in sediment cores from the outer sector of the Bilbao Estuary (N Spain), attributed to the extreme floodings of 1983. Moreover, as observed from the sedimentological features (Fig. S4), the flooding could have also diluted the content of coal microparticles transported from upstream mines by the Nalón River. Historically, its catchment basin has experienced numerous floodings (Fernández et al., 2017; Lastra et al., 2008), and the 1938 flooding is considered one of the most significant inundations occurred in northern Spain during the 20th

century. In fact, the Spanish Meteorological Observatory estimated that day rainfall on September 8th was equivalent to the total annual rainfall that year.

The most recent interval Jun-3 (34–0 cm) is made of black muddy sediments, with a low sand content (8 %) and abundant presence of coal microparticles, similar to the basal unit Jun-1 (Fig. S4c). Foraminiferal densities experience a significant increase upwards (900 tests/15 g). The assemblages comprise variable abundances of the principal species *E. macrescens* and *T. inflata*, and secondarily of *H. wilberti* and *M. fusca* (Table 3). SAR exhibits lower values than those determined in the previous unit Jun-2, with an average of $\sim 0.46 \text{ cm yr}^{-1}$, and a rather constant profile (Fig. 2). After the 1938 inundation, previous environmental dynamics seem to be recovered, with predominantly muddy sedimentation and higher foraminiferal densities. The upcore increasing abundances of *T. inflata* could indicate a transition toward a marsh with lower topographic elevation (Leorri et al., 2010). Nevertheless, tests of this taxon exhibit abundant partial breakage signals in chamber walls and septa, and exposure of inner linings downcore (Fig. S5a, b, c.1 and c.2). Furthermore, the microtexture of *T. inflata* tests revealed the existence of numerous uncemented and disaggregated grains (Fig. S5d.1 and d.2, e.1 and e.2). These characteristics suggest strong taphonomic processes conditioning the preservation of the assemblages and a selective loss downcore.

Vertical patterns displayed by TOC and Al-normalised Hg, Cu and Pb are fairly similar with maximum values between 34 and 24 cm depth, notably higher than those observed in the basal interval Jun-1 (Fig. 2). The maximum concentrations of Hg in the Junquera marsh, occurring

Table 3

Summary of sedimentological and micropaleontological (foraminifera) data from cores drilled in the Nalón Estuary (Asturias, N Spain). The single value represents the average and those in parentheses the range.

Junquera	Castillo	Muros	San Juan
Jun-3 (34–0 cm)	Cast-2 (11–0 cm)	Mur-2 (13–0 cm)	SJuan-2 (28–0 cm)
Mud 92 (82–98) %	Mud 63 (51–83) %	Mud 80 (57–90) %	Mud 50 (33–84) %
Sand 8 (2–19) %	Sand 37 (17–49) %	Sand 20 (10–43) %	Sand 50 (16–67) %
High coal microparticles content	Moderate coal microparticles content	Moderate coal microparticles content	High coal microparticles content
No. Tests/15 g 900 (41–3840)	No. Tests/15 g 33 (16–40)	No. Tests/15 g 93 (19–175)	No. Tests/15 g 56 (6–211)
Marine 0 %	Marine 0 (0–0.4) %	Marine 0 (0–1) %	Marine 0 %
Hyaline 0 %	Hyaline 91 (78–99) %	Hyaline 77 (57–85) %	Hyaline 89 (78–98) %
Agglutinated 100 %	Agglutinated 9 (1–22) %	Agglutinated 23 (15–44) %	Agglutinated 11 (2–22) %
E. macrescens 54 (19–92) %	H. germanica 61 (56–69) %	H. germanica 53 (37–61) %	E. macrescens 43 (1–94) %
T. inflata 39 (8–70) %	A. morphogroup tepida 18 (8–25) %	A. morphogroup tepida 20 (15–28) %	T. inflata 38 (0–77) %
H. wilberti 5 (0–19) %	E. williamsoni 7 (2–12) %	T. inflata 9 (7–11) %	H. wilberti 13 (0–41) %
M. fusca 1 (0–7) %	E. excavatum 5 (1–12) %	M. fusca 8 (2–32) %	M. fusca 11 (2–33) %
	M. fusca 3 (0.4–7) %	E. macrescens 5 (2–11) %	
	E. macrescens 3 (0–13) %	E. excavatum 2 (1–5) %	
	T. inflata 3 (0.4–5) %		
Jun-2 (40–34 cm) (40–36 cm < 100 tests/sample)	Cast-1 (50–11 cm)	Mur-1 (50–13 cm) (45–30 cm, 26–23 cm and 19–13 cm < 100 tests/sample)	SJuan-1 (50–28 cm) (<100 tests/sample)
Mud 83 (71–92) %	Mud 49 (23–75) %	Mud 88 (59–97) %	Mud 30 () %
Sand 17 (8–29) %	Sand 51 (25–77) %	Sand 12 (3–41) %	Sand 70 (60–80) %
Very low coal microparticles content	Very low coal microparticles content	High coal microparticles content	High coal microparticles content
No. Tests/15 g 15 (6–29)	No. Tests/15 g 36 (10–117)	No. Tests/15 g 18 (0–72)	No. Tests/15 g 3 (0–11)
Marine - %	Marine 1 (0–5) %	Marine 4 (0–9) %	Marine - %
Hyaline - %	Hyaline 99 (98–100) %	Hyaline 83 (64–93) %	Hyaline - %
Porcellaneous - %	Agglutinated 1 (0–2) %	Porcellaneous 2 (0–7) %	Agglutinated - %
Agglutinated - %	H. germanica 52 (21–74) %	Agglutinated 15 (4–35) %	H. germanica 0–43 tests
E. macrescens 18–52 tests	E. excavatum 21 (8–54) %	H. germanica 65 (44–77) %	A. morphogroup tepida 0–19 tests
T. inflata 16–35 tests	A. morphogroup tepida 14 (1–32) %	A. morphogroup tepida 8 (4–13) %	E. excavatum 0–8 tests
H. wilberti 2–5 tests	E. williamsoni 10 (4–20) %	E. excavatum 6 (0–11) %	M. fusca 0–44 tests
M. fusca 0–2 tests		T. inflata 5 (1–10) %	
		M. fusca 5 (0–24) %	
		E. macrescens 4 (1–6) %	
Jun-1 (50–40 cm)			
Mud 95 (93–98) %			
Sand 5 (2–7) %			
High coal microparticles content			

Table 3 (continued)

Junquera	Castillo	Muros	San Juan
No. Tests/15 g 135 (46–208)			
Marine 0 %			
Hyaline 0 (0–1) %			
%			
Agglutinated 100 (99–100) %			
E. macrescens 77 (73–84) %			
T. inflata 18 (14–23) %			
H. wilberti 2 (0–6) %			
%			
M. fusca 2 (0–6) %			

between 1947 ± 19 and 1973 ± 10 CE, coincide with the highest historical production patterns of Hg flasks in Asturias (Luque Cabal and Gutiérrez Claverol, 2006). Subsequently, the clear decreasing trend in Hg since 1973 ± 10 CE is compatible with the closure of the mercury mines located upstream (Ordóñez et al., 2013). Magnetic susceptibility behaves differently, exhibiting a progressive and very significant increase from 34 cm upwards, and reaching very high values between 26 and 8 cm depth (1966–2004 CE) (Fig. 2). Thus, maximum ferromagnetic input to the sediments seems to be reached after the 1960s and does not abate until the 2000s, when susceptibility clearly decreases. This behaviour is compatible with an anthropogenic ferromagnetic source related to abundant magnetite-rich microspherules in the fly ash coming from coal-fired power plants, located nearby the estuary and in the catchment area (Fig. 1). The majority of these plants reached their peak activity between the late 1960s and the beginning of the 21st century. The decreasing trend in magnetic susceptibility observed since the 2000s coincides with the enactment of stricter UE and national environmental legislation and when the power plant closest to the Nalón estuary (ENSIDESA, Avilés) ceased its operations (Fernández Montes, 2016; PRTR-España, 2024).

4.3. Castillo core

Radiometric data, with the near total absence of any artificial radionuclide activity (Fig. 3), do not allow establishing an absolute and direct chronology in this core. ²¹⁰Pb_{xs} is cancelled at ~15 cm indicating that sediments below this depth were deposited before 110 to 150 years ago (Appleby and Oldfield, 1978). This core can be divided into two different units or depth intervals (Fig. 3). The basal interval Cast-1 (50–11 cm) comprises brownish sediments with a high sand content (average 51 %), low to very low foraminiferal densities (36 tests/15 g) and scarce marine allochthonous taxa (range 0–5 %) (Table 3). Hyaline forms compose an average 99 %, dominated by *H. germanica*, *E. excavatum*, *A. morphogroup tepida*, and *Elphidium williamsoni* (Table 3). All this suggests a brackish sandy intertidal flat as depositional environment (Pascual et al., 2019; Rodríguez-Lázaro et al., 2013). Vertical profiles of Cu, Pb, Zn, Ni are fairly constant and low, similar to scarcely anthropized historical stages of the Nalón and the nearby Avilés estuaries (García-Ordiales et al., 2020; Gardoki et al., 2021). In fact, the overall values are below the regional geochemical background (Rodríguez et al., 2006; Sanz-Prada et al., 2022). Magnetic susceptibility levels are the lowest found in all the analyzed sediment cores, and the sediments present a very low coal microparticles content. All these characteristics support a chronology before the maximum exploitation of upstream coal and mercury deposits and industrial activities developed in the 20th century. However, the first noticeable appearance of Hg occurring at 32 cm depth (Fig. 3) seems to indicate the beginning of mercury mining activities, which started after mid-19th century (Luque Cabal and Gutiérrez Claverol, 2006).

The upper interval Cast-2 (11–0 cm), constituted by dark-brownish

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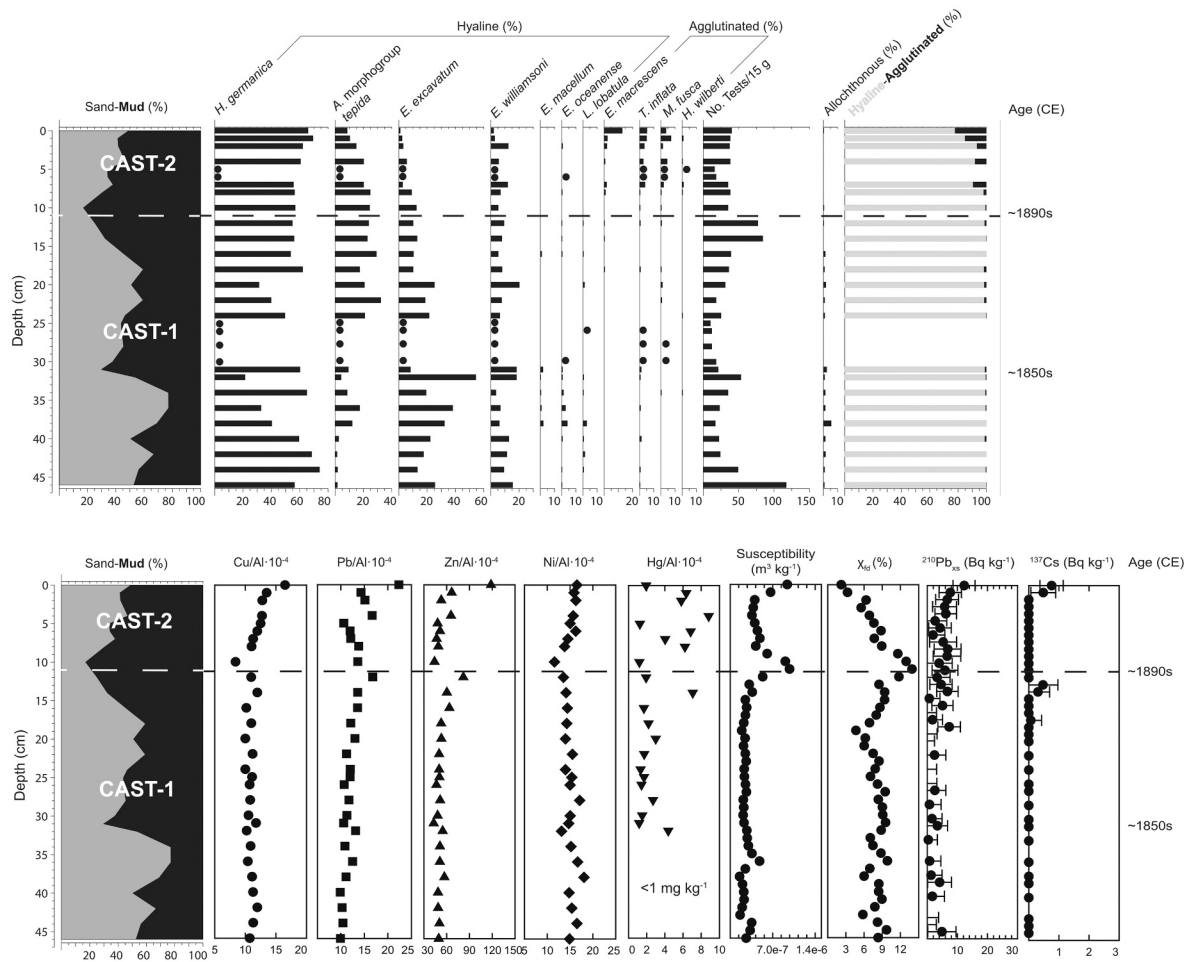


Fig. 3. Vertical distribution of grain-size contents (%), relative abundance of main foraminiferal species (%), Al-normalised trace metals, magnetic susceptibility ($\text{m}^3 \text{kg}^{-1}$) and frequency-dependent susceptibility (%), $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities (Bq kg^{-1}) in the Castillo core, Nalón Estuary. Black dots represent samples with <100 tests.

sediments, shows a lower average sand content (37 %) with the prevailing dominance of *H. germanica* and *A. morphogroup tepida*, and the reduction of *E. williamsoni* and *E. excavatum* (Table 3). In contrast, it is noteworthy the greater presence of agglutinated *T. inflata*, *E. macrescens* and *M. fusca* tests, comprising >22 % of the assemblage in the uppermost part. The abundances of agglutinated low-salinity forms, the reduction of *Elphidium* spp. and the absence of allochthonous taxa (Fig. 3) suggest a shift toward a more restricted intertidal depositional environment, possibly very close to a vegetated marsh (Cearreta, 1994).

Moreover, this interval is characterized by slightly higher coal microparticles content (Table 3) and magnetic susceptibility values compared to the lower unit (Fig. 3). Likewise, the maximum values of the χ_{fd} magnetic parameter are recorded at the base of Cast-2 (Fig. 3), suggesting an initial elevated input of pedogenic materials, probably anthropogenically induced. These changes, concurrently with an overall significant increase in Hg concentrations (Table 2), indicate enhanced coal and mercury mining processes upstream. The increasing mining activities in Asturias were favored by a continued technological development occurring by the end of the 19th century (IGME, 2006; Luque Cabal and Gutiérrez Claverol, 2006). Consequently, the transition to interval Cast-2 could be chronologically framed in the late 19th century, aligning with this period of growth. However, the low $^{210}\text{Pb}_{\text{xs}}$ activity and its cancellation at ~15 cm depth, likely coincides with the final segment of a $^{210}\text{Pb}_{\text{xs}}$ -derived exponential decreasing curve. This

indicates that sediments corresponding to maximum mining and industrial activities have not been recorded due to depositional hiatus and/or postdepositional erosion, in contrast to the continuous record of the Junquera core. As observed by the analysis of the surface samples and field surveys, erosive processes are currently affecting the marshes creating eroding cliffs on both estuarine banks (Fig. S1). Thus, the potential environmental driver explaining the absence of the most recent record could be the currently enhanced fluvial erosion.

4.4. Muros core

The distribution of $^{210}\text{Pb}_{\text{xs}}$ activity is relatively constant in the upper 5 cm, with decreasing values from this depth downward, although its activity is not cancelled at the core base (Fig. 4). The absolute chronology has been obtained by estimating the total inventory (Sanchez-Cabeza and Ruiz-Fernández, 2012), and can be framed between 1965 ± 2 and 2020 CE. The extended presence of large microplastics (Table 2) supports the recent chronology established by radiotracers' activity. Plastic fibers (85 %) constitute the majority of the assemblages, accompanied secondarily by filaments (7 %) and films (6 %) (Fig. S3). Regarding concentrations of trace metals, the overall values are more elevated than those determined in Castillo, whilst the concentrations of Hg ($<1 \text{ mg kg}^{-1}$) are clearly lower. These features, together with the high magnetic susceptibility values (Table 2) and the overall abundant

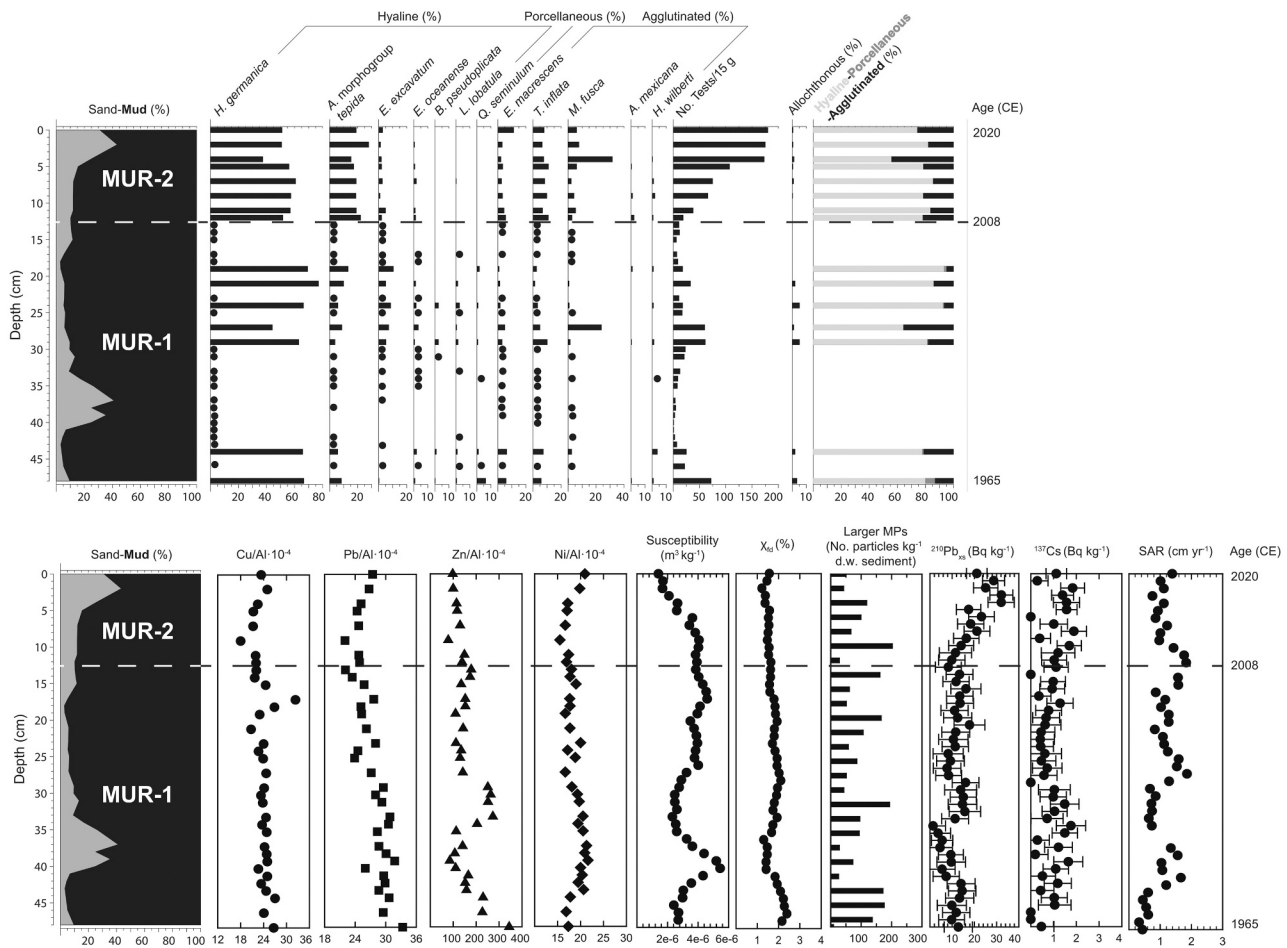


Fig. 4. Vertical distribution of grain-size contents (%), relative abundance of main foraminiferal species (%), Al-normalised trace metals, magnetic susceptibility ($\text{m}^3 \text{kg}^{-1}$) and frequency-dependent susceptibility (%), microplastics content (particles kg^{-1} d.w. sediment), $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities (Bq kg^{-1}), and Sediment Accumulation Rate (cm yr^{-1}) in the Muros core, Nalón Estuary. Black dots represent samples with <100 tests.

coal microparticles in the sediments (Table 3) further support a recent chronology for the entire record.

This core consists of two different depth intervals (Fig. 4). Mur-1 (50–13 cm), characterized by black muddy sediments, presents a low sand content (12 %), and extremely low foraminiferal densities (18 tests/15 g). Only seven samples yielded >100 tests. These samples present *H. germanica* as the dominant species, with secondary abundances of *A. morphogroup tepida*, *E. excavatum*, *T. inflata*, *M. fusca* and *E. macrescens* (Table 3). The rest of the samples with <100 tests contain the same foraminiferal species. The constant and abundant presence of agglutinated marsh taxa throughout this intertidal flat core (Fig. 4) would suggest the erosion of the nearby marshes and the introduction of their buried microfossil content (Fig. S1).

Three sections with negligible presence of microfauna and greater abundances of coal microparticles have been detected in this unit at 45–30 cm, 26–23 cm and 19–13 cm depth, broadly coinciding with increases in sediment accumulation rates of 1.66 cm yr^{-1} , 1.59 cm yr^{-1} and 1.58 cm yr^{-1} respectively (Fig. 4). These episodes and their reduced foraminiferal densities could have arisen due to increases in sediment load, leading to the dilution of the micropaleontological signature. The sediment load would be influenced by the interplay of the naturally elevated transport capacity of the Nalón River, together with historical contributions from coal mining activities (Diz Tirado, 1906; Vera González, 1958). Besides its fluvial and torrential conditions, the effluents coming from inefficient coal washing plants and dumps located

near riverine banks discharged large quantities of coal wastes during the 20th century (Cabrera Ceñal and Flor, 1993). In fact, it was estimated that, despite mining decline in the late 20th century, a total volume of $5,000,000 \text{ m}^3 \text{ yr}^{-1}$ containing $300,000 \text{ t yr}^{-1}$ of fine coal tailings (<0.5 mm) was discharged regionally into public watercourses (Fernández Valcarce, 1992). As a result, coal particle concentrations higher than $40,000 \text{ mg L}^{-1}$ were detected in upstream waters (Fernández Valcarce, 1992), when environmental quality criteria imposed by the Spanish Government limit total suspended solids to a maximum of 100 mg L^{-1} (González and Miranda, 1988). Hence, the sediment infilling, highly conditioned by fine-grained coal wastes, led to extensive dredging activities along the lower Nalón Estuary (Flor et al., 2018; Vera González, 1958).

On the other hand, interval Mur-2 (13–0 cm) exhibits a slightly higher sand content (20 %) and foraminiferal density, increasing from 19 to nearly 200 tests/15 g in the uppermost samples. Hyaline and agglutinated forms constitute an average 77 % and 23 %, respectively. *Haynesina germanica* and *A. morphogroup tepida* appear as main species, whereas agglutinated *T. inflata*, *M. fusca* and *E. macrescens* also stand out with secondary abundances (Table 3). In general terms, Al-normalised concentrations of metals present slightly lower values than in the basal unit, whereas the magnetic susceptibility exhibits a decreasing trend in the upper part. These changes, along with the slight increase in foraminiferal densities and a lower coal microparticles content observed since the early 2000s, could be likely attributed to declining coal mining

activities, the closure of coal washing plants and the enactment of stricter environmental regulations limiting fly ash emissions (Fernández Montes, 2016; Núñez et al., 2008; PRTR-España, 2024).

4.5. San Juan core

Radiometric data exhibit a $^{210}\text{Pb}_{\text{xs}}$ activity close to zero and although ^{137}Cs artificial radiotracer displays a peak near 20 cm depth, this might be attributed to granulometric differences within an overall sandy nature (Fig. 5). Hence, establishing an accurate absolute chronology for this sedimentary sequence is not possible. Vertical profiles of different trace metals (Pb, Zn, Cu and Ni) are quite similar (Fig. 5), characterized by relatively constant values, but significantly higher than those found in Castillo. Hg presents lower concentrations ($<1 \text{ mg kg}^{-1}$) along the entire core, as in Muros intertidal flat deposit. These characteristics, together with the abundant presence of coal microparticles and the high magnetic susceptibility values (Table 2; Table 3), indicate a recent chronology for these sediments.

Two different stratigraphic intervals have been distinguished in this core (Fig. 5), both made of black sediments. The lowermost SJuan-1 (50–28 cm) interval presents an extremely low foraminiferal density (3 tests/15 g), and an average sand content of 70 %. Despite low numbers did not allow calculating relative abundances, the unit is a mixture of *H. germanica*, *M. fusca*, *A. morphogroup tepida* and

E. excavatum, that can be interpreted as a brackish, sandy intertidal flat environment (Pascual et al., 2019), similar to the Nalón modern tidal flats (Table 1). On the contrary, the SJuan-2 upper interval (28–0 cm) exhibits an overall increase in its mud content (50 %) and in the foraminiferal densities (56 tests/15 g). Agglutinated *E. macrescens* and *T. inflata* stand out as the dominant species, together with *H. wilberti* and *M. fusca* (Table 3). These micropaleontological features indicate a transition toward a vegetated marsh (Cearreta, 1994; Leorri et al., 2010; Patterson, 1990).

According to the paleoecological information, this sediment core reveals a temporal evolution pattern toward a well-developed vegetated marsh in a relatively short time span. The basal unit, SJuan-1, could represent the original sandy intertidal flat from the lower estuary bay, prior to the development of the San Juan de la Arena marsh. In fact, according to historical graphic documents, in the early 19th century extensive intertidal flats with restricted marshes characterized this estuarine bay (Adaro, 1979) (Fig. S6). More recently, aerial photographs reveal significant changes in the area during the 20th and 21st centuries (Fig. S6). In 1956, small patches of vegetated marsh areas with a heterogeneous distribution could be observed in the former intertidal flat of the bay (Fig. S6). However, by 1970 the marsh reached its maximum development, with a distinctive crescent-moon shape and a quite dense vegetation (Fig. S6). Therefore, it is reasonable to assume that the shift from SJuan-1 to SJuan-2 intervals occurred rapidly between 1956 and

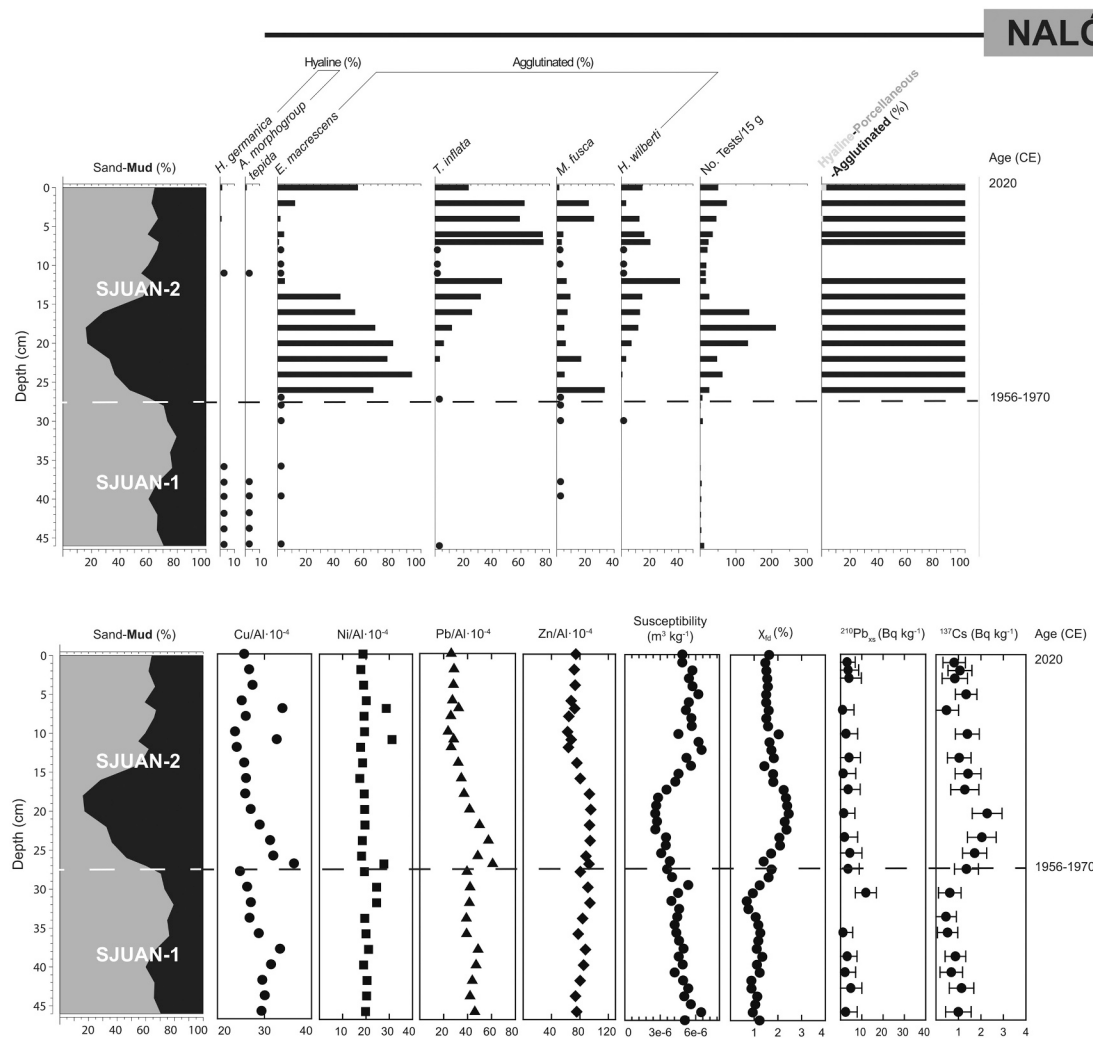


Fig. 5. Vertical distribution of grain-size contents (%), relative abundance of main foraminiferal species (%), Al-normalised trace metals, magnetic susceptibility ($\text{m}^3 \text{kg}^{-1}$) and frequency-dependent susceptibility (%), and $^{210}\text{Pb}_{\text{xs}}$ and ^{137}Cs activities (Bq kg^{-1}) in the San Juan core, Nalón Estuary. Black dots represent samples with <100 tests.

1970 CE. The observed evolution suggests that sediment supply was sufficient to allow rapid vertical accretion, transitioning from a sandy intertidal flat to a well-developed vegetated marsh in a short period of time. As observed by the abundant coal microparticles present along the entire core, sediment availability could be conditioned by the abundant coal wastes from upstream mines and the nearby San Esteban de Pravia port activities during the 20th century (Fig. 1) (Marín Toyos, 1930; Vera González, 1958). Subsequently, between 2004 and 2006, a small dock (130 m in length) was constructed for the mooring of recreational boats in front of the marsh (Fig. S6). Since then, there has been a marked retreat of the marsh due to accelerated erosional processes (Fig. S6). The construction of this dock and the cessation of coal mining waste dumping, probably triggered its current erosion and retreat (Fig. S1c). To date, the San Juan de la Arena marsh is no longer active, presenting a highly developed and stepped eroding cliff with remains of dead vegetation (Fig. S1c). This can also be deduced from the extremely low numbers of living foraminifera populating these marsh sediments (Table 1).

4.6. Environmental assessment of the Nalón Estuary: implications and concerns

Assessing the recent evolution of historically-contaminated coastal systems is an intricate task owing to the large number of perturbations and the complex anthropogenic interactions masking the natural influence. The combined analysis of sediment surface samples and cores has allowed us to achieve a historical and spatial perspective of the sedimentation-erosion processes shaping the evolution of the Nalón Estuary due to merged natural and anthropogenic factors. Fluvial dominance was the primary natural mechanism shaping the estuary during the last centuries, also driving consistently low absolute abundances of benthic foraminifera in the sediments, along with current scarcity of allochthonous and porcelaneous taxa in the middle sector. Natural sediment load of the Nalón Estuary interacted with historical coal mining disposals driving increased sedimentation in its tidal domains. These phenomena were detected by increases in sediment accumulation rates and abundant coal microparticles in the middle estuary, favoring the dilution of geological signatures in their sedimentary records. In the lower estuary bay, rapid sedimentation conditioned vertical accretionary processes during the 20th century, ultimately leading to the formation of the San Juan de la Arena marsh between 1956 and 1970 CE. However, the construction of a port dock and ceasing coal mining disposals led to a negative sedimentary balance, inducing its erosion since 2004–2006. Conversely, our environmental reconstruction has exposed that fluvial activity in the middle sector enhanced erosion in tidal flat sediments with the removal of the most recent sequence recording historical mining activities. These erosive patterns also impacted on marshes from both margins, causing the collapse of large blocks and mixing reworked agglutinated tests with in situ microfauna.

In terms of the environmental quality of the Nalón Estuary, contents of Cu, Pb, Zn and Ni in most samples remain near background values, being distinctly lower than those found in nearby highly polluted estuaries (Table 2) (Cearreta et al., 2000; Irabien et al., 2020; Serrano, 2020; Sierra et al., 2014). In turn, Hg could be considered as an element of environmental concern, given that concentrations in surface sediments from marshes ($n = 2$) and tidal flats ($n = 6$) exceed the toxicological threshold Effect Range Median (ERM; Long et al., 1995), above which adverse effects would frequently occur. Furthermore, both estuarine subenvironments present high abundances of coal microparticles. The currently increased erosion of the marshes is having a noticeable effect on foraminiferal assemblages, but it is unclear whether the marsh deposits are having a direct effect on the concentration of mercury and coal microparticle in contiguous tidal flats. In fact, the highest concentration of Hg in the surface sediments is found at tidal flat station 10 (5.31 mg kg⁻¹), which is not particularly close to the eroding marshes (Fig. 1), further supporting the long-lasting presence of this contaminant

(Janssen et al., 2021; Yeager et al., 2018), even after the closure of mercury mines five decades ago. The persistent presence of mercury in the Nalón Estuary sediments might be attributed to its redistribution from other estuarine environmental compartments or its transport from upstream mining districts in the particulate and dissolved fractions. Initially, mercury transport in the dissolved fractions from to upstream spoil heap leachates and mine water drainages was inferred to be relatively low due to its reduced solubility (Loredo et al., 2010); although later studies reported its presence in these coastal waters (Pavoni et al., 2021).

Regarding the vertical distribution of Hg in the sedimentary deposits, the intervals where concentrations of this metalloid exceed the ERM exhibit a high variability: in the Castillo intertidal flat they are constrained to the uppermost 33 cm ($n = 22$), from mid- to late 19th century, while in the Junquera marsh they can be identified from 0 to 50 cm depth ($n = 22$), 1858 $\pm$ 54 to 2022 CE. Although concentrations are lower than average values determined in mining waste dumps located in upstream districts (Loredo et al., 1999), these estuarine sediments are so enriched in Hg that numerous samples are above the Action Level C (2.84 mg kg⁻¹). According to the Spanish framework for the classification of dredged materials, when values exceed this upper threshold, they could be included within the category of “contaminated materials”, which are not susceptible to offshore dumping (Buceta et al., 2015). Based on this same national regulation, they are still far from the “Non-hazardous sediment” threshold (Table 2). Moreover, the abundant presence of microplastics in intertidal flat and marsh deposits is also noteworthy (Table 2). Given that microplastics have the capacity to transport pollutants (Brennecke et al., 2016), their stratigraphic coincidence in the middle sector marsh with high Hg concentrations raises the urgent need to assess their combined potential ecotoxicological implications. Besides, the extensive presence of unburnt coal particles in this estuary, which are recognized to carry polycyclic aromatic hydrocarbons (Achten and Hofmann, 2009) or induce severe impacts on biota (Berry et al., 2016), also represents a potential environmental threat. Therefore, further studies on the environmental management of these materials are recommended.

Benthic foraminifera, in broad terms, do not seem to show any ecological response to mercury inputs since the mid-19th century until nowadays, neither by changing abundances and diversity nor by morphological deformities of tests in tidal flats and marshes. Foraminiferal densities were already quite low before the onset of mercury mining in the 1850, as observed in the Castillo sedimentary sequence (Fig. 3). On the other hand, taphonomic biases and the 1938 flooding likely caused diminished downcore abundances concomitant with elevated Hg values (Fig. 2), as documented in the Junquera marsh. However, the exceedingly low standing crop at station 10 (63 individuals/80 cm³) (Fig. 1), coexisting with very high Hg levels (Fig. S2d), contributes to the uncertainty surrounding this environmental issue. In fact, detrimental effects of Hg toxicity on benthic foraminifera have been documented in field studies and culture experiments (e.g., Coccioni et al., 2009; Frontalini et al., 2018). Therefore, effects on microfauna exerted by this metalloid at certain areas within the estuary cannot be completely dismissed. Although García-Ordiales et al. (2018) and Pavoni et al. (2021) determined the general reduced formation of methylmercury in the Nalón Estuary, the aforementioned finding underscores the necessity of evaluating the potential ecotoxicity stemming from the different species of this metalloid on benthic (micro) fauna.

Nevertheless, coastal sedimentary deposits are not static geochemical recorders and sinks of pollutants (O'Shea et al., 2018), given their susceptibility to natural and anthropogenic phenomena. In this context, our environmental reconstruction has exposed this estuary as a dynamic and changing system, strongly subjected to fluvial sedimentation and erosion patterns, while vulnerable to riverine floodings. The erosion detected in the intertidal flats from the middle sector has exposed Hg-legacy levels exceeding the ERM threshold by eighteenfold. Therefore,

contaminated sedimentary materials are being remobilized and transferred to other environmental compartments, likely attributed to the significant fluvial influence on the Nalón Estuary. This finding indicates that the Hg-enriched marshes, as well as the potentially affected intertidal flats, can be considered a 'ticking timebomb' acting as a secondary source of contamination. Pollutants, primarily Hg, coal microparticles and microplastics, encapsulated in the sediment layers of these environments can be easily released due to the ongoing natural erosion and the frequent flooding events observed in this estuary. Consequently, in a global climate change and sea-level rise context where extreme hydrological phenomena could be enhanced, the geomorphic processes affecting these deposits with 'trapped' pollutants should be closely monitored.

5. Conclusions

The sedimentary deposits of the Nalón Estuary constitute a multi-archive record of a complex environmental transformation resulting from the interaction of anthropogenic and merged natural processes. The multidisciplinary analysis of surface and core samples from marshes and tidal flats has allowed us to:

- Recognize the high fluvial influence as the primary natural environmental driver that has shaped the estuary during recent times. In this regard, the recorded historical flooding of 1938 stands out as an excellent example of an extreme and short-lived hydrological and geomorphic phenomenon.
- Identify the human-induced changes that condition the recent environmental and morphosedimentary evolution of the estuary. Natural processes (fluvial influence and extreme riverine flooding) interacted with anthropogenic inputs (coal and mercury mining disposals) leading to increasing sedimentation processes, which conditioned high sediment accumulation rates, the dilution of other geological signatures, and the formation of a new marsh in the lower estuary.
- Determine the micropaleontological fingerprints provided by the current erosion of historically-polluted marshes to contiguous tidal flats from the middle sector of the estuary. Further erosion of these marshes can affect the geochemical signature of the intertidal flat sediments, although our data support the long-lasting presence and redistribution of Hg despite the closure of mercury mines five decades ago.
- Deduce that benthic foraminiferal assemblages were mostly conditioned by natural drivers, involving fluvial activity, extreme riverine floodings and taphonomic biases. Furthermore, current Hg toxicity could pose a potential environmental risk to microfauna within the estuary.
- Confirm the remobilization of contaminated sedimentary materials and surface exposure of older sediments with legacy pollutants due to fluvial influence. This finding highlights the importance, in a global climate change scenario, of monitoring the geomorphic

drivers affecting these previously polluted marsh deposits containing also abundant coal microparticles and microplastics.

CRediT authorship contribution statement

Jon Gardoki: Writing – review & editing, Writing – original draft, Visualization, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Alejandro Cearreta:** Writing – review & editing, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **María Jesús Irabien:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Investigation, Funding acquisition, Data curation, Conceptualization. **José Gómez-Arozamena:** Writing – review & editing, Methodology, Investigation. **Víctor Villasante-Marcos:** Writing – review & editing, Methodology, Investigation, Formal analysis. **Ane García-Artola:** Writing – review & editing, Validation, Supervision, Methodology, Data curation. **Filipa Bessa:** Writing – review & editing, Methodology, Investigation.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A

Table A.1

Reference list of estuarine (autochthonous) and marine (allochthonous) foraminiferal species identified in the Nalón Estuary (Asturias, Spain) grouped by wall type.

ESTUARINE (Autochthonous)

Agglutinated test

Ammotium morenoi (Acosta) = *Ammobaculites morenoi* Acosta, 1940

Arenoparrella mexicana (Kornfeld) = *Trochammina inflata* var. *mexicana* Kornfeld, 1931

Balticammina pseudomacrescens Brönnimann, Lutze & Whittaker, 1989

(continued on next page)

Table A.1 (continued)

<i>Entzia macrescens</i> (Brady) = <i>Trochammina inflata</i> (Montagu) var. <i>macrescens</i> Brady, 1870
<i>Haplophragmoides marialensis</i> Andersen, 1953
<i>Haplophragmoides wilberti</i> Andersen, 1953
<i>Lepidodeuterammina ochracea</i> (Williamson) = <i>Rotalina ochracea</i> Williamson, 1858
<i>Miliammina fusca</i> (Brady) = <i>Quinqueloculina fusca</i> Brady, 1870
<i>Scherchorella moniliformis</i> (Siddall) = <i>Reophax moniliforme</i> Siddall, 1886
<i>Siphotrochammina lobata</i> Saunders, 1957
<i>Tiphotrocha comprimata</i> (Cushman & Brönnimann) = <i>Trochammina comprimata</i> Cushman & Brönnimann, 1948
<i>Trochammina inflata</i> (Montagu) = <i>Nautilus inflatus</i> Montagu, 1808
Porcellaneous test
<i>Quinqueloculina seminulum</i> (Linnaeus) = <i>Serpula seminulum</i> Linnaeus, 1758
Hyaline test
<i>Ammonia</i> morphogroup <i>tepida</i> (Cushman) = <i>Rotalia beccarii</i> var. <i>tepida</i> Cushman, 1926
<i>Bolivina britannica</i> Macfadyen, 1942
<i>Bolivina pseudoplicata</i> Heron-Allen & Earland, 1930
<i>Elphidium excavatum</i> (Terquem) = <i>Polystomella excavata</i> Terquem, 1875
<i>Elphidium margaritaceum</i> Cushman, 1930
<i>Elphidium oceanense</i> (d'Orbigny in Fornasini) = <i>Polystomella oceanensis</i> d'Orbigny, 1826
<i>Elphidium williamsoni</i> Haynes, 1973
<i>Haynesina germanica</i> (Ehrenberg) = <i>Nonionina germanica</i> Ehrenberg, 1840
 MARINE (Allochthonous)
Agglutinated test
<i>Ammobaculites agglutinans</i> (d'Orbigny) = <i>Spirolina agglutinans</i> d'Orbigny, 1846
<i>Cribristomoides jeffreysii</i> (Williamson) = <i>Nonionina jeffreysii</i> Williamson, 1858
<i>Eggerelloides scabrum</i> (Williamson) = <i>Bulimina scabra</i> Williamson, 1858
<i>Lagenammina diffugiformis</i> (Brady) = <i>Reophax diffugiformis</i> Brady, 1879
<i>Textularia earlandi</i> Parker, 1952
<i>Textularia sagittula</i> DeFrance, 1824
Porcellaneous test
<i>Adelosina bicornis</i> (Walker & Jacob) = <i>Serpula bicornis</i> Adams, 1798
<i>Adelosina laevigata</i> d'Orbigny, 1826
<i>Massilina secans</i> (d'Orbigny) = <i>Quinqueloculina secans</i> d'Orbigny, 1826
<i>Milionella subrotunda</i> (Montagu) = <i>Vermiculum subrotundum</i> Montagu, 1803
<i>Quinqueloculina dimidiata</i> Terquem, 1876
<i>Quinqueloculina lamarckiana</i> d'Orbigny, 1839
<i>Quinqueloculina lata</i> Terquem, 1876
<i>Spiroloculina depressa</i> d'Orbigny, 1826
<i>Triloculina oblonga</i> (Montagu) = <i>Vermiculum oblongum</i> Montagu, 1803
Hyaline test
<i>Acervulina inhaerens</i> Schulze, 1854
<i>Aubignyna perlucida</i> (Heron-Allen & Earland) = <i>Rotalia perlucida</i> Heron-Allen & Earland, 1913
<i>Bolivina difformis</i> (Williamson) = <i>Textularia variabilis</i> var. <i>difformis</i> Williamson, 1858
<i>Bolivina spathulata</i> (Williamson) = <i>Textularia variabilis</i> var. <i>spathulata</i> Williamson, 1858
<i>Bolivina variabilis</i> (Williamson) = <i>Textularia variabilis</i> Williamson, 1858
<i>Bolivina pseudopunctata</i> (Höglund) = <i>Bolivina pseudopunctata</i> Höglund, 1947
<i>Bulimina gibba</i> Fornasini, 1902
<i>Bulimina marginata</i> d'Orbigny, 1826
<i>Cassidulina laevigata</i> d'Orbigny, 1826
<i>Cassidulina obtusa</i> Williamson, 1858
<i>Cibicides refulgens</i> Montfort, 1808
<i>Cribrorhynchium magellanicum</i> (Heron-Allen & Earland) = <i>Elphidium (Polystomella) magellanicum</i> Heron-Allen & Earland, 1932
<i>Cornuspira involvens</i> (Reuss) = <i>Operculina involvens</i> Reuss, 1850
<i>Elphidium crispum</i> (Linnaeus) = <i>Nautilus crispus</i> Linnaeus, 1758
<i>Elphidium earlandi</i> Cushman, 1936
<i>Elphidium gherti</i> van Voorthuysen, 1957
<i>Elphidium macellum</i> (Fichtel & Moll) = <i>Nautilus macellum</i> Fitchell & Moll, 1798
<i>Elphidium</i> sp. 1
<i>Elphidium</i> sp. 2
<i>Favulina melo</i> (d'Orbigny) = <i>Oolina melo</i> d'Orbigny, 1839
<i>Fursenkoina subacuta</i> (d'Orbigny) = <i>Polymorphina subacuta</i> d'Orbigny, 1852
<i>Gavelinopsis praegeri</i> (Heron-Allen & Earland) = <i>Discorbina praegeri</i> Heron-Allen & Earland, 1913
<i>Haynesina depressula</i> (Walker & Jacob) = <i>Nautilus depressulus</i> Walker & Jacob, 1798
<i>Lagena</i> sp.
<i>Lagenosolenia seguenziana</i> (Fornasini) = <i>Lagena seguenziana</i> Fornasini, 1886
<i>Lobatula lobatula</i> (Walker & Jacob) = <i>Nautilus lobatulus</i> Walker & Jacob, 1798
<i>Planorbulina mediterraneensis</i> d'Orbigny, 1826
<i>Rosalina anomala</i> Terquem, 1875
<i>Rosalina globularis</i> d'Orbigny, 1826
<i>Rosalina irregularis</i> (Rhumbler) = <i>Discorbina irregularis</i> Rhumbler, 1906
<i>Rosalina</i> sp.
<i>Stainforthia fusiformis</i> (Williamson) = <i>Bulimina pupoides</i> var. <i>fusiformis</i> Williamson, 1858
<i>Uvigerina peregrina</i> Cushman, 1923

Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.scitotenv.2024.173792>.

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