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MARINE SEDIMENT POLLUTION IN THE BLACK SEA: SPATIAL DISTRIBUTION, SOURCES, AND CONTAMINATION ASSESSMENT ON THE ROMANIAN SHELF

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ABSTRACT

The Black Sea ecosystem is highly vulnerable to anthropogenic pressures, with its semi-enclosed character favoring the accumulation of pollutants. This study provides an updated and integrated assessment of sediment contamination on the Romanian continental shelf, focusing on persistent hazardous substances: heavy metals (HMs), organochlorine pesticides (OCPs), polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs), and total petroleum hydrocarbons (TPH). Surface sediments collected during 2023 monitoring expeditions were analyzed using standardized and validated techniques, ensuring data reliability and comparability with previous assessments. Results reveal that metals such as Cu, Cd, and Pb show localized exceedances near port areas and river-influenced zones, while OCPs and PCBs display both legacy pollution signals and indications of recent inputs. PAHs were generally below ecological thresholds, although specific hotspots near industrial and maritime activity suggest ongoing sources. TPH concentrations exceeded regulatory limits in a small percentage of samples, particularly in the Constanța Port sector. By integrating multiple contaminant groups and linking the results to ecological quality standards, this work provides the most recent and spatially comprehensive dataset for Romanian Black Sea sediments. The findings highlight persistent and emerging pressures, reinforce the need for continuous monitoring, and directly support national and regional marine strategies under the Marine Strategy Framework Directive (MSFD) and the Black Sea Commission.

Keywords: Black Sea pollution, marine sediments, heavy metals, persistent organic pollutants, contamination assessment

INTRODUCTION

The Black Sea is a semi-enclosed marine basin characterized by limited water exchange and the permanent presence of anoxic conditions below the oxic layer, making it particularly vulnerable to the accumulation of hazardous substances. Due to its natural features and the increasing anthropogenic pressures from riverine inputs, maritime activities, and coastal urbanization, the northwestern Black Sea, especially the Romanian sector (Lazar *et al.*, 2024; Bisinicu *et al.*, 2024), acts as a long-term sink for various contaminants, including heavy metals (HMs), polycyclic aromatic hydrocarbons (PAHs), organochlorine pesticides (OCPs), and polychlorinated biphenyls (PCBs).

Marine sediments play a dual role in the aquatic environment: they serve both as repositories for historical pollution and as potential secondary sources of contaminants, especially under conditions of physical or chemical disturbance. Thus, sediment quality is a key component of marine environmental assessments, particularly within the framework of the Marine Strategy Framework Directive (MSFD), which promotes an ecosystem-based approach to achieving and maintaining Good Environmental Status (GES) of European marine waters.

Previous studies have highlighted the heterogeneous distribution of contaminants in Black Sea sediments, influenced by multiple factors including sediment type, hydrodynamic regime, proximity to pollution sources, and natural geochemical background (Secrieru, 2002; Boicenco *et al*., 2018). These factors must be carefully considered when interpreting concentration levels and assessing ecological risk, particularly in transitional and coastal zones subject to cumulative pressures.

Despite existing research on sediments in the northwestern Black Sea, the majority of available studies remain fragmented and limited to individual groups of pollutants, without providing an integrated assessment across pollutant groups and matrices (Readman et al., 2002; Fillmann et al., 2002; Ţigănuş *et al*., 2013; Nemirovskaya et al., 2024; Bucse et al, 2024; Soloveva et al., 2024; Ristea *et al*., 2025). Comprehensive evaluations that integrate multiple categories of hazardous substances, including HMs, OCPs, PCBs, PAHs, and TPH, in marine sediments across the Romanian continental shelf remain limited, with only a few recent studies addressing this need, such as the integrated assessment based on 2020–2022 monitoring data (Oros et al., 2024).

The limited availability of integrated studies restricts the ability to track emerging contamination hotspots, assess ecological risks in a holistic way, and evaluate progress toward achieving GES under the MSFD, highlighting the importance of assessments that consider multiple pollutant groups within the same environmental matrix.

To address this gap, the present study was guided by the following research questions:

- what is the current contamination status of surface sediments on the Romanian shelf with respect to priority hazardous substances?
- how do spatial distribution patterns of these substances reflect anthropogenic pressures such as riverine discharge, port activities, and industrial sources?
- to what extent do the 2023 data indicate compliance with MSFD Descriptor 8 criteria for Good Environmental Status, and do they reveal areas of potential concern?

In this context, the present study aims to evaluate the contamination levels of key hazardous substances in surface sediments from the Romanian Black Sea shelf, based on monitoring conducted in 2023. The assessment includes spatial distribution analysis of HMs, OCPs, PCBs, PAHs, and total petroleum hydrocarbons (TPH), with comparisons against applicable environmental quality standards. The objective is to identify possible exceedances, assess the achievement of Good Environmental Status (GES) under Descriptor 8 of the MSFD, and provide a scientific basis for future marine monitoring and management efforts.

Overall, this study provides updated and comprehensive data on hazardous substances (HMs, POPs, PAHs, and TPH) in Romanian Black Sea sediments, building upon previous assessments and addressing the ongoing need for continuous environmental monitoring. By integrating the most recent data from 2023, this research contributes essential information to support the evaluation of environmental status under MSFD objectives and establishes a foundation for future long-term assessments. The 2023 dataset provides valuable insights into the current contamination status of Romanian Black Sea sediments, reflecting both ongoing anthropogenic pressures and potential environmental changes in recent years. Compared to previous assessments conducted in the 2012–2017 period (Boicenco *et al.*, 2018), these updated results offer an opportunity to evaluate temporal trends and identify any emerging pollution hotspots that may be linked to shifts in land-based activities, climatic variability, or evolving riverine inputs, particularly from the Danube.

Moreover, recent policy developments at national and regional levels, such as the integration of new priority substances under the Water Framework Directive and the refinement of Good Environmental Status (GES) criteria (Tornero *et al*., 2021), further underscore the need for sustained and harmonized monitoring efforts (Katsanevakis *et al.,* 2025; Nikolaou *et al*., 2025). Maintaining continuity in sediment quality assessments is essential not only for verifying the effectiveness of mitigation strategies but also for anticipating long-term ecological risks (Dorleon *et al*., 2025). Such continuity ensures data comparability, supports adaptive marine management, and contributes to regional cooperation within the Black Sea basin.

In the following sections, the study area and methods are described, the results on sediment contamination are presented and discussed across multiple pollutant groups (HMs, OCPs, PCBs, PAHs, and TPH) and the key findings are synthesized together with their limitations, implications, and recommendations to support future monitoring and management.

MATERIALS AND METHODS

Study Area and Sampling

A monitoring expedition was conducted from 4 to 13 October 2023 along the Romanian Black Sea coast, covering the continental shelf up to the 50–60 m isobath. A total of 32 surface sediment samples were collected from a variety of locations (Fig. 1), spanning transitional, coastal and marine waters. Stations belong to the network of the long-term monitoring program coordinated by the National Institute for Marine Research and Development (NIMRD) “Grigore Antipa”. Their locations were selected to ensure representative spatial coverage of transitional, coastal, and marine areas on the Romanian continental shelf.

Sediment samples were obtained using a Van Veen grab sampler. Only the top layer of the sediment was carefully sub-sampled, transferred into pre-labeled containers or polyethylene bags, and stored at –20 to –24 °C until laboratory analysis, following UNEP/FAO/IOC/IAEA (1995) standard protocols.

Sample Preparation

In the laboratory, sediments were freeze-dried, homogenized, and sieved to remove coarse material (>1 mm). All samples destined for contaminant analysis were processed in accordance with established protocols (UNEP/FAO/IOC/IAEA, 1995).

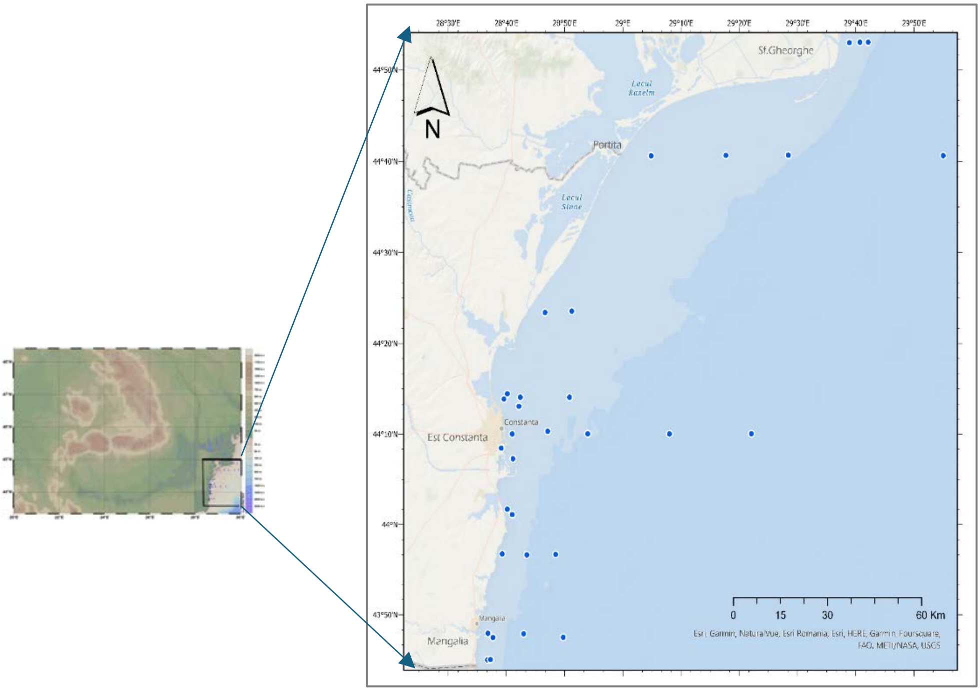

Fig. 1. Map of sediment sampling stations on the Romanian Black Sea coast – October 2023

Heavy Metals (HMs)

Concentrations of heavy metals (Cu, Cd, Pb, Ni, Cr, and Co) were determined following acid digestion using HNO_3 (65%, Suprapur) in closed Teflon vessels. Digestion was carried out using either a microwave system or a hotplate. After digestion and cooling, samples were diluted to a final volume of 100 mL with deionized water. Metals were quantified using graphite furnace atomic absorption spectrometry (GF-AAS) on a High-Resolution Continuum Source AAS (HR-CS ContrAA 800 G equipment, Analytik Jena, Jena, Germany). Calibration curves were established for each element using 5-point standards ranging from 0–100 µg/L. Each sample was analyzed in triplicate, and mean concentrations were reported. Detection limits range from 0.001 to 0.01 µg/L depending on the element (IAEA-MEL, 1999).

Persistent Organic Pollutants (POPs): Organochlorine pesticides (OCPs) and Polychlorinated biphenyls (PCBs)

Organochlorine pesticides (OCPs) and polychlorinated biphenyls (PCBs) were extracted from sediment samples using a hexane/acetone (1:1 v/v) mixture with microwave-assisted extraction (30 minutes at 120 °C; Start E Milestone system). Extracts were concentrated by rotary evaporation, purified using Florisil columns, and subjected to sulfur removal with copper. Final analysis was performed using gas chromatography with an electron capture detector (GC-ECD), as described in IAEA-MEL (1995), on a Perkin Elmer (USA) gas chromatograph equipped with electron capture and mass spectrometry detectors. Target analytes included HCB, lindane, aldrin, dieldrin, endrin, DDT and its metabolites, and seven indicator PCBs (PCB 28, 52, 101, 118, 138, 153, 180).

Polycyclic Aromatic Hydrocarbons (PAHs)

PAHs were extracted using a similar protocol to POPs, with hexane/acetone (1:1 v/v) and microwave-assisted extraction (30 minutes at 120 °C). After concentration and cleanup, samples were analyzed by gas chromatography coupled with mass spectrometry (GC-MS). Analytes included 16 priority PAHs such as fluorene, phenanthrene, anthracene, and benzo[a]pyrene, following methods validated by IAEA-MEL (1995).

Total Petroleum Hydrocarbons (TPH)

TPH in sediment was extracted with a hexane/dichloromethane mixture (7:3 v/v), and the concentrated extracts were analyzed fluorimetrically using the Fluorat-02-3M analyzer (200–950 nm detection range), as per IAEA-MEL (1995).

Data Analysis and Mapping

Statistical analysis was performed using MS Excel 365, Statistica (TIBCO Software Version 14.0.1.25), Ocean Data View (ODV) version 5.1.7., and ArcGIS Desktop 10.7 software. Distribution maps in ArcGIS were generated through inverse distance weighting (IDW) interpolation, a method that calculates cell values based on a weighted combination of nearby sampling points.

A schematic workflow summarizing the sequence of methodological steps applied in this study is presented in Fig. 2.

Fig. 2. Workflow of the research methodology applied in this study, from study area and sampling to sample preparation, laboratory analyses of pollutants (HM, POP, PAH, TPH), and final data analysis and mapping.

RESULTS AND DISCUSSION

The data resulted from the investigations carried out in 2023 on the levels of contaminants in the Romanian Black Sea sediments are presented herein, structured within 4 tables and 22 figures (9 distribution maps, 7 variability plots, 6 histograms).

Heavy Metals

The investigation conducted in 2023 on HM levels in surface marine sediments along the Romanian Black Sea coast revealed the following average concentrations and ranges (Table 1):

- Copper: 11.263 ± 10.882 µg/g (range: 1.622–61.140 µg/g),
- Cadmium: 0.414 ± 0.595 µg/g (0.053–2.837 µg/g),
- Lead: 14.364 ± 8.930 µg/g (3.095–43.160 µg/g),
- Nickel: 14.765 ± 6.760 µg/g (5.431–28.730 µg/g),
- Chromium: 11.328 ± 4.691 µg/g (4.962–24.260 µg/g),
- Cobalt: 2.896 ± 0.838 µg/g (1.403–4.815 µg/g).

The ecological status of marine sediments was evaluated according to threshold values proposed for defining Good Environmental Status (GES) under the Marine Strategy Framework Directive. For this purpose, Effects Range-Low (ERL) values were applied, as recommended for assessing sediment quality and the ecological relevance of hazardous substance concentrations (Long *et al*., 1998).

Tabel 1. Concentrations of heavy metals determined in marine sediments from the Romanian coast in 2023

	Average	Median	Min	Max	Percentile 25th	Percentile 75th	Std.Dev.
Cu (µg/g)	11.263	7.929	1.622	61.140	5.202	15.160	10.882
Cd (µg/g)	0.414	0.232	0.053	2.837	0.121	0.342	0.595
Pb (µg/g)	14.364	12.585	3.095	43.160	7.385	20.665	8.930
Ni (µg/g)	14.765	14.155	5.431	28.730	9.068	19.120	6.760
Cr (µg/g)	11.328	10.525	4.962	24.260	7.8495	13.780	4.691
Co (µg/g)	2.896	3.019	1.403	4.815	2.3875	3.507	0.838

For copper, 91% of concentrations, especially in coastal areas, were below 20 µg/g. Slightly elevated values (up to 25 µg/g) were recorded in 6% of samples from river-influenced transitional waters and deeper marine areas beyond the 30 m isobath, notably in the Portița transect (northern sector), East Constanța at 50–60 m (central sector), and Mangalia 50 m (southern sector). However, extreme values exceeding the 40 µg/g GES threshold were measured in 3% of samples, specifically at Constanța Port stations (Constanța Sud 10 m, 20 m), indicating strong anthropogenic influence (e.g., wastewater discharge, port activities) (Fig. 3).

Cadmium concentrations were below 0.5 µg/g in 84% of samples. Slightly elevated levels (0.5–1.0 µg/g) were observed in 6% of samples, particularly in transitional waters (e.g., Portița 20 m) and coastal sediments within the Constanța Port area (Constanța Sud 10 m). Notably, the ERL of 1.2 µg/g was exceeded in 9% of samples, including East Constanța transect: 10 m (harbor area), 50 m, and 60 m, likely linked to increased maritime traffic and historical accumulation in fine-grained sediments (Fig. 4).

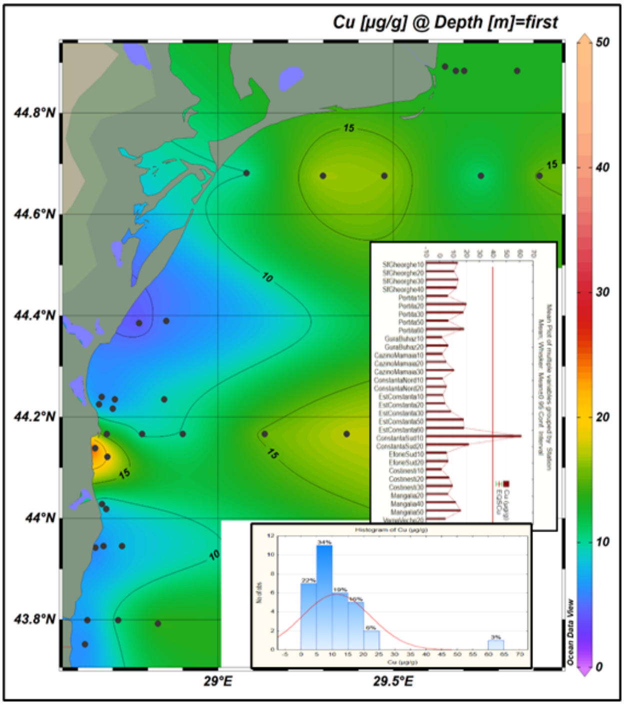

Fig. 3. Distribution of copper concentrations in sediments from the Romanian seaside in 2023 (distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

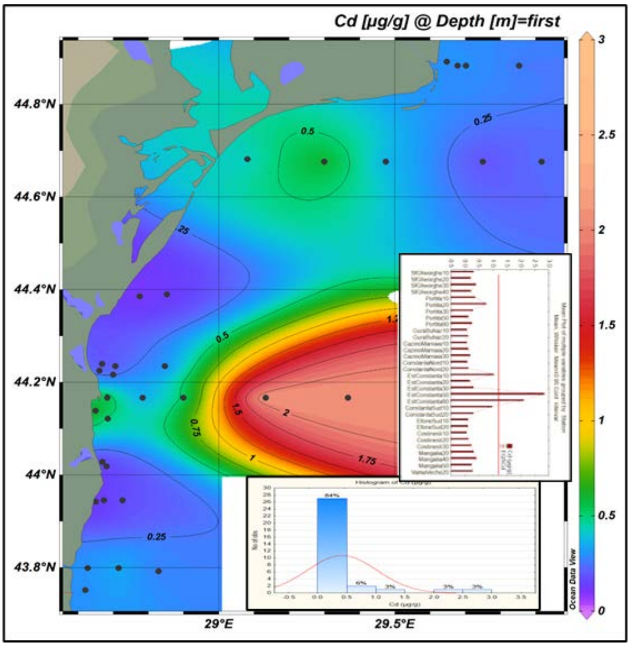

Fig. 4. Distribution of cadmium concentrations in sediments from the Romanian coast in 2023 (distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

Lead displayed a gradient of increasing concentrations from coastal to offshore stations, with the exception of the highest values (> 40 µg/g) recorded near Constanța Port. These represented 3% of total samples. However, the ERL value of 47 µg/g was not exceeded, and 97% of all samples had Pb concentrations below 30 µg/g, indicating generally low ecological risk (Fig. 5).

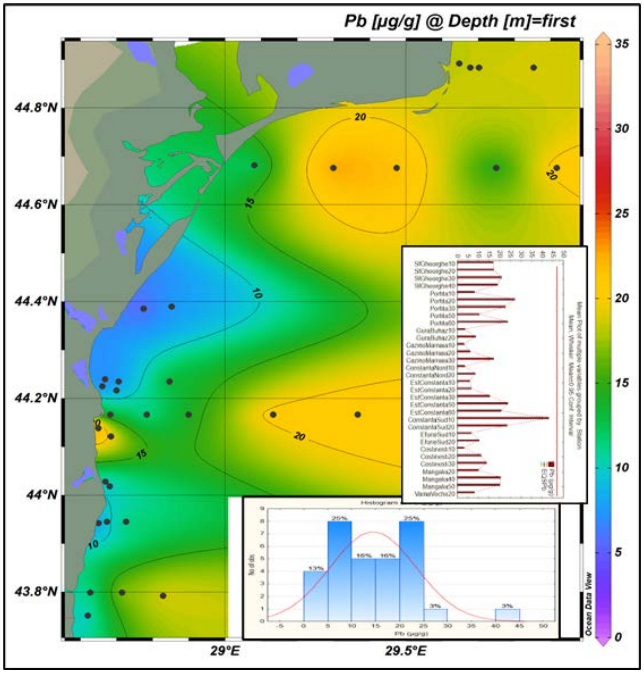

Fig. 5. Distribution of lead concentrations in sediments from the Romanian seaside in 2023 (distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

Nickel concentrations were slightly higher in the northern sector and deeper offshore sediments, with a clear gradient from coastal to marine areas. Most values (88%) were below 22 µg/g, while 12% of samples, from Portița 20 m, 30 m; East Constanța 60 m; Constanța Sud 20 m, showed moderately elevated levels. Nevertheless, all concentration remained below the GES threshold of 35 µg/g (Fig. 6).

Chromium followed a distribution pattern similar to nickel, with higher concentrations observed in transitional and offshore sediments compared to nearshore zones. Most samples (91%) showed Cr concentrations below 16 µg/g, while 9% exceeded 20 µg/g, notably at Portița 20 m, 30 m, and Constanța Sud 10 m, yet all remained well below the ERL of 81 µg/g (Fig. 7).

Cobalt showed slightly elevated values in the fluvially influenced zone, particularly at Sf. Gheorghe 30 m and Portița 30 m, with 6% of samples exceeding 4 µg/g. The lowest concentrations were observed in central coastal sediments, with a general increasing gradient toward marine areas. In the southern sector, values were more uniformly distributed (Fig. 8).

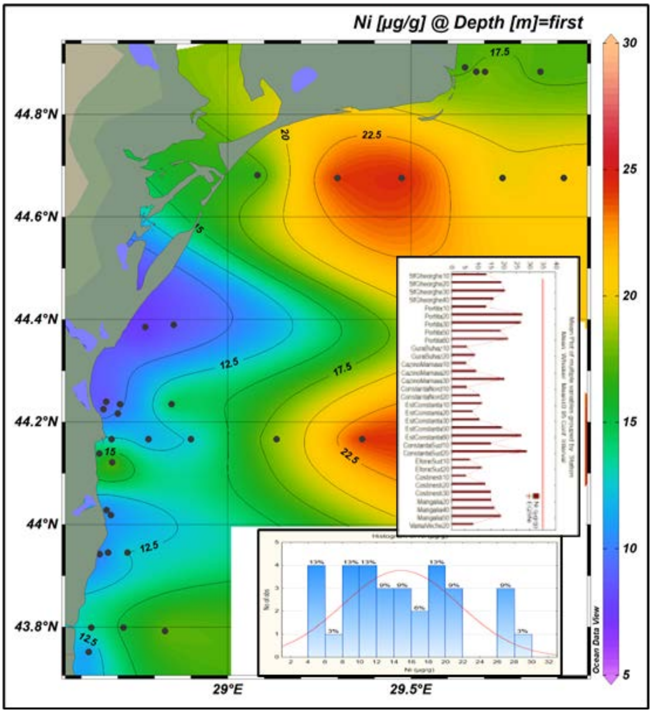

Fig. 6. Distribution of nickel concentrations in sediments from the Romanian seaside in 2023
(distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

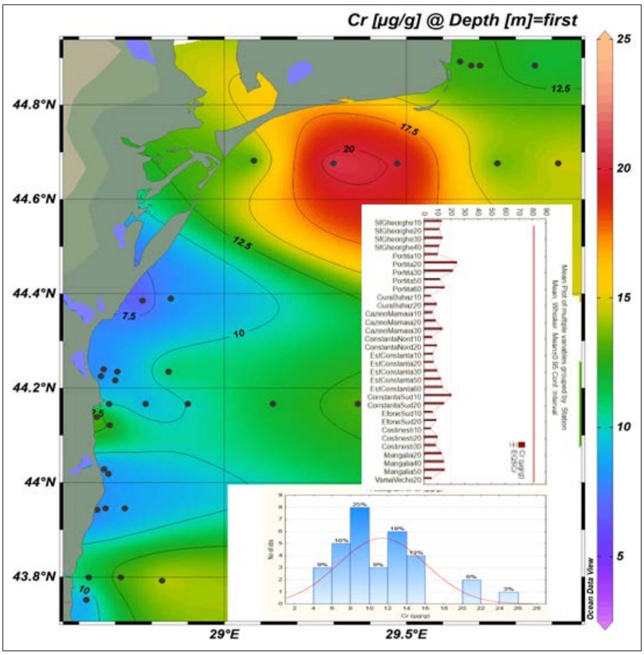

Fig. 7. Distribution of chromium concentrations in sediments from the Romanian coast in 2023
(distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

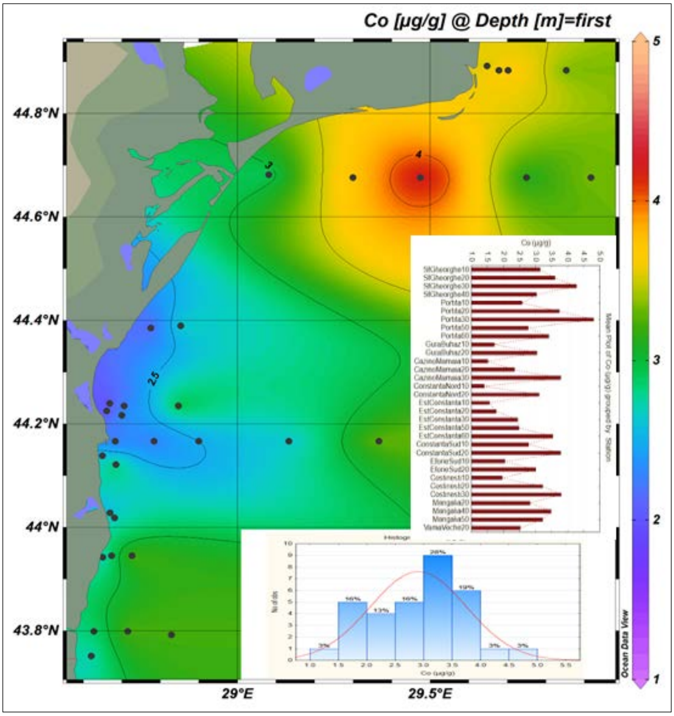

Fig. 8. Distribution of cobalt concentrations in sediments from the Romanian coast in 2023 (distribution map (ODV 5.1.7); variability plot and histogram (Statistica 14.0.1.25)

The highest accumulation of heavy metals was observed in river-influenced transitional waters and deeper marine stations beyond 30 m depth, where sediment characteristics (fine particles, high organic matter) promote retention. In contrast, coastal sediments generally exhibited lower concentrations, except within and near Constanța Port, where strong anthropogenic pressures (e.g., wastewater discharge, port operations, maritime traffic) were evident. Despite localized exceedances, the percentage of samples exceeding quality standards was below the 25% threshold set for achieving GES. Therefore, the 2023 evaluation of the HM indicator in sediments reflected Good Environmental Status (GES) for all monitored elements (copper, cadmium, lead, nickel, chromium, cobalt).

Overall, HMs assessed in marine sediments along the Romanian coast showed highest accumulations primarily in river-influenced transitional zones and deeper marine stations (>30 m), likely due to sediment characteristics favorable to pollutant retention. While localized exceedances occurred, particularly near Constanța Port, overall metal concentrations indicated a generally GES under MSFD criteria.

Heavy metals (HMs) are naturally occurring components of the marine environment; however, human activities such as extraction, industrial use, and waste discharge can lead to their dangerous accumulation in marine ecosystems (El-Sharkawy *et al.* 2025).

Marine sediments play a critical role as sinks for metals, which may accumulate concentrations toxic to both marine organisms and humans (Özșeker *et al*., 2022).

Metals can be classified into two categories: essential metals, such as zinc (Zn), selenium (Se), iron (Fe), copper (Cu), chromium (Cr), and molybdenum (Mo), which are involved in biological functions; and non-essential metals, such as lead (Pb) and cadmium (Cd), which have no known physiological roles and can be toxic even at low concentrations. Through the process of remobilization, metals stored in sediments may re-enter the water column and affect the marine environment over the long term (White & Rainbow, 1985; Depledge & Rainbow, 1990; OSPAR, 1992).

The primary sources of HM contamination in the marine environment include urban and industrial discharges, port and offshore activities, and riverine inputs. Rivers transports significant quantities of metals in particulate form into the sea, especially during extreme hydrological events such as floods (UNEP, 2002; UNEP, 2006). Atmospheric deposition also plays an important role in supplying metals to coastal zones and the wider basin and is influenced by climate and meteorological variability (Fashola *et al*., 2016; Sakson *et al*., 2018).

Because HM are non-degradable pollutants, they can disrupt the biogeochemical balance of contaminated environments. Through processes such as bio-adsorption, metals accumulate within cells, where they can exert toxic effects (Ding *et al.,* 2022). Mechanisms such as membrane transport and passive diffusion facilitate their internal distribution within organisms (Wang & Fisher, 1997). Although marine sediments initially trap metals through deposition onto fine particles, they can also become a secondary source of pollution through remobilization processes, including resuspension, bioturbation, and molecular diffusion (Hazrat *et al*., 2019; Richir & Gobert, 2016).

Under changing environmental conditions, sediments may release metals back into the overlying water, which underscores the importance of monitoring metal concentrations in sediments to assess ecological risks. These concentrations are influenced by both anthropogenic and natural sources, as well as sediment characteristics such as grain size and organic matter content (Zhuang & Gao, 2014; Wang et al., 2023).

Biogeochemical studies in the northwestern Black Sea emphasize the key role of the Danube River and other local sources in delivering metals to the continental shelf, where fine, organic-rich sediments tend to accumulate higher levels of heavy metals (Tankere S.P.C., 2001; Bucse *et al.,* 2024; Oros *et al.,* 2025).

The most recent evaluations of the HM indicator in sediments, conducted for the 2012–2017 period (Boicenco *et al*., 2018), indicated Good Environmental Status (GES) for some elements in specific marine reporting units. However, when applying the threshold criterion of no more than 25% of samples exceeding the environmental limit values, the data showed failures to achieve GES for certain metals: copper showed poor status in both marine and transitional waters, cadmium showed poor status in transitional areas, nickel failed to meet GES in all surveyed regions.

HMs tend to accumulate over time in marine sediments as a result of deposition from the water column. Their concentrations are strongly influenced by sediment granulometry and organic matter content, both of which affect the retention and mobility of metals (Han *et al*., 2024; Chen *et al*., 2025). However, it is important to note that, especially in the case of nickel, the natural background levels in Black Sea marine sediments may exceed commonly recommended threshold values, due to local geological and geochemical conditions (Secrieru, 2002). Therefore, when assessing heavy metal concentrations in sediments, integrated evaluations of ecological status should be approached with caution and must take into account the specific characteristics of the region (Oros, 2019).

Persistent Organic Pollutants (POPs): Organochlorine pesticides (OCPs) and Polychlorinated biphenyls (PCBs)

In 2023, concentrations of organochlorine pesticides (OCPs) determined in surface marine sediments along the Romanian coast of the Black Sea ranged from the limit of detection (LoD) to the following maximum values: HCB: LoD–0.050 µg/g d.w., lindane: LoD–6.688 µg/g d.w., heptachlor: LoD–2.863 µg/g d.w., aldrin: LoD–4.313 µg/g d.w., dieldrin: LoD–0.557 µg/g d.w., endrin: LoD–0.776 µg/g d.w., p,p’ DDE: LoD–1.180 µg/g d.w., p,p’ DDD: LoD–8.583 µg/g d.w., and p,p’ DDT: LoD–5.786 µg/g d.w. (Table 2).

Tabel 2. Concentrations of organochlorine pesticides in marine sediments in 2023

Compound (µg/g)	Average	Median	Minimum*	Maximum	Percentile 75th	Percentile 90th	Dev. Std.
HCB	0.0041	0.0003	0.0003	0.0507	0.0003	0.0166	0.0113
Lindan	0.7022	0.0003	0.0003	6.6882	0.0003	3.7534	1.7794
Heptaclor	0.2574	0.0002	0.0002	2.8631	0.0002	0.5469	0.7634
Aldrin	0.2873	0.0002	0.0002	4.3134	0.0957	0.4465	0.8634
Dieldrin	0.0439	0.0002	0.0002	0.5579	0.0663	0.1142	0.1056
Endrin	0.0276	0.0003	0.0003	0.7765	0.0003	0.0189	0.1369
p.p’DDE	0.0425	0.0002	0.0002	1.1806	0.0002	0.0002	0.2099
p.p’DDD	1.6488	0.8477	0.0002	8.5832	1.9967	5.3566	2.2709
p.p’DDT	0.3276	0.0002	0.0002	5.7864	0.0002	1.2262	1.0997

*Limit of detection

The Romanian Black Sea coastal sediments were dominated by the presence of p,p’ DDD, especially in the northern sector (Sfântu Gheorghe profile), but more significantly in the central and southern sectors (Constanța Sud, Eforie, Costinești, Mangalia, and Vama Veche profiles). This distribution pattern reflects historical pollution with p,p’ DDT, which, over time, degrades into its metabolites p,p’ DDD and p,p’ DDE. Evidence of more recent p,p’ DDT pollution was observed in 2023, notably in the Portița area and the East Constanța profile (Fig 9).

Overall, in 2023, sediment samples were dominated by the presence of p,p’ DDD, clearly indicating historical pollution with p,p’ DDT, a compound that, over time, degrades into its metabolites p,p’ DDD and p,p’ DDE. However, evidence of recent contamination was also identified, based on the elevated concentrations of p,p’ DDT found in sediments from the northern and central sectors, particularly in the East Constanța area. These findings suggest ongoing inputs from diffuse sources, most likely through atmospheric deposition.

The ecological status of marine sediments was assessed based on threshold values proposed for defining Good Environmental Status (GES) under the Marine Strategy Framework Directive. For this purpose, the Effects Range-Low (ERL) values were used to evaluate the ecological relevance of the hazardous substances detected in sediments (Long *et al.,* 1998). Exceedances of GES thresholds were recorded particularly for lindane (22%) and dieldrin (31%), while HCB and p,p' DDE exceeded threshold values in 6% of samples.

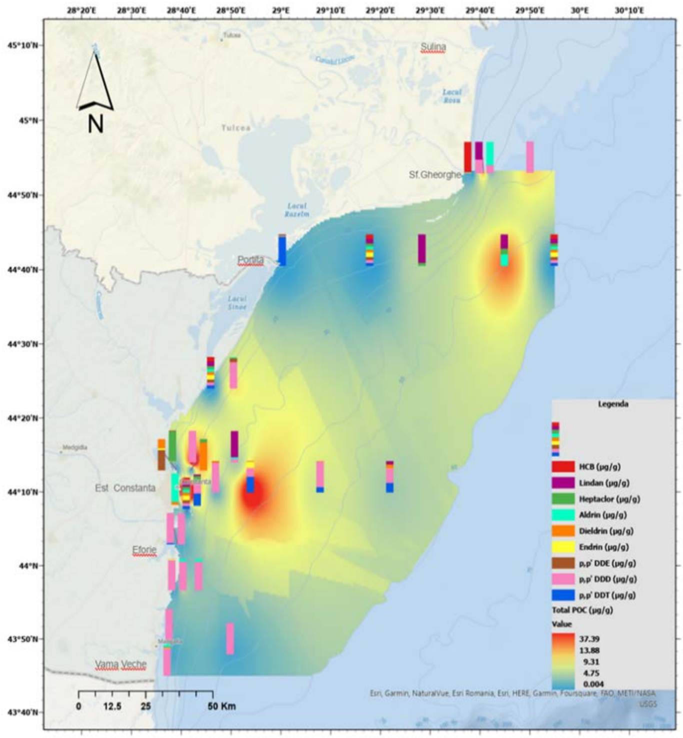

Fig. 9. Distribution of organochlorine pesticides (µg/g) and the percentage contribution of each compound to the total (%) in sediments from the Romanian coast, in 2023 (ArcGIS Desktop 10.7)

For the other compounds: heptachlor, aldrin, endrin, p,p' DDD, and p,p' DDT, threshold values for defining GES have not yet been established under the MSFD framework. In 2023, concentrations of polychlorinated biphenyls (PCBs) determined in surface marine sediments from the Romanian Black Sea coast ranged from the limit of detection (LoD) to the following maximum values: PCB28: LoD–0.168 µg/g d.w., PCB52: LoD–6.526 µg/g d.w., PCB101: LoD–7.437 µg/g d.w., PCB118: LoD–8.778 µg/g d.w., PCB138: LoD–11.120 µg/g d.w., PCB153: LoD–0.205 µg/g d.w., and PCB180: LoD–0.177 µg/g d.w. (Table 3).

Sediments from the central and southern sectors of the Romanian Black Sea coast were dominated by the presence of PCB 153, PCB 118, and PCB 101, with the highest PCB accumulations observed in the vicinity of Midia Port and the Rompetrol refinery, as well as in the southernmost areas, from Eforie to Vama Veche. In the northern sector, PCB 52 was predominant; however, PCB 153, PCB 118, and PCB 101 were also detected in significant concentrations at several stations (Fig. 10).

Frequent exceedances of the threshold values proposed for defining Good Environmental Status (GES) were recorded for multiple congeners: PCB 153 exceeded the threshold in 69% of samples, PCB 101 in 59%, PCB 118 in 50%, and PCB 52 in 41% of samples. Other PCB congeners also showed exceedances, but at much lower frequencies.

Tabel 3. Concentrations of polychlorinated biphenyls in shallow marine sediments in 2023

Compound (µg/g)	Average	Median	Minimum*	Maximum	Percentile 75th	Percentile 90th	Dev. Std.
PCB 28	0.0085	0.0004	0.0004	0.1681	0.0004	0.0004	0.0313
PCB 52	0.4377	0.0003	0.0003	6.5262	0.0528	0.4048	1.4371
PCB 101	1.0034	0.0123	0.0003	7.4371	0.2587	4.8434	2.0781
PCB 118	0.7463	0.0041	0.0004	8.7785	0.0350	0.0831	2.3390
PCB 138	1.7407	0.0417	0.0006	11.1201	0.0936	7.5231	3.3925
PCB 153	0.0105	0.0007	0.0007	0.2054	0.0007	0.0145	0.0367
PCB 180	0.0058	0.0003	0.0003	0.1773	0.0003	0.0003	0.0313

*Limit of detection

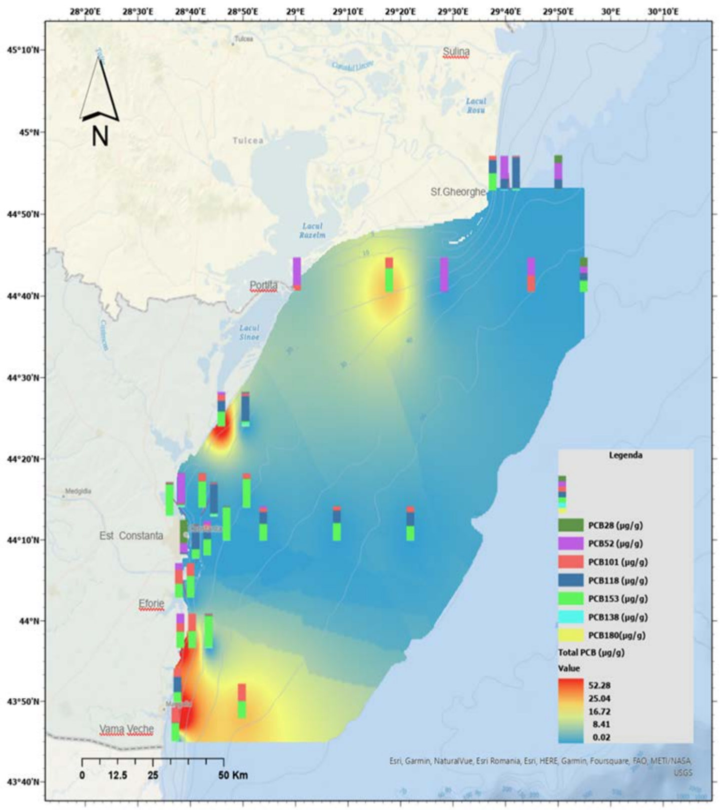

Fig. 10. Distribution of polychlorinated biphenyls (µg/g) and the percentage contribution of each compound to the total (%) in marine sediments on the Romanian coast, in 2023 (ArcGIS Desktop 10.7)

The highest PCB accumulations in sediments were observed in the vicinity of Midia Port and the Rompetrol refinery, as well as in the southernmost coastal areas, from Eforie to Vama Veche, where frequent exceedances of the threshold values proposed for defining Good Environmental Status were recorded.

Persistent organic pollutants (POPs), such as organochlorine pesticides and polychlorinated biphenyls (PCBs), are a class of synthetic chemicals characterized by their high toxicity and exceptional resistance to environmental degradation (Kumar *et al*., 2022). These compounds, known for their carcinogenic, neurotoxic, and endocrine-disrupting properties, have the ability to bioaccumulate organisms and are transported globally via air, water, and migratory species (Zhang *et al*., 2024). As a result, they pose long-term risks to both aquatic and terrestrial ecosystems (Ritter *et al*., 1995; Xing *et al*., 2005; Muhammad Aqeel Ashraf, 2017).

Studies conducted in the Romanian sector of the Black Sea have consistently documented the persistence of POPs in marine sediments, particularly in heavily anthropized coastal areas, where observed concentrations frequently exceed environmental quality standards (Boicenco *et al.,* 2012; Coatu *et al*., 2013; ANEMONE Deliverable 2.1, 2021).

Although Romania ratified the Stockholm Convention on POPs, adopting national restrictions on substances such as DDT, aldrin, heptachlor, and PCBs (Law No. 261/2004), the continued detection of these compounds in marine sediments suggests the presence of ongoing or residual contamination sources. The retention of POPs in sediments is strongly influenced by sediment characteristics, especially fine grain size and high organic matter content, which promote their accumulation. Furthermore, remobilization processes, including bioturbation and resuspension, can lead to the release of POPs back into the water column (Mikheeva *et al,* 2022). This not only increases ecological risk but also underscores the importance of regular sediment monitoring to assess the long-term impacts of these substances on the marine environment (ANEMONE Deliverable 2.3, 2021).

Overall, the distribution patterns of POPs, including OCPs and PCBs, highlighted persistent historical contamination coupled with ongoing input sources, particularly near heavily anthropized areas such as Constanța Port and Midia. Elevated levels of specific pollutants (e.g., lindane, dieldrin, PCB congeners) suggest the continued need for targeted management measures to reduce ecological risks.

Although many organochlorine pesticides (e.g., DDT, aldrin, heptachlor) and PCBs have been banned under international conventions, their continued presence in marine sediments indicates the persistence of legacy contamination. These compounds are known to remain in the environment for decades due to their chemical stability and strong affinity for organic matter in sediments (Asaoka *et al.,* 2019). However, recent detections of parent compounds such as p,p'-DDT, as opposed to only its degradation products (p,p'-DDD and p,p'-DDE), suggest the possibility of more recent inputs. Potential current sources may include leakage from obsolete pesticide stockpiles, illegal or uncontrolled use in agriculture, and long-range atmospheric transport from regions where restrictions may be less stringent. This dual origin, legacy and recent, emphasizes the need for continued monitoring and control, as even low-level but sustained inputs can compromise sediment quality and pose risks to marine biota over time.

Polycyclic Aromatic Hydrocarbons (PAHs)

In 2023, the concentrations of polycyclic aromatic hydrocarbons (PAHs) determined in surface marine sediments along the Romanian Black Sea coast ranged from the limit of detection up to a maximum of 0.428 µg/g dry weight (Table 4). With the exception of fluorene, anthracene, and indeno(1,2,3-c,d)pyrene, most PAH compounds were detected at concentrations close to the detection limit.

Tabel 4. Polycyclic aromatic hydrocarbon concentrations in sediments from the Romanian Black Sea coast in 2023

Compound (µg/g)	Average	Median	Min*	Max	Percentile 25th	Percentile 75th	Dev. Std
Naftalina	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Acenaftilen	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Acenaften	0.000097	0.0001	0.0000	0.0001	0.0001	0.0001	0.000018
Fluoren	0.010006	0.0001	0.0001	0.14783	0.0001	0.0001	0.036844
Fenantren	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Antracen	0.014656	0.0001	0.0001	0.428375	0.0001	0.0001	0.076847
Fluoranten	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Piren	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Benzo[a]antracen	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Crisen (µg/g)	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Benzo[b]fluoranten	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Benzo[k]fluoranten	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Benzo[a]piren	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Benzo (g.h.i)perilen	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Dibenzo(a.h)antracen	0.0001	0.0001	0.0001	0.0001	0.0001	0.0001	0.0000
Indeno(1.2.3-c.d)piren	0.014459	0.0001	0.0001	0.320350	0.000011	0.0001	0.059969

The dominant PAH compounds in sediments were fluorene, anthracene, and indeno(1,2,3-c,d)pyrene, primarily found in the Danube-influenced area, particularly along the Sfântu Gheorghe profile (Fig. 11). The ecological status of the marine sediments was assessed using threshold values proposed for defining Good Environmental Status (GES) under the Marine Strategy Framework Directive (MSFD).

For this assessment, Effects Range-Low (ERL) values were applied to evaluate sediment quality and the ecological significance of the measured concentrations of hazardous substances (Long et al., 1998). It is noteworthy that none of the PAH concentrations exceeded the thresholds established for defining GES. Therefore, based on this indicator, it can be concluded that in 2023 the quality of marine sediments along the Romanian coast was good with respect to polycyclic aromatic hydrocarbon contamination.

Overall, PAH concentrations across the Romanian Black Sea coast were generally low, with no exceedances of ecological thresholds. Dominance by certain compounds (fluorene, anthracene, indeno(1,2,3-c,d)pyrene) mainly near the Danube-influenced area reflects limited but specific pollution inputs, supporting an overall Good Environmental Status.

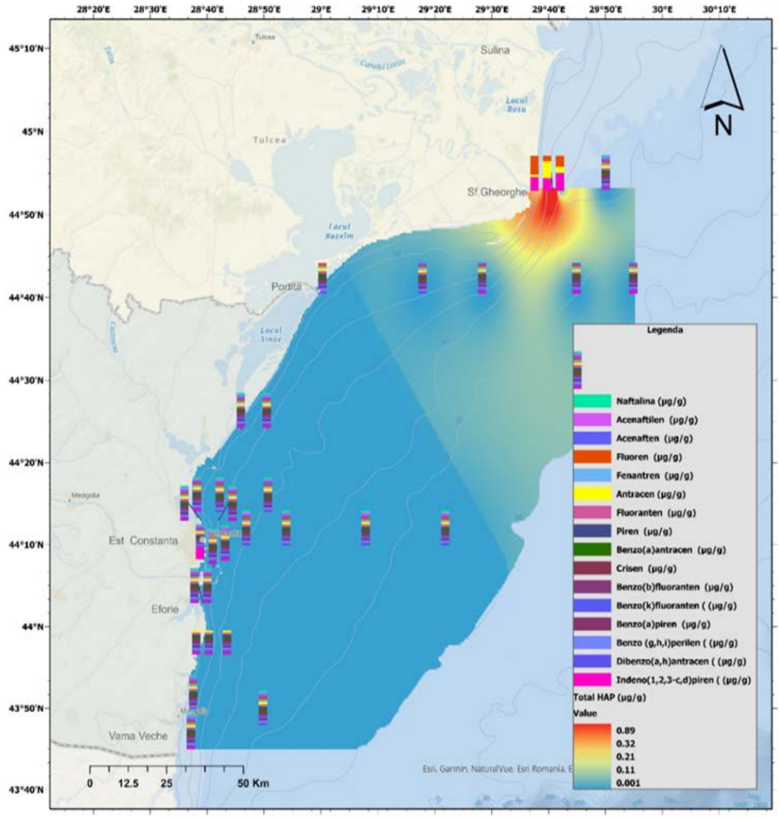

Fig. 11. Spatial distribution of polycyclic aromatic hydrocarbons (µg/g) and the percentage contribution of each compound to the total (%) in sediments on the Romanian Black Sea coast in 2023 (ArcGIS Desktop 10.7)

Polycyclic aromatic hydrocarbons (PAHs) are chemical compounds primarily produced during the combustion of organic materials containing carbon and hydrogen, such as coal, petroleum, gasoline, and wood (Montano *et al.,* 2025). While most environmental PAHs originate from anthropogenic activities, including maritime traffic and industrial processes, there are also natural sources, such as forest fires, volcanic activity, and biogenic processes associated with plants and microorganisms (Patel *et al*., 2020). In the Black Sea, PAHs are widespread in sediments, reflecting both pyrolytic sources (predominant in the Bosphorus region) and petrogenic inputs (dominant in the northeastern area near Sochi), as well as mixed pollution profiles along other coastal zones (ANEMONE Deliverable 2.3, 2021; Readman et al., 2002). PAHs occur naturally in crude oil and represent a major component of petroleum products, from which they are introduced into the environment through both intentional releases and accidental spills (Daisey *et al*., 1981; Gagosian *et al*., 1981).

In the Romanian sector of the Black Sea, marine sediments have been regularly monitored over the past two decades, and PAH concentrations frequently reach levels that may negatively affect the ecological status of the ecosystem. Particularly concerning are compounds such as fluorene, phenanthrene, anthracene, benzo(a)perylene, benzo(a)pyrene, benzo(g,h,i)perylene, and dibenzo(a,h)anthracene, which have been detected in sediments at concentrations that frequently exceed ecological safety thresholds (ANEMONE Deliverables 2.1 and 2.3, 2021; Nicolaev et al., 2019). Among these, benzo(a)pyrene is considered a key indicator of PAHs from petroleum-related inputs and is often measured at concentrations above the maximum allowable limits, primarily due to maritime traffic and offshore operations (Denga et al., 2017).

Total PAH concentrations in Black Sea sediments are comparable to those found in relatively unpolluted areas of the Mediterranean Sea but are significantly lower than those recorded in the polluted estuarine areas of the North Sea and Irish Sea, including the Mersey, Tyne, and Thames estuaries (Readman et al., 2002). The lack of correlation between total petroleum hydrocarbons (TPH) and PAH concentrations in sediments suggests that these two pollutant groups may originate from different primary sources, highlighting the complexity of PAH distribution in the marine environment.

Overall, the dominance of specific PAH compounds, such as fluorene, anthracene, and indeno[1,2,3-c,d]pyrene, in sediments from the Danube-influenced northern sector suggest mixed sources of contamination, with both pyrogenic and petrogenic origins (Davis et al., 2019). Pyrogenic PAHs are typically associated with high-temperature combustion processes, such as fossil fuel burning, vehicular emissions, and industrial activities, and are often characterized by higher molecular weight compounds like indeno[1,2,3-c,d]pyrene (Balmer et al., 2029). In contrast, petrogenic PAHs, which include lower molecular weight compounds such as fluorene and phenanthrene, originate mainly from unburned petroleum products, oil spills, and discharges from maritime traffic or port activities (Olivares-Rubio et al., 2025). The presence of both types in Romanian coastal sediments points to combined inputs from urban runoff, riverine discharge, and maritime operations. The identification of these source signatures is essential for informing targeted mitigation strategies and assessing the potential ecological impact of PAH contamination.

Total Petroleum Hydrocarbons (TPH)

In 2023, the concentrations of total petroleum hydrocarbons (TPH) in sediments along the Romanian Black Sea coast ranged between 2.44 and 389.34 µg/g. The highest values were recorded in sediments from the southern sector, particularly in the Constanța Sud port area, reflecting significant anthropogenic influence, including wastewater discharges and port activities. TPH concentrations exceeded the maximum admissible limit of 100 µg/g, as stipulated by MAPPM Order No. 756/1997 regarding environmental pollution assessment, in 3% of all samples investigated (Fig. 12). TPH concentrations remained below the legal limit in 97% of the analyzed sediment samples, supporting the conclusion that the majority of the Romanian coastal zone exhibited good sediment quality with respect to hydrocarbon pollution in 2023.

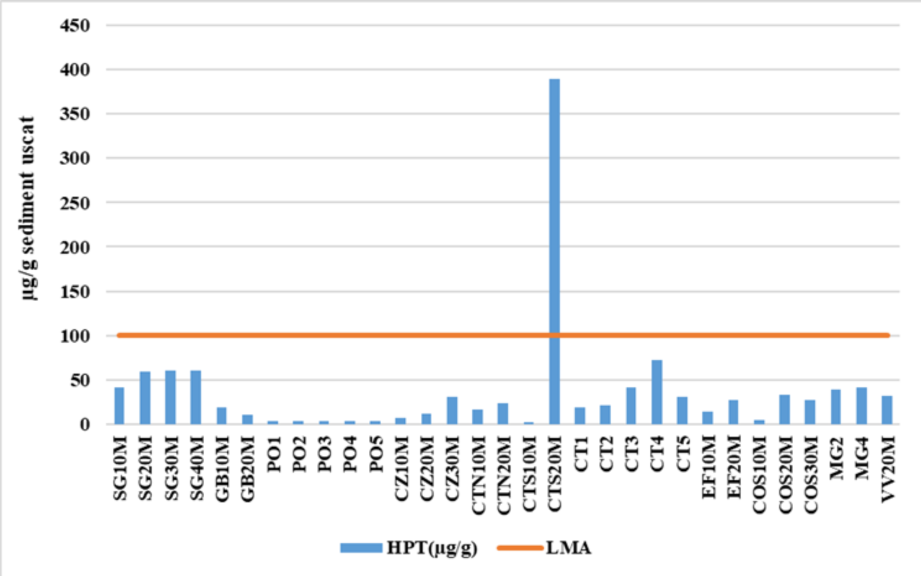

Fig. 12. Average concentrations of total petroleum hydrocarbons in sediments from the Romanian Black Sea coast in 2023, in relation to the maximum permissible limits provided by MAPPM Order no. 756/1997 for the approval of the Regulation on environmental pollution assessment

Overall, in 2023 TPH concentrations were below regulatory limits at most sampling sites, indicating generally low hydrocarbon contamination along the Romanian shelf. However, localized areas with elevated levels, particularly in Constanța Sud port area, highlight ongoing anthropogenic pressures, emphasizing the necessity of maintaining targeted monitoring and mitigation strategies.

Constanța Port remains a prominent hotspot for sediment contamination along the Romanian Black Sea coast. As one of the largest and most active commercial ports in the region, it hosts a wide range of industrial, petrochemical, and maritime activities, including cargo handling, ship maintenance, oil product storage, and wastewater discharge (Juganaru I, 2021). These operations contribute to a complex and continuous input of pollutants, like petroleum hydrocarbons, heavy metals and persistent organic pollutants. In addition to current emissions, the area is also characterized by the presence of legacy contaminated sediments that may be resuspended or remobilized through dredging, bioturbation, or hydrodynamic disturbance. Furthermore, challenges related to wastewater infrastructure, accidental spills, and stormwater runoff exacerbate the pollution burden in this sector. The persistence of high contaminant levels at Constanța Sud station highlights the need for enhanced site-specific management, improved port infrastructure, and targeted remediation strategies to reduce ecological risks and support compliance with environmental quality standards.

The pollutants investigated in this study have important implications for marine organisms and ecosystem health. HMs, even at relatively low concentrations, can bioaccumulate in marine organisms, potentially causing chronic toxicity and disrupting physiological processes, particularly in benthic communities (Wei et al., 2025). Persistent organic pollutants (POPs), such as OCPs and PCBs, pose long-term risks due to their persistence, bioaccumulative properties, and ability to interfere with reproductive, endocrine, and immune functions in marine biota (Nasri et al., 2022). Although concentrations of PAHs and TPH were generally below ecological thresholds in most sampled areas, localized hotspots identified near ports and river mouths remain concerned due to potential impacts on sensitive habitats and commercially important species. Continuous exposure, even at sublethal levels, can lead to cumulative stress, altering species composition, biodiversity, and overall ecosystem resilience (Mayer-Pinto, 2020). Thus, sustained monitoring and management strategies are critical to mitigating these risks and preserving the ecological integrity of the Romanian Black Sea ecosystem.

The contaminant levels (HMs, POPs, PAHs, TPHs) identified in the 2023 sediment samples are consistent with previously reported ranges for the Romanian Black Sea shelf, reinforcing the persistence of certain pollutants in specific areas, particularly near industrial and river-influenced zones. While the current study was not designed to assess temporal trends, the new data represents an important contribution to the existing long-term monitoring framework and will serve as a reference point for future assessments. By expanding the spatial resolution and analytical coverage of sediment contamination, this study supports the broader goals of national and regional marine strategies, including future evaluations under the Marine Strategy Framework Directive (MSFD). Comprehensive trend analyses and interannual comparisons will be addressed in subsequent studies, enabling a more detailed understanding of long-term changes in sediment quality and associated ecological risks.

Summary of Main Findings and Implications

This study provides a comprehensive assessment of hazardous substances in Romanian Black Sea sediments, highlighting several key findings. Heavy metals (HMs) generally indicated low ecological risks, with localized exceedances in port and river-influenced areas. Persistent organic pollutants (POPs), including lindane, dieldrin, and specific PCB congeners, frequently exceed thresholds, reflecting both historical contamination and ongoing inputs. Polycyclic aromatic hydrocarbons (PAHs) and total petroleum hydrocarbons (TPHs) were largely below ecological standards, although localized hotspots suggest continued pressures near anthropized sites.

The main strengths of this study lie in the integration of multiple pollutant groups, the use of harmonized methodologies consistent with international standards, and the broad spatial coverage of the Romanian shelf. Together, these aspects improve comparability with previous assessments and contribute to regional evaluations under the Marine Strategy Framework Directive (MSFD).

However, some limitations should be acknowledged. First, the assessment is based on a single survey year (2023), which restricts the ability to capture interannual variability or establish long-term trends. Second, the dataset is limited to surface sediments, without considering sediment cores that could provide information on legacy contamination and historical deposition patterns.

Third, the absence of concurrent biological or ecotoxicological data constrains the direct evaluation of pollutant effects on marine organisms. These aspects should be addressed in future research to provide a more comprehensive understanding of sediment quality and ecosystem risk.

Despite these limitations, the new dataset provides an updated benchmark for the Romanian Black Sea shelf, confirming the persistence of legacy pollutants in certain coastal sectors while also highlighting emerging contamination hotspots associated with port and riverine influences. Continued monitoring and integration of future data will be essential to track trends and strengthen the long-term assessment of sediment quality.

The implications for marine sediment pollution in the Black Sea are significant. The continued detection of hazardous substances demonstrates the need for sustained monitoring, improved wastewater and port management practices, and enhanced regional cooperation to reduce inputs. These results provide a scientific basis for adaptive management strategies, supporting the achievement and maintenance of Good Environmental Status (GES) in the Black Sea ecosystem.

CONCLUSIONS

The findings of this study are consistent with the stated objectives and research questions. Quantification of heavy metals, POPs, PAHs, and TPH established the current contamination status of surface sediments on the Romanian shelf. The spatial distribution patterns reflected the influence of major anthropogenic pressures, particularly riverine discharges, port activities, and industrial sources. Comparisons with MSFD Descriptor 8 thresholds indicated that, although Good Environmental Status is generally achieved for most compounds, exceedances of specific POPs and localized hotspots point to areas of concern. Taken together, these results strengthen the scientific basis for long-term monitoring and management of sediment quality in the Black Sea.

This research provides updated and comprehensive data on sediment contamination along the Romanian continental shelf of the Black Sea, underscoring several key findings:

- Heavy metals exhibited generally low ecological risks, with localized exceedances mainly near Constanța Port. Overall, less than 25% of samples exceeded Good Environmental Status (GES) thresholds, indicating acceptable ecological conditions.
- Persistent organic pollutants (POPs), particularly lindane (22% of samples), dieldrin (31%), and specific PCB congeners such as PCB 153 (69%), PCB 101 (59%), PCB 118 (50%), and PCB 52 (41%), frequently exceeded ecological thresholds, reflecting historical contamination and ongoing anthropogenic inputs.
- Polycyclic aromatic hydrocarbons (PAHs) were mostly below ecological threshold values, indicating generally good sediment quality; however, localized presence near river-influenced areas suggests continued inputs from specific sources.
- Total petroleum hydrocarbons (TPH) showed limited exceedances (3% of samples), particularly near port areas, highlighting localized anthropogenic pressures.

Our findings highlight critical aspects concerning the distribution and accumulation of pollutants in marine sediments from the Romanian continental shelf, enhancing understanding of their ecological impacts and informing long-term management strategies. Given the multiple pressures affecting the Black Sea, we propose the following recommendations to support pollution management and the maintenance of GES. These recommendations emphasize integrated measures and robust ecosystem-based approaches adapted to regional realities:

- Reducing pollution from diffuse sources:

Pollutants originating from diffuse sources—such as atmospheric deposition and accidental spills—pose cumulative long-term risks. Implementing progressive reduction measures is essential, including stricter regulation of atmospheric emissions, improved waste and wastewater management, and stringent control over petroleum spills and discharges.

- Continuous monitoring and periodic assessment:

Given the spatial and temporal variability of contaminants, maintaining ongoing monitoring programs is necessary. Periodic assessment of heavy metals, PAHs, and POPs will quickly identify emerging issues. Integrated national and regional monitoring is essential to achieving and maintaining GES, in line with the Marine Strategy Framework Directive (MSFD).

- Implementing ecosystem-based management:

The marine ecosystem faces multiple interconnected pressures—coastal development, climate change, overfishing, and diffuse pollution—making an integrated watershed-to-marine basin management approach crucial. Ecosystem-based strategies can help mitigate cumulative impacts and preserve ecological balance.

- Enhancing stakeholder collaboration:

Effective pollution management and conservation require close cooperation between researchers, environmental authorities, policymakers, and local communities. Data sharing and joint efforts between these stakeholders will enable informed decision-making and adaptive, region-specific strategies.

- Adapting conservation measures to climate change:

The effects of climate change, including higher temperatures, frequent extreme events, and sea-level rise—can intensify pollutant impacts on marine ecosystems. Pollution reduction strategies must remain flexible and responsive to changing climate scenarios to ensure long-term protection of marine biodiversity.

While chemical analyses provide essential information on the concentrations and spatial distribution of pollutants in marine sediments, they do not fully capture the potential biological effects on marine organisms and ecosystems.

To strengthen environmental assessments and support a more comprehensive evaluation of ecological risk, future monitoring efforts should incorporate ecotoxicological bioassays (Losso *et al.*, 2010). These tests, using representative benthic or pelagic species, can provide direct evidence of sublethal or toxic effects caused by complex pollutant mixtures, including those below chemical detection thresholds (Reichelt-Brushett, 2023).

By integrating chemical and biological lines of evidence, ecotoxicological bioassays would enhance the interpretation of sediment quality data, help identify biologically relevant hotspots, and support more effective decision-making under the Marine Strategy Framework Directive (MSFD). This dual approach is particularly valuable in heavily anthropized areas such as ports and river mouths, where chemical mixtures may act synergistically and pose unanticipated ecological risks.

These recommendations underscore the importance of an integrated approach to marine pollution management. Sustained and coordinated efforts supported by clear policy frameworks and interinstitutional collaboration will be crucial for protecting the Black Sea ecosystem and achieving ecological sustainability objectives.

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