



Combining single and complex indices of pollution with grain size trend analysis of surficial sediments in Edremit Gulf, western Turkey

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Abstract

Pollution risk assessment of heavy metals (Pb, Cr, Cu, Ni, Zn, As) in sediment samples from 68 locations in the Gulf of Edremit, western Turkey, was performed using single and complex indices of pollution assessment approaches. The pollution indices used in the sediment analyses included pollution load index, contamination factor, contamination degree for individual metals, geoaccumulation index (I_{geo}), enrichment factors, toxic risk index, modified pollution index, frequency of adverse biological effects (TEL-PEL), percent incidence of biological effects (ERL-ERM), and total risk quotients. Results indicated a low degree of contamination for the entire surveyed area except moderate contamination factor for arsenic using single indices. Enrichment factors and geoaccumulation indices indicated moderate to significant enrichment and uncontaminated to moderately contaminated both for arsenic (As) and lead (Pb), respectively. According to the results of ecological risk assessment based on TELs and PELs, the adverse biological effects of heavy metals on aquatic organisms; are for As, Cr, Ni frequently, Cu and Pb occasionally, and Zn rare. The toxic risk index values displayed low to considerably toxic risk for the entire Gulf and calculated toxic risk index results showed that arsenic accounted for most of the entire sediment toxicity. Transport patterns of sediment grain size trend analyses were correlated with the results of pollution assessment models. Transport trends are compatible with the sources of lithogenic, anthropogenic, mining activities and wastewater treatment/ geothermal originated arsenic.

Keywords Metals · Sediment contamination · Pollution index · Background levels · Sediment transport · Edremit Gulf

Abbreviations

FDEP	Florida Environmental Protection Agency	SQG	Sediment quality guideline
NOAA	National Oceanic and Atmospheric Administration	TEL	Threshold effect level
EF	Enrichment factor	PEL	Probable effect level
I_{geo}	Geoaccumulation index	ERL	Effect range low
Cf	Contamination factor	ERM	Effect range medium
PLI	Pollution Load Index	TRI	Toxic risk index
Cdeg	Degree of contamination	Q_{TEL} and Q_{PEL}	The total risk quotients of the heavy metals based on the TELs and PELs
mCdeg	Modified degree of contamination	TOC	Total organic carbon
MPI	Modified Pollution Index	CA	Cluster analysis
		PCA	Principal component analysis
		GSTA	Grain size trend analysis

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Introduction

Metal pollution in coastal areas, estuaries, and bays has recently received increasing attention since it could be unhealthy for humans and living organisms in the marine ecosystem. The degrading impact of rapid urbanization and industrialization on the quality of water resources has

emerged as a matter of concern across the globe (Khan et al. 2021). A significant number of industrial wastes containing metals have caused substantial and fast environmental degradation, especially in coastal environments. Accordingly, the ecosystem may possibly be spoiled and threaten all types of organisms including human beings. Metals accumulate in sediments and can be adsorbed via benthic organisms or/and released into the water, causing deterioration of water quality. Sediments are sensitive indicators for monitoring pollutants in aquatic environments and are typically regarded as the ultimate sink for heavy metals discharged into these environments (Zhang et al. 2014; Khan et al. 2017; Jeong et al. 2021).

Several studies regarding metal contamination and its connected environmental problems have been conducted in the marine coastal areas and rivers around the world (Feng et al. 2004; Hyun et al. 2007; Gong et al. (2008), Niencheski et al. 2014; Wu et al. 2014; Aguilar Pesantes et al. 2019). Recent studies have shown that pollutants such as organic pollutants, heavy metals, pathogens, nitrogen, and phosphorus in sediments are ubiquitous and cause health risks to human and macrobenthos (Passos et al. 2010; Tornero et al. 2014; Saleem et al. 2015). The persistency and toxicity of inorganic pollutants in the marine environment, the index-based pollution approach for categorizing of sediments become very crucial. Distribution of metals in the sediment supplies knowledge about their sources (Chen et al. 2015; Pejman et al. 2015; Sołek-Podwika et al. 2016). Historical human activities could be reflected, sediment enrichment with metals could give explanations about historical human activities (Tang et al. 2015; Mazurek et al. 2017).

The influence of the pollutants on the concentration of metals in the sediments requires the convenient evaluation as well as detection of their ecological risk (Hakanson 1980; Zhong et al. 2010; Gao and Chen 2012; Baran et al. 2018). Sediment quality guidelines and background levels are extensively utilized in ecological risk evaluations to assign metal pollution in the marine environment (Xu et al. 2015). The effective evaluation of sediment pollution with heavy metals is performed using several pollution indices such as enrichment factor (EF), geoaccumulation index (I_{geo}), contamination factor (Cf), degree of contamination (Cdeg), modified degree of contamination (mCd), modified pollution index (MPI), and toxic risk index (TRI). The indices assist to find out whether the accumulation of metals was due to anthropogenic activities or natural resources (Sutherland 2000; Caeiro et al. 2005; Elias and Gbadegesin 2011).

Although there are several studies on concentrations of heavy metals and ecological risk assessment based on concentrations of trace elements along the Turkish coasts, there has not been extensive research concerned with the metal contamination levels and ecological risk assessment in Edremit Gulf. The current study investigates distribution

of metal concentrations, to assess possible enrichment, to evaluate metal pollution of the surface sediments using several indices, to compare to the results from other studies around the world, to identify possible sources and ecological risks of metals using established sediment quality guideline in Edremit Gulf.

Material and methods

Study area

The area of study is the northernmost Gulf on the Aegean coast of Turkey and located between the Ida Mountain and Madra Mountains. Edremit Gulf, separates the Anatolian Peninsula and Lesbos Island (Fig. 1). There are many settlements such as Küçükkuyu, Altınoluk, Akçay, Burhaniye, Gömeç, and Ayvalık around the bay and tourism activities increase especially in the summer period in this area. Edremit and surrounding areas are famous for their olive cultivation and oil production as well as ore deposits in Turkey (Efe et al. 2009, 2012, 2013; Ozturk et al. 2021). Agricultural activities (especially fruit and vegetable growing) and fishery are of great importance in this area (Alevkayali and Tagil 2020; Ceyhan et al. 2006; Sağlam et al. 2014). There are several active metal mines (Yiğit 2012) and several studies involving ore deposits around the investigation area that have been conducted (Çolakoğlu 2000; Yiğit 2006, 2009; Yılmaz 2007; Oyman 2010; Akiska et al. 2013; Somay 2016).

Sampling and measurement

The sediments were taken using a Box-corer from a total of 68 sampling station (Fig. 1) during the surveys of R/V Dokuz Eylül 3 under the framework of a scientific project in the autumn of 2017 (TUBİTAK-115Y180 2018). Collected samples from Box corer were used for element, grain size and other analysis. Surface sediments (1–2 cm; 200 g) which are scooped off the undistributed top layer were used for element analysis, while 2–3 kg of sediment samples were used for the rest of analyses. The samples were transported in the ice bags and kept in a deep freeze until the drying step. Then, sediments were dried in freeze-dryer (LAB-CONCO), homogenized and were sieved through 63 μm for grain size correction. A total of 45 element concentrations were analyzed using four-acid digestion (HCl, HNO₃, HF, and HClO₄) in microwave digestion system followed by inductively coupled plasma mass spectrometry (ICP-MS) (MA200 package) (Analytical Labs ACME, Vancouver, Canada). Reference sediment sample (STD OREAS25A-4A, STD OREAS45E) was utilized as quality control of data for the analytical method. The recovery rates for the studied metals in OREAS25A-4A and OREAS45E were in the range

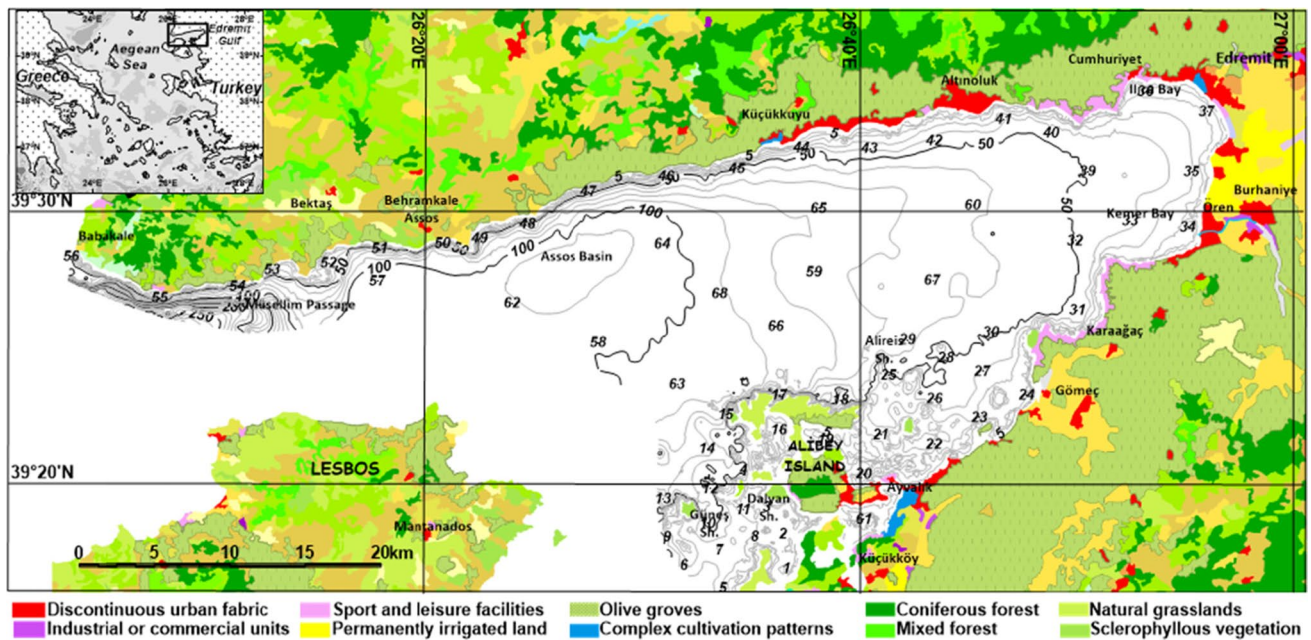


Fig. 1 Land use and bathymetric map of the Edremit Gulf with surface sediment sampling station network

of 92–110% and 88–108%, respectively. The relative standard deviation (RSD) was < 4%, and reagent blanks showed a negligible effect on element determination.

The total carbonate and organic carbon levels were determined in the sediments by adopting the standard methods (Loring and Rantala 1992). The analysis of total organic carbon (TOC) was performed by sulfochromic oxidation analytical procedure in spectrophotometry and the accuracy of this method is given as 0.25% organic substance. The total carbonate level was measured using a gasometric method, which was adopted from Scheibler gasometer system (Müller 1967). This method is based on the volumetric determination of CO₂ released by acidification of the dry sample with 10% HCl solution. The precision estimated using standard and replicate samples is $\pm 0.5\%$ for total carbonate.

The particle size analysis was used to detect gravel and sand fractions which are bigger than 63 μm . The fine grained fractions (clay and silt), which are smaller than 63 μm , were found by hydrometer procedure. The grain size classification of the sediments depends on the relative percentages of clay (< 0.002 mm), silt (0.002–0.063 mm), sand (0.063–2 mm), and coarse sand–gravel (> 2 mm). The sediment textural classes are examined using the procedure of Folk (1974).

Pollution indices

Metals can accumulate in ecosystems as a result of natural and/or anthropogenic effects. The accumulation of pollutants in marine environments occurs predominantly within the sediment in depositional areas on seabed. Many methods

are used to determine anthropogenic, environmental, and other effects. Evaluating the quality of sediment relies on a number of substantial indices suggested by investigators. Among them, such as enrichment factor suggested by Reimann and de Caritat (2000), geoaccumulation index by Muller (1969), contamination factor and degree of contamination by Hakanson (1980), modified degree of contamination by Abraham and Parker (2008), and Modified Pollution Index by Brady et al. (2015) were used in the current study to assess the risks associated with the trace metals. The calculation formulae for used indices are summarized in Table 1, and Table S1 includes the classes of pollution.

Sediment quality parameters

To determine the potential toxicity of sediments in aquatic ecosystems, sediment quality criteria developed by Florida Environmental Protection Agency (FDEP) and National Oceanic and Atmospheric Administration (NOAA), particularly the threshold effect level (TEL) and probable effect level (PEL), and the effect range low (ERL) and effect range medium (ERM) (Table 2), are used, respectively (Long et al. 1998). Three ranges of element concentrations were used to classify biological adverse effects for the TEL-PEL and ERL-ERM (David and Cosio 2021). First, the lowest range of levels, within which adverse effects are rarely detected (below TEL and ERL). Second, the possible effects range, between the TEL to PEL and ERL to ERM, within which adverse effects are occasionally monitored; and third, the probable effects range above PEL and ERM, within which

Table 1 Single and total complex pollution indices

Index	Formula	Eq	References
Enrichment factor (EF)	$EF = (C_n/C_{ref})/(B_n/B_{ref(Al)})$	(1)	Reimann and de Caritat (2000), Sutherland (2000), Gong et al. (2008), Kowalska et al. (2018)
Geoaccumulation index (I_{geo})	$I_{geo} = \log_2(C_n/1.5 \times B_n)$	(2)	Müller (1969), Loska et al. (2003), Gong et al. (2008), Ji et al. (2008), Lu and Bai (2010)
Contamination factor (Cf)	$C_f = C_m/C_{np}$	(3)	Hakanson (1980), Loska et al. (2004), Liu et al. (2005)
Pollution load index (PLI)	$PLI = \sqrt[n]{Cf_1 \times Cf_2 \times \dots \times Cf_n}$		
Degree of contamination (Cdeg)	$C_{deg} = \sum_{i=1}^n C_f^i$	(4)	Hakanson (1980), Caeiro et al. (2005)
Modified degree of contamination (mCd)	$mC_{deg} = \frac{1}{n} \times \sum_{i=1}^n C_f^i$	(5)	Abraham and Parker (2008)
Modified Pollution Index (MPI)	$MPI = \sqrt{\frac{(EF_{average})^2 + (EF_{maximum})^2}{2}}$	(6)	Brady et al. (2015)

Table 2 Summary statistics (minimum, maximum, mean, standard deviation) of sediment grain-size and measured parameters, avg shale, local background values and threshold-midrange effect of sediment quality guidelines for metals

	As	Cr	Cu	Ni	Pb	Zn	Al	Fe	Sand	Silt	Clay	C _{org}	Carb
	(mg/kg)						(%)						
Minimum	12	26	6.4	14.6	16.8	31	1.8	1.24	0.99	2	0	0.37	7.24
Maximum	63	200	51.5	136.9	63.7	123	8.17	4.86	96.9	98.72	40.16	3.44	65.19
Mean	29.2	86.2	22.7	55.2	35.9	72.3	6.15	3.21	45.2	42.7	9.95	1.67	26.9
SD	9.73	39.79	9.2	28.19	8.24	19.72	1.43	0.77	30.48	26.02	8.91	0.59	13.87
Background 57	18	105	27.5	88.7	36.4	85	7.36	3.90					
Background 58	23	103	32.7	83.6	37.3	84	7.42	4.06					
Background 60	33	117	48.9	77.8	45.2	107	7.64	4.97					
Avg. Shale*	13	90	45	68	20	95	8.1	4.72					
TEL**	7.2	52.3	18.7	15.9	30.2	124							
PEL**	41.6	160.4	108.2	42.8	112	271							
ERL***	8.2	81	34	20.9	46.7	150							
ERM***	70	370	270	51.6	218	410							

Background 57, 58, and 60: Background values from deepest sections of Gravity Core samples in Station No: 57, 58, and 60 (Table S3). *TEL*, Threshold effect level; *PEL*, Probable effect level; *ERL*, Effect range low; *ERM*, Effect range medium

*Turekian and Wedepohl (1961)

**MacDonald et al. (2000), Xu et al. (2016)

***Long et al. (1995, 1998)

adverse biological effects are frequently observed (Long et al. 1995; MacDonald et al. 2000) (Table S2).

Risk assessment and toxicity prediction

One of the two methods used to evaluate the toxic risks of sediments polluted with heavy metals is based on TEL and PELs. In this method, the total risk segments of heavy metals, Q_{TEL} and Q_{PEL} , are expressed by the following formulas.

$$Q_{TEL} = \sum_{i=1}^n \frac{C_i}{TEL} \quad (7)$$

$$Q_{PEL} = \sum_{i=1}^n \frac{C_i}{PEL} \quad (8)$$

where, C_i is the concentration (mg/kg) of each heavy metal measured in sediment and n represents the number of selected heavy metals. The TELs for As, Cr, Cu, Ni, Pb and Zn were 7.2, 52.3, 18.7, 15.9, 30.2, and 124 mg/kg and the PELs were 41.6, 160.4, 108.2, 42.8, 112, and 271 mg/kg, respectively (MacDonald et al. 2000, Table 2). If $Q_{PEL} > 1$ sediment is defined as highly “toxic”. If $Q_{PEL} \leq 1 \leq Q_{TEL}$, the risk is variable and if the Q_{TEL} is < 1 , the heavy metal content in the sediment is not harmful for benthic organisms. The sediments were classified as “non-toxic” (toxicity $< 10\%$), “uncertain” ($10\% \leq$ toxicity $\leq 30\%$), and “toxic”

(toxicity > 30%), according to the results of toxicity tests (Eqs. (7) and (8)—Zhang et al. 2017).

Toxic risk index (TRI) was used to evaluate the toxic risk based on both the TEL and PEL effects of trace metals. The TRI of a certain metal was calculated using the following equation as TRI_{*i*}:

$$TRI_i = \sqrt{\frac{\left(\frac{C_i}{C_{iTEL}}\right)^2 + \left(\frac{C_i}{C_{iPEL}}\right)^2}{2}} \quad (9)$$

where, C_{*i*} is metal concentration *i* (mgkg⁻¹) in the sediment sample, and C_{*i*TEL} and C_{*i*PEL} are the TEL and PEL of metal *i* (mgkg⁻¹), respectively. The integrated toxic risk index for various metals in one sediment sample could be calculated using Eqs. (9), (10)—(Zhang et al. 2016).

$$TRI = \sum_{i=1}^n TRI_i \quad (10)$$

TRI represents the potential acute toxicity of the heavy metals in a sediment sample can be assessed as the sum of TRI_{*i*}, and *n* represents the number of target metals. Toxic risks were categorized into five categories: (1) TRI ≤ 5, no toxic risk; (2) TRI 5–10, low toxic risk; (3) TRI 10–15, moderate toxic risk; (4) TRI 15–20, considerable toxic risk; and (5) TRI ≥ 20, very high toxic risk (Zhang et al. 2016).

Multivariate analysis

Since PCA and CA are the most extensive multivariate statistical methods used in marine investigations (Filzmoser et al. 2009; Shen et al. 2019; Jansson et al. 2022), to determine the correlation between metals in marine sediments and pollution sources, cluster analysis (CA) and principal component analysis (PCA) were conducted using the statistics software package IBM SPSS Modeler (Windows, version 25.0). PCA was used to decrease the size of data and investigate highly associated heavy metal elements into independent factors. CA sorted heavy metals into different classes, and the classes required for clustering were unknown (Bro and Smilde 2014; Dominech et al. 2022).

Grain size trend analysis (GSTA)

Sediment trend size analyses is a Fortran program as developed by Gao and Collins (1992) and was used to describe sediment transport patterns and deposition where the grain-size trends within a grid of surficial sediments are converted into a residual pattern displaying net sediment transport pathways. These vectors define the directions of sediment transport, even though their magnitudes are meaningless. Within the scope of this study, QGIS 2.8–GiSedTrend Ver. 2.5 developed program (Poizot and Mear 2010) based on the

Fortran software GSTA (Grain size trend analysis) adapted by Gao (1996) to model sediment transport, and deposition was used and sediment transport vectors were defined for the entire study area.

The grain-size parameters of surface sediments have been used to determine their transport directions over the last three decades. This method is based on producing changes in the grain size parameters (size, sorting and skewness) in the direction of transport during the sedimentation process. These trends have been properly used to establish sediment distribution patterns in the gulfs and shallow coastal areas (Gao et al. 1994; Gao 1996; Chang et al. 2001; Duman et al. 2004, 2006; Héquette et al. 2008; Poizot et al. 2008; Papa-theodorou et al. 2012; McLaren and Tear 2014; Duc et al. 2016).

Results and discussion

Sediment type, carbonate and organic carbon

Grain size composition of recent sediments in Edremit Gulf displays high sand contents as 45% (mean) followed by silt and clay with a mean value of 43% and 10%, respectively (Table 2; Table S3). Their composition is mainly silty and sandy, with varying rates of coarse and fine, thus they include the entire range from mud to silt, sandy silt, silty sand, and sand (Fig. 2a). Considering the spatial distribution of the sand-sized material, except for Ayvalık port and Gömeç coastal areas, the southern part of Edremit Gulf and the northwest coastal zone around Müsellim Passage where it is connected with the open Aegean Sea has a high concentration of over 50% (Fig. 2b). Silt contents (> 50%) prevailing in the zone between the southern shore of Edremit inner gulf and Assos basin, high values (> 75%) are observed in Assos basin and Altınoluk regions (Fig. 2c). While clay, the finest sedimentary unit with a maximum content of 35%, shows distribution in deeper areas of the Assos basin and southern offshore section of Küçükkuyu, it also has high values in coastal areas of Ayvalık, Gömeç and Ören (Fig. 2d).

Fine-grained sediments (silt–clay) show higher contents in deeper sections of Edremit Gulf with exceptions on south-eastern coastal areas between Ayvalık and Burhaniye. Usually, grain-size distribution trends were expressed by depth variations, with finer particles being found in the deeper sections (Assos basin and offshore Küçükkuyu) and coarser sediments in shallow areas (northern and southern entrance of Gulf). Coarse grained sediments are widespread on the shelf area, which is normally shallower than 50 m, at the southern and northern entrances (around Alibey Island and Musellim passage) where Edremit Gulf is connected with the open Aegean Sea. This configuration can be evaluated as a result of

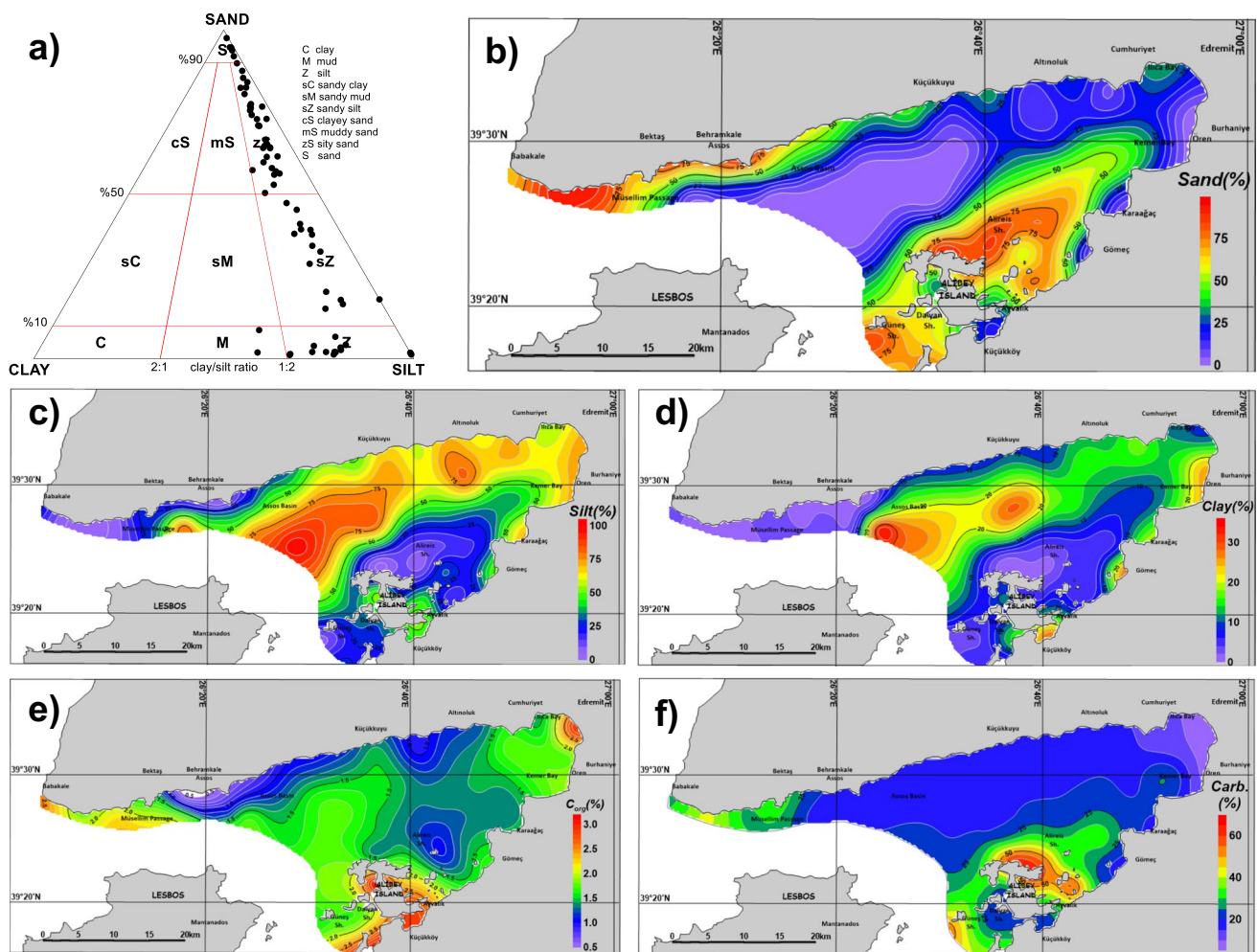


Fig. 2 Sediment classification according to Folk (1974) triangle diagram (a) and areal distribution of sand (b), silt (c), clay (d), organic carbon (e) and carbonate contents (f)

bottom water movements developed under the effect of cyclonic water circulation (Giamali et al. 2020), which is characteristic of the northern Aegean Sea.

Organic carbon (C_{org}) contents show high variability, in between 0.37% and 3.44%, and mean value of 1.67% (Table 2, Table S3); the highest organic carbon values (up to 3.0%) are detected in both coastal areas of south and north-western sections and additionally the eastern part of the inner Gulf (Fig. 2e). While the higher content of the C_{org} is a factor of limited circulation in Ayvalık Bay, other areas of local maxima are possibly related to waste-water treatment plants (WWTP) located around the Edremit Gulf. There is no available reference related with locations of higher TOC contents that is close to waste-water treatment plants (WWTP) in work area. On the other hand, Karageorgis et al. (2020) mentioned that the areal distribution patterns of C_{org} content reveal the area of local maxima, which is situated close to WWTP of the Psyttaieia Island, western Aegean Sea (2.62%) in their article. Potential effects of these locations

on C_{org} , carbonate and trace element contents (e.g. As, Cr, Cu, Ni, Pb, Zn) will be discussed in the following sections.

Total carbonate content of Edremit Gulf sediments showed high variability, with a maximum level of 65%, minimum level of 7% with mean value of 27% (Table 2, Table S3) that can be assumed to reflect the quantity of biogenic, and lesser degree terrigenous components. According to Yümün and Önce (2017), Edremit Gulf sediments contain autochthonous biogenic carbonates, such as foraminifera, bivalves, gastropod shells, and their fragments. Carbonate distribution (Fig. 2f) shows different trends in comparison to C_{org} , with maxima (> 50%) in the southern entrance of the Edremit Gulf around Alibey island and northwestern part of the Gulf. These higher carbonate contents are probably the result mainly of the higher benthic productivity and less mud sedimentation prevailing in these areas. Nevertheless, there is overall significant and negative (−0.6) correlation between the carbonate contents and fine-sized grain fraction of the sediments (Table S4, Fig. 2c,d). For example, low

calcareous (<20% CaCO₃) sediments were usually found at locations where the fine (silt, clay) portions of the samples were high.

Contents and areal distribution of trace elements

Investigated heavy metal levels of 68 surface sediments of Edremit Gulf are presented in Table 2. The overall results, which are given in Table S3 of the Supplementary information, demonstrated that the concentration (mg/kg) of trace elements in recent sediments were in the range of Al: 1.8–8.17%, Fe: 1.24–4.86%, As: 12–63 mg/kg, Cr: 26–200 mg/kg, Cu: 6.4–51.5 mg/kg, Ni: 14.6–137 mg/kg, Pb: 16.8–63.7 mg/kg, Zn: 31–123 mg/kg. The comparison between metals concentrations in the current study and the most recently published data at marine environments in the Mediterranean Sea are given in Table 3. In the limited number of studies in which results on As concentrations were given, the mean As values were lower than the values measured in this study (Martínez-Guijarro et al. 2019; Karageorgis et al. 2020; Radomirović et al. 2021). There are several reasons to explain why As levels in these studies were relatively low. Different sampling depths and sampling methods might be the main reason, which are replicated and then mixed, as well as the fact that the sediment samplings were carried out at different times over a period of approximately

5 to 20 years. Contrary to these studies, where the sampling was made in different seasons and time intervals, the total sampling period in this study is limited to a few months. The equipment used for sediment sampling generally preferred as Box corer and Grab sampler while water depths of the sampling locations, the characteristics and differences of the coastal zones are among the other reasons. In all these studies, it was stated that As pollution in the sediment was mostly of anthropogenic origin. While the average values of Cr, Cu, and Ni concentrations in this study are generally lower than many areas such as Thessaloniki Bay (Christophoridis et al. (2019), Saronikos Gulf (Karageorgis et al. 2020), and Boka Kotorska Bay (Radomirović et al. 2021); conversely, they were higher from Valencian Community (Martínez-Guijarro et al. 2019), Alexandroupolis Gulf (Karditsa et al. 2014), Northern Cyprus (Abbasi and Mirekhtiary 2020), Gulf of Tunis (Ben Amor et al. 2019), and Güllük Bay (Balkis et al. 2020). The average values of Pb and Zn are only higher than Heraklion Gulf (Koukounari et al. 2020), Valencian Community (Martínez-Guijarro et al. 2019), Northern Cyprus (Abbasi and Mirekhtiary 2020), and lower than the others listed in Table 3. Overall, the metal concentrations studied in this study showed significant fluctuations compared to some of the most recent studies on coastal areas mentioned in Table 3. When the mean concentrations of heavy metals in the study region are examined, Cr has been

Table 3 Ranges and mean or median values of trace metal concentrations (mg/kg) in Edremit Gulf and the Mediterranean Sea (na: not available)

Area (reference)	Number of samples (Mean/ Median)	As	Cr	Cu	Ni	Pb	Zn
Edremit Gulf (This study)	68 Mean	12–63 (29.2)	26–200 (86.2)	6.4–51.5 (22.7)	14.6–136.9 (55.2)	16.8–63.7 (35.9)	31–123 (72.3)
Heraklion Gulf (Koukounari et al. 2020)	55 Median	na	83.8–152 (114)	8.9–27.6 (18.9)	na	6.5–23.0 (11.3)	24.6–68.5 (45.0)
Alexandroupolis Gulf, NE Aegean (Karditsa et al. 2014)	53 Median	na	23–221 (66)	3–78 (20)	na	9–113 (38)	37–248 (93)
Valencian Community, Spain (Martínez-Guijarro et al. 2019)	288 Mean	1.8–47.1 (15.7)	2.4–37.5 (15.6)	1.1–7.9 (3.2)	1.3–18.3 (8.0)	1.7–12.6 (5.4)	6.8–66.7 (30.5)
Kavala Gulf (Stamatis et al. 2019)	60 Mean	na	23.1–185.3 (80.79)	0.78–154.0 (25.14)	0.84–50.0 (22.32)	18.12–203.3 (52.79)	48.1–1024.7 (139.78)
Thessaloniki Bay, Greece (Christophoridis et al. 2019)	25 Median	na	65.5–173.5 (121.9)	21.3–180.1 (54.4)	41.8–171.3 (79.8)	29.4–195.4 (62.4)	48.6–538.3 (144.6)
Saronikos Gulf (Karageorgis et al. 2020)	216 Mean	2–179 (19)	19–544 (152)	7–365 (52)	9–392 (77)	5–374 (69)	15–982 (169)
Northern Cyprus (Abbasi and Mirekhtiary 2020)	9 na	na	15–93 (na)	11–49 (na)	11–37 (na)	11–22 (na)	26–72 (na)
Gulf of Tunis (Ben Amor et al. 2019)	20 na	na	15–55 (na)	1.5–19 (na)	14–51 (na)	16–107 (na)	27–450 (na)
Boka Kotorska Bay (Radomirović et al. 2021)	12 Mean	11.1–90.8 (23.2)	65–1652 (295)	18.2–1296 (170)	30–389 (177)	16.5–351 (74.6)	66.6–830 (249)
Güllük Bay (Balkis et al. 2020)	23 na	na	0.1–46 (na)	1–59 (na)	na	0.1–209 (na)	10–259 (na)

the highest value with 86.2 mg/kg followed by Zn (72.3 mg/kg), Ni (55.2 mg/kg), Pb (35.9 mg/kg), As (29.2 mg/kg), Cu (22.7 mg/kg), and Al (6.15%), Fe (3.21%). Areal distribution of trace elements is illustrated in Fig. S1. Certain elements exhibit similar distribution patterns indicating their common origin, Arsenic, Cr, and Ni show distinct local maxima in the northern and east to southeastern sectors of Edremit Gulf, with decreasing trend from the same coastal sources to deep basin. Finally, As is also locally enriched around the Alibey Island and southern shore (Fig. S1). The remaining group of elements showing similarities in their distribution patterns that consists of Cu, Pb, Zn, Al, and Fe, exhibits maximum contents in deep basin and at eastern end of Gulf (Fig. S1), with E-W decreasing trend. In the eastern half of the Gulf, both northern and southern sides, the levels of metals were relatively high, showing that discharges of small rivers and streams were the possible pollution sources. Contents of some trace elements, as As in some areas have four times higher than their background levels. Highest metal (Cr, Zn, and Ni) concentrations distributed mainly eastern half of the north side of the Gulf and central part of the southern shore around Gömeç. While high As contents are mainly focused on the coastal regions of southern side, in the north they spread only at the center of Küçükköy area and decrease relatively without showing any depocenter in the offshore part of the Gulf. The distribution model of the center with high As values are predominantly in accordance with the local wastewater treatment plant discharge locations.

Since the coastal areas with open sea connections in the west of the Gulf are exposed to more sediment movement compared to the inner parts of the bay, they have mainly erosional/transport rather than depositional areas. The average concentrations of heavy metal contents of the inner coastal canyons that depositional trends are more active are in the order $Cr > Zn > Ni > Pb > As > Cu$. The only study done on the basis of limited sediment samples across the Edremit Gulf belongs to Meriç et al. (2012). According to this study, as a result of Co, Cu, Cr, Mn, Ni, Pb, Fe, and Al elements, it was concluded that the metal enrichment in Edremit Gulf originated from the metal ores located in the northwest Aegean Region.

Relationship between element concentrations and sediment grain size

Organic matter and sediment grain size are the most important parameters of sediments that affect the concentration of metals (Giordano et al. 1999; Cai et al. 2011). Fine particles absorb soluble metals in seawater and transport them to the surface sediments of the seafloor. In general, the levels of heavy metals increase with decreasing grain size (Förstner and Wittmann 1983) and the fine fraction ($< 63 \mu\text{m}$) is generally preferred in environmental research as it has the

ability to bind elements through complex adsorption and/or absorption mechanisms (Tessier et al. 1984). Among all metals examined in this study, Al ($r = 0.569$, $P < 0.001$), Cr ($r = 0.515$, $P < 0.001$), Cu ($r = 0.774$, $P < 0.001$), Ni ($r = 0.673$, $P < 0.001$), Pb ($r = 0.534$, $P < 0.001$). Zn ($r = 0.783$, $P < 0.001$) showed an extremely positive correlation with fine material consisting of silt and clay (Table S4). In contrast, the content of As ($r = 0.232$, $p < 0.10$), which correlates poorly with the fine material, is also not associated with the coarse sized material. This indicates that grain size is not the predominant factor controlling the As distribution in the surficial sediments of the Gulf and probably a different source of As from all other metals. The total organic carbon (TOC) levels in the sediments varied from 0.37 to 3.44%. Heavy metals tend to be absorbed by biological matters (Ianni et al. 2010), so the organic matter is a good metal filter (Wasserman et al. 1998; Weiping et al. 2014) and plays an important role in metal reserves in sediments. Edremit Gulf sediment TOC and heavy metal data show no such correlation with investigated metals except for Cu and Pb, which positively correlated with very low values (Table S4). This clearly indicates anthropogenic and/or lithogenic source other than biological source for total organic content.

Trace element interrelations and source identification

In Pearson's correlation coefficients (Table S4), a positive and significant correlation was observed between the fine material and lithogenic Al, Fe, and other elements except arsenic. The carbonate, (CaCO_3) which positively correlated (0.60) with the coarse-grained material, is negatively correlated (< -0.84) with all these lithogenic elements and represent biogenic carbonates (see Figs. 2b and d). All trace elements except As are distinctly related to Al and Fe, suggesting that their common origin is due to lithogenic or similar transport patterns. Various anthropogenic effects on the region were determined by evaluating the relationships between trace elements, organic carbon, carbonate, and grain size. For this purpose, by grouping variables with common geochemical behavior. Principal Component Analysis (PCA), in which the varimax rotation is applied with Kaiser Normalization, has been performed to the existing data to define the heavy metal sources in Edremit Gulf surface sediments. By extracting the eigen values and eigen vectors from the correlation matrix, the number of important factors and the percentage of variance described by each were calculated.

Table S5 demonstrates the results of varimax transformed factor values and Eigen values and communalities. The results showed that there are five Eigen values higher than 1 and these five factors cover 83% of the total variance.

The first factor accounts for 51.5% of the total variance and depends strongly on Ni, Cr, Co (>0.7) and moderately to low for Fe, V, Al, Mn, and Cu which are loaded >0.4. Factor 1, which is partly positively loaded lithogenic elements associated with fine-grained soil including silt (0.6) and clay (0.2) indicates an anthropogenic-lithogenic origin with high Cr and Ni values predominantly from mining waste. As can be seen from Fig. 3a, it points to the important input originating from the northern shores of the bay and affecting a large section of the Gulf. Factor 2 constitutes 12.9% of the total variance, with Sn, Zn, Pb, Cu scores above >0.7 and secondarily with relatively high values of silt and clay dominated by Al, Fe and V (>0.4). Considering the relevant metals and the spatial distribution model given in Fig. 3b, together with the partial lithogenic source, it indicates an anthropogenic pollution associated with the Ayvalık cantered maritime traffic. Traffic could be considered a notable source of Cu, Ni, Pb, Sn, and Zn pollution, particularly since there are urban areas around the gulf. Moreover, runoff is an important source of heavy metals in the marine surrounding, while contents of the heavy metals from transporting

emissions and ships are correlated with the density of traffic (Christophoridis et al. 2019). Factor 3 is loaded primarily by Ba, Al, and Nb >0.7, as well as moderately by Rb, V, Pb, and Fe (0.4), making up 9.5% of the total variance and representing the lithogenic factor with high Al value (Fan et al. 2022). Low (>0.4) level loadings (Al, Fe,) within the first two factors indicate the partial lithological effect within the respective group. In total, these three factors account for 73.9% of the total variance and are shown in a three-dimensional space (Fig. 3). As can be seen from Fig. 3 (top left), the first three factors were found to overlap locally in regions around Al and Fe. This combination consisting of the contributions of Al and Fe (Table S5), which are among the basic parameters of all three factors, indicates that the main factor in Edremit Gulf is predominantly lithogenic. The main element group of the first three factors includes primary heavy metals (Ni, Cr, Co, Cu, Pb and Zn) and nutrients (organic carbon), which are normally thought as the anthropogenic pollutants mostly related to the discharge of agricultural, industrial wastes, and untreated domestic sewage and also marine and land traffic activities (Qi et al. 2010;

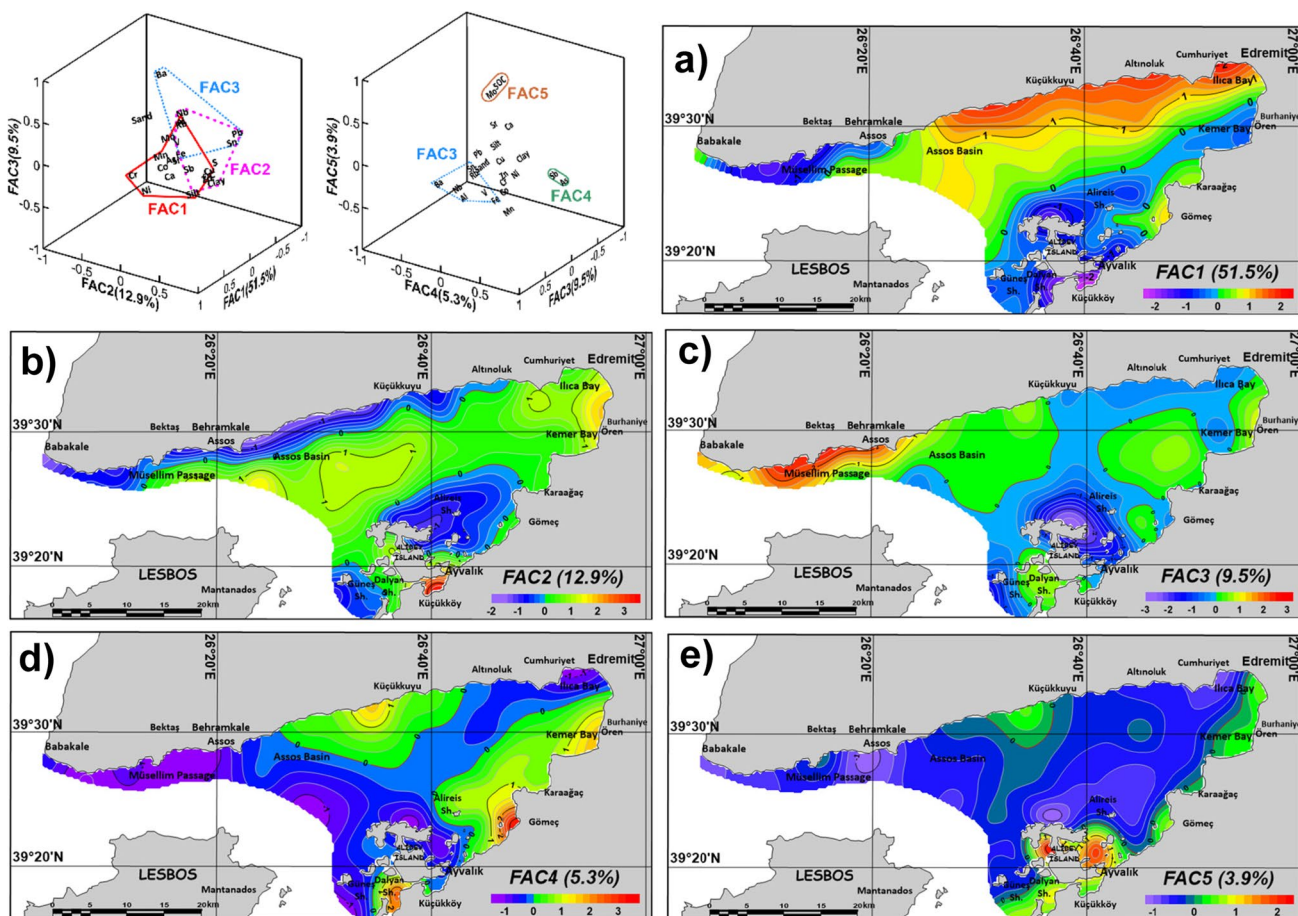


Fig. 3 Spatial distribution of the 5 Factors obtained from Principal Component Analysis; (a) F1: Mining, (b) F2: Marine Traffic, (c) F3: Terrigenous, Lithogenic, (d) F4: wwtp/geothermal, (e) F5: OM sulfurization factors

Lu et al. 2010). Anthropogenic sources of heavy metals are indicated in the worldwide marine areas in numerous former studies (Abbas et al. 2016; Birch 2017; Martínez-Guijarro et al. 2019; Karageorgis et al. 2020).

Factor 4 accounts for 5.3% of the total variance with high contribution of As and Sb and low contribution of Mn, Co, Ni, Fe, Cr, Cu, respectively. In the spatial distribution model given in Fig. 3d, high values are mainly concentrated in the locations where wastewater treatment systems and geothermal resources are located, and the relevant positive metal charges indicate the wastewater discharge — geothermal factor in this study. As with high loading (0.9), which is the highest representative of factor 4, was suggested to be associated with geothermal waters in Somay (2016). Factor 5, which accounts for the lowest 3.9% of the total variance, is strongly correlated with Mo, S, organic carbon, almost all of which have high loading values (> 0.68). This factor has been defined as the “organic matter sulfurization” factor due to the following environmental characteristics (Fig. 3e). According to Qi et al. (2010) S has negative correlations with anthropogenic and lithogenic components and positive correlations with biogenic elements, showing a marine source instead of terrigenous origin. High TOC, high Mo and low As conditions demonstrate possible circumstances of sulfurized organic material in sediments. High Mo levels point out that reactive iron was limiting factor, preventing As capture (Tribouillard 2020).

Establishing natural background levels in the study area

Local geochemical background concerns the element content in the sediments without any anthropogenic enrichment, i.e., the element content is supplied by natural sources such as biogenic and terrigenous components. After preliminary trials using three different sediment cores, it was found that there is no significant difference in the background levels between the three locations of the Edremit Gulf. To distinguish the pre-industrial deposits in the Gulf, available two ¹⁴C dated core data taken from northeastern part of Edremit Gulf were used from Yümün and Önce (2017). In the Edremit Gulf, the sediment accumulation rates were calculated at 0.57 cm y⁻¹ and 0.75 cm y⁻¹, thus element contents of deepest core levels (88–150 cm) are used as background values. Table 2 displays the pre-industrial values of metals defined as background values for each core locations.

Risk assessment and contamination

The pollution indices are useful tools to assess the sediment contamination. In this study, several reliable contamination and risk indicators utilized in previous investigations, as well as new indices were used to assess pollution degree

and potential ecological risk (Bourliva et al. 2018; Christophoridis et al. 2019). The Enrichment Factor (EF), Geoaccumulation Index (I_{geo}), Contamination Factor (Cf), Modified Pollution Index (MPI), and Toxic Risk Index (TRI) were used. Moreover, SQGs along with the Q_{TEL} and Q_{PEL} were used to estimate the seabed surficial sediment contamination in sampling region.

Enrichment Factor (EF)

To analyze if the heavy metals in Edremit Gulf surficial sediments were affected by anthropogenic activities, enrichment factors (EFs) based on the earth's crust were computed using Eq. (1) and the results are shown in Fig. 4. Calculated EF values exhibited ranges of 1.3–8.9 for As, 0.4–2.7 for Cr, 0.3–1.3 for Cu, 0.3–2.3 for Ni, 1.4–5.1 for Pb, and 0.6–1.8 for Zn. The highest EFs were calculated by As followed by Pb, while average EFs indicated decreasing order of As > Pb > Cr > Ni > Zn > Cu with respective values as 3.1, 2.5, 1.3, 1.1, 1.0, and 0.7. The EF values of all determined heavy metals (except Cu and Zn) are higher than 1.5 suggesting their anthropogenic source. According to Zhang and Liu (2002), EF values between 0.5 and 1.5 indicate that the existence of the metal was entirely from materials from the Earth's crust or natural processes, whereas EF values > 1.5 suggest the contribution of anthropogenic sources. Alternatively, the EF values of As, Pb > 5 and Cr, Ni > 2 indicated significant to moderate enrichment, respectively. The basic distribution of EF values between stations (a) and within each heavy metal (b) were shown in Fig. 4. While moderate and significant enrichments are observed on EF values of As, Pb, Cr, and Ni, all values for Cu and Zn indicate minimal enrichment. When the spatial distribution of EF-As values, which constitute the highest values among all metals studied, is examined, almost all of the southern coastal areas and the central part of the northern coast of around Küçükkuyu demonstrated higher EF values than those in the central part of the Gulf (Fig. 4). The high EF-As values observed in the northern part where EF-Pb values are minimal, were evaluated as untreated urban sewage and geothermal origin. EF increases mainly with anthropogenic activities such as agriculture, industrialization, urbanization, mining, disposal of industrial wastes (Gao and Chen 2012), and others like higher sewage runoff (Zhang and Liu 2002). EF of Cr and Ni display generally minimal values except the northern coast around Küçükköy and Edremit, which shows moderate enrichments related with mining activity (Fig. 4, bottom).

Geoaccumulation Index (I_{geo})

The I_{geo} values of the heavy metals in Edremit Gulf's surficial sediments were computed based on the crust of the earth using Eq. (2) and results are presented in Fig. 5. All analyzed

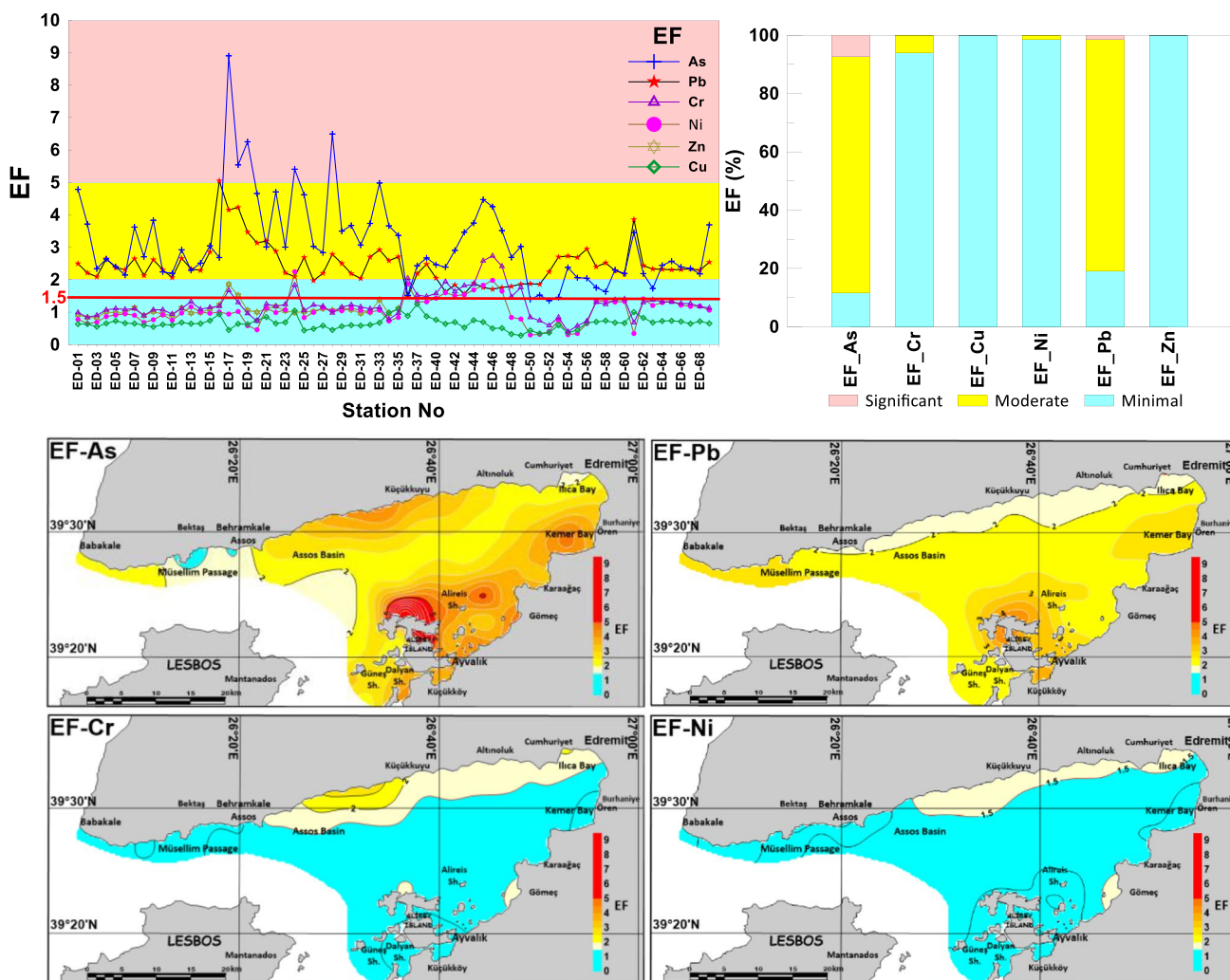


Fig. 4 Stationary (top left) and elementary (top right) distribution of EF values. Areal distribution of EF values for As, Pb, Cr and Ni are shown in below

samples displayed negative I_{geo} values of Cu and Zn and were defined as “non-polluted” regarding these two metals. Calculated I_{geo} values ranged from -0.7 to 1.7 for As, -2.4 to 0.6 for Cr, -3.4 to -0.4 for Cu, -2.8 to 0.4 for Ni, -0.8 to 1.0 for Pb, -2.2 to -0.2 for Zn. The highest I_{geo} values were calculated by As followed by Pb, while average I_{geo} values indicated decreasing order of $As > Pb > Cr > Ni > Zn > Cu$ with respective values as $0.5, 0.2, -0.8, -1.0, -1.1$ and -1.7 , which was parallel to the trend traced in the enrichment factor values. The I_{geo} values of Cu and Zn and majority of Cr and Ni in the Edremit Gulf, which suggested that the sediments are not polluted by anthropogenic origins. The general distribution of I_{geo} values between stations (a) and within each heavy metal (b) is shown in Fig. 5. While moderate contamination is observed on I_{geo} values of only As, nearly all values for Pb indicate uncontamination to moderate contamination. While a few uncontaminated to

moderately contaminated values are observed in I_{geo} values of Cr and Ni, a significant part of the remaining values consists of uncontaminated as in Cu and Zn.

When the spatial distribution of I_{geo} -As values is examined, which constitute the highest values among all metals studied, almost all of the southern coastal areas and the central part of the northern coast around Küçükuyu exhibited higher I_{geo} values than those in the central part of the gulf (Fig. 5), the same trend as EF values (Fig. 4). It has been observed that the I_{geo} values of the Pb data throughout the area and the Cr and Ni values are uncontaminated to moderately contaminated only in a few stations, especially around Küçükuyu, on the north coast. Except for the high values of Cr and Ni observed in the limited area around Küçükuyu, all of the data are uncontaminated ($I_{geo} < 0$) throughout the bay as well as Cu and Zn (Fig. 5). As shown in Fig. 5, both EF and I_{geo} showed similar results for trace element

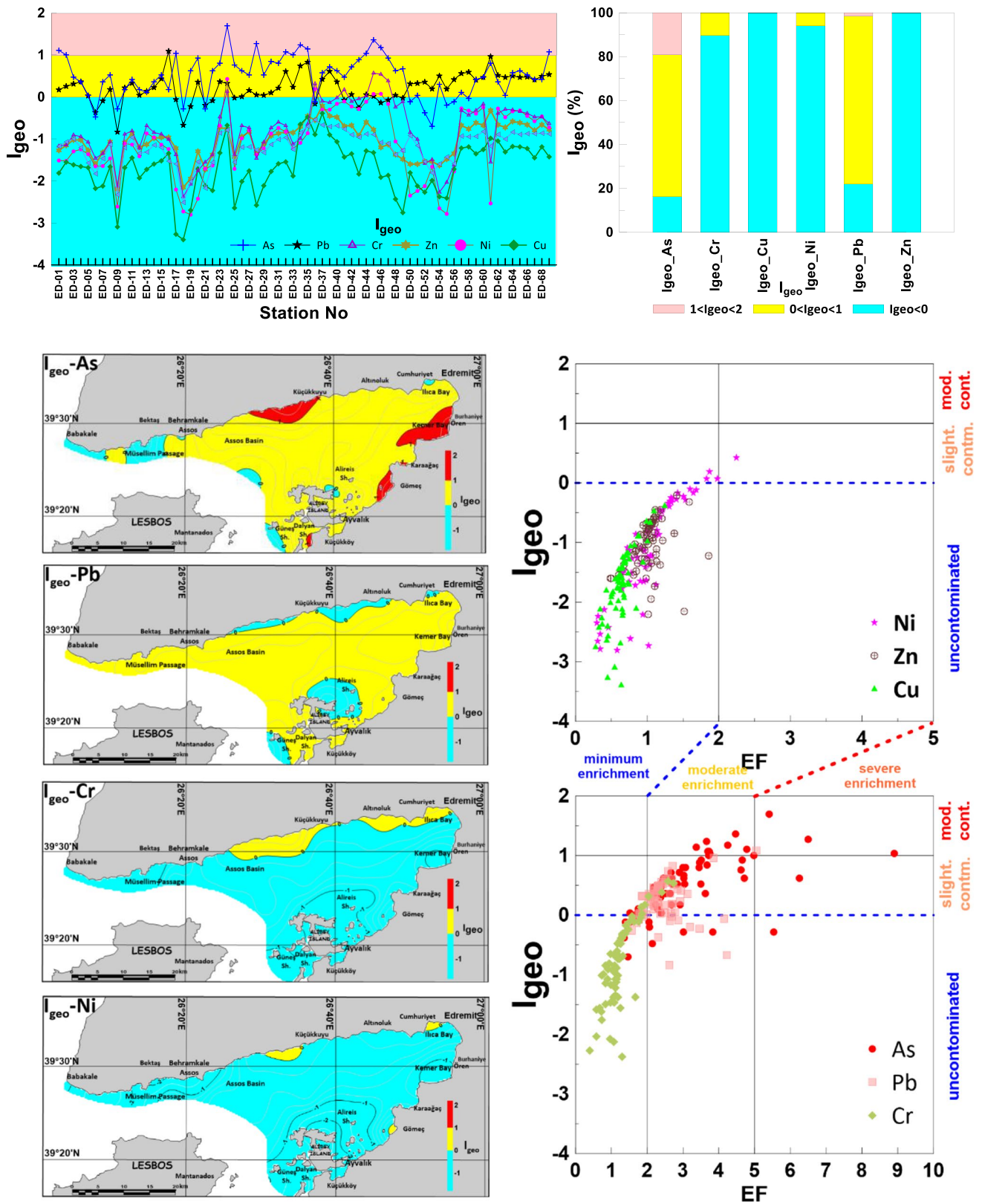


Fig. 5 Stationary (top left) and elementary (top-right) distribution of I_{geo} values. Areal distribution of I_{geo} values for As, Pb, Cr, and Ni are shown below (left). $I_{geo} < 0$ Uncontaminated; $0 < I_{geo} < 1$: Uncontami-

nated to moderately contaminated; $1 < I_{geo} < 2$: Moderately contaminated. EF versus I_{geo} in surface sediments of the Edremit Gulf (lower right)

contamination levels in the sediments of Edremit Gulf. As and Pb were dominating contamination, Cr and Ni pollution were also observed in limited coastal regions, proposing that main driving factor for the enrichment of As, Pb, Cr, and Ni was anthropogenic origin. Nevertheless, Cu and Zn were firstly originated from natural sources like those mentioned at recent works (Martínez-Guijarro et al. 2019; Wang et al. 2020). According to Liao et al. (2022) I_{geo} values appear to be independent of geological background differences, which can compare the risk of the same heavy metal in different regions, whereas EF values are suggested to be more appropriate for comparison of different heavy metals in the same sampling region.

Pollution indices

The calculated values of Cf, PLI (Eq. (3)), C_{deg} (Eq. (4)) and mCd (Eq. (5)) values for heavy metals are given in Table S6. Cf values of the trace elements were found in the order of As (1.62) > Pb (0.99) > Zn (0.86) > Cr (0.84) > Cu (0.82) > Ni (0.71). Between the calculated, Cf values only displayed the moderate contamination for Arsenic while the rest of the Cf values exhibited low contamination for Pb, Zn, Cr, Cu, and Ni. The average PLI was nearly 1 (0.99), which indicated baseline level of pollution for the entire area of investigation. The interpretation of PLI value is defined as polluted (PLI > 1); baseline level of pollution (PLI = 1) and not polluted (PLI < 1) (Liu et al. 2005; Chakravarty and Patgiri 2009). The degree of contamination (C_{deg}) and mCd values were calculated as 5.84 and 0.97, respectively. These values correspond with low degree of contamination for C_{deg} and nil to very low contamination for mCd. Cf in Eq. (3) is used for the assessment of sediment pollution (Savvides et al. 1995; Raj and Jayaprakash 2008; Rajkumar et al. 2019; Liao et al. 2022). Hakanson (1980) suggested to compare average metal value of all measured concentrations in sampling stations at work area and local reference value for measured element. In this way, there is a single Cf value for each metal which represents the whole investigated area, not for a single location. In most of the recent studies like Rajkumar et al. (2019); Liao et al. (2022), Rahman et al. (2022) Cf values of each element calculated as a separate value for each station and they produced number of Cf values instead of single value for whole area. In this work, calculated Cf, PLI, C_{deg} , and mCd values given in Table S6 consist of only a single value and describe the related parameters for the entire survey area.

Multi-elemental pollution indices are beneficial to single element ones, as for instance EF, as they combine the additional impact of multiple pollutants, which are frequently existing in industrialized and urbanized surroundings, in sediment quality evaluations. Among other indices within the study, the Modified Pollution Index (MPI) presented by

Brady et al. (2015) has also been applied. Several researchers tested MPI in varied sampling regions and deduced that it is more confident than other unidimensional and multi-dimensional indices. This idea has been lately reported by Karageorgis et al. (2020). MPI was computed by Eq. (6) and could be thought of as an improvement of the Håkanson's modified degree of contamination index (mCd) (Eq. (5)) (Håkanson 1980) in the sense that a suite of elements is combined to generate a single value, in the way that it uses the $C_{f_{max}}$ to develop a weighted average, hence considering the effect of contamination of one element, that otherwise could be decreased. Sediment samples with $10 < MPI$, $5 < MPI < 10$, $3 < MPI < 5$, $2 < MPI < 3$, $1 < MPI < 2$ and $MPI < 1$ were categorized as extremely polluted, heavily, moderately-heavily, moderately, slightly and unpolluted, respectively (Brady et al. 2015). According to Fig. S2, indicating the spatial distribution pattern of MPI, the Edremit Gulf, is entirely polluted starting from slightly to heavily. In the southwestern corner of the area, the northern side of the Alibey island heavily polluted sediments were observed close to old Pb mining area (MPI: 6.6). Similarly to the spatial distribution of the EF-As and EF-Pb (Fig. 4), the MPI is decreasing to moderately and slightly polluted ranges toward the east and north, respectively. In the northern part of the Edremit Gulf moderately heavily polluted sediments are found close to Küçükuyu WWTP outfall and the same as Gömeç and Küçükuyu outfalls in the southern coast. Similarly, to the spatial distribution of the As, Cr, and partly Ni (Fig. S1), the MPI is reducing to moderately and slightly polluted ranges in parallel to coastline (Fig. S2). This pattern is in line with the abnormalities observed in the foraminifer shells and the yellow and/or black-colored shells of both the foraminifers and ostracods were the results of natural and anthropogenic pollution around Küçükuyu and Kemer Bay (Yümün and Önce 2017). The disruption of sediment quality is conformable with the cyclonic hydrological regime forcing the distribution of treated wastewater of the outfall systems both southern and northern side of the Gulf, similar to the work of Karageorgis et al. (2020) at Saronikos Gulf.

Sediment quality parameters

The limit values determined in the sediment quality guidelines (SQG) and the results of the metal analysis performed in the samples were given in Table 2. As seen in Fig. 6 showing the entire data, our study showed that all measured Zn concentrations in sediment samples were below the corresponding TEL value indicating rare (9%) toxic effect that is not associated with adverse biological effects. Mean levels of As, Cr, Cu, and Pb were between corresponding TELs and PELs in which only the mean value of Ni exceeds PEL. As, Pb, Cr, Cu, and Ni concentrations were in the range between the corresponding TELs and PELs by 90%, 78%,

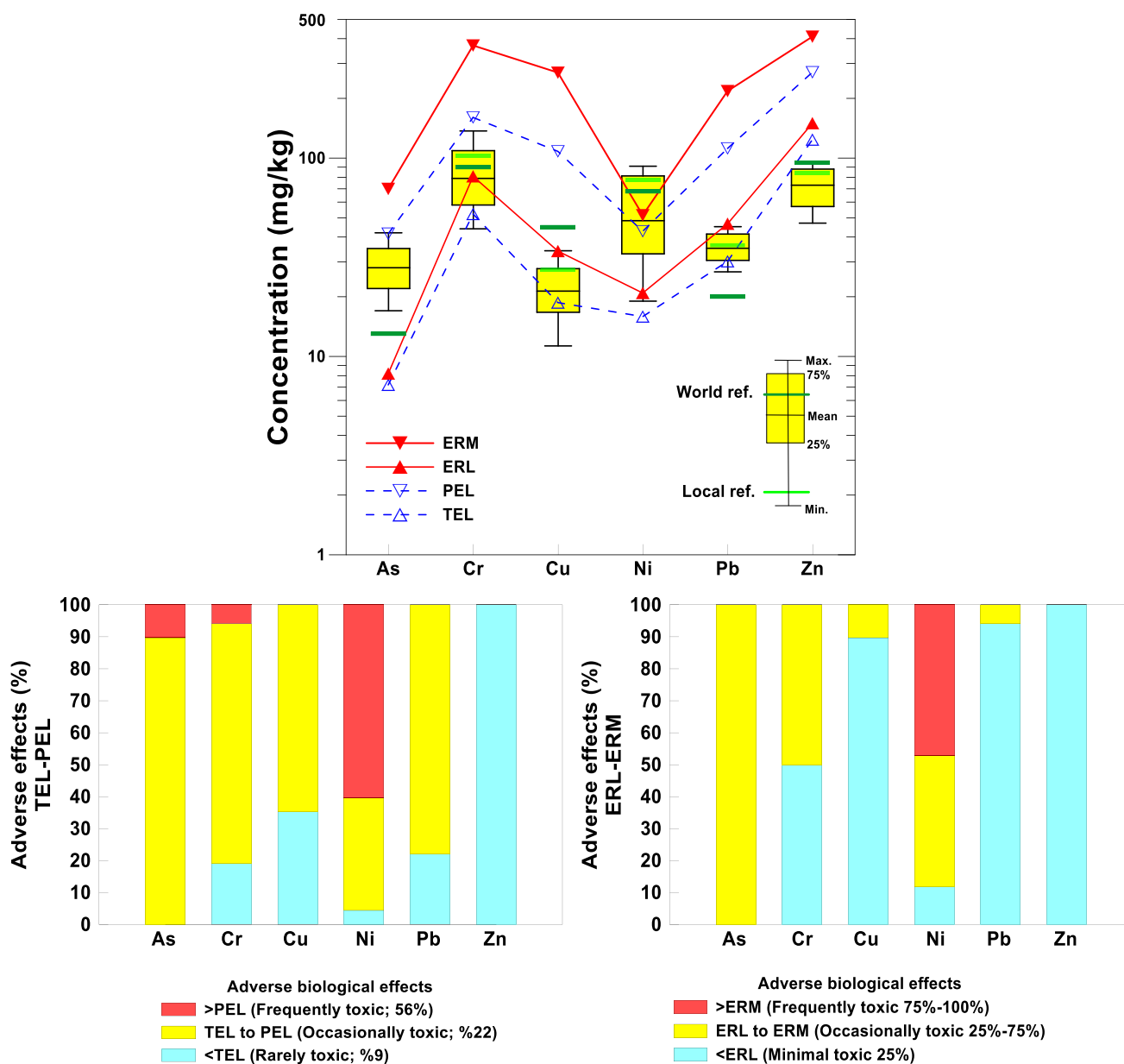


Fig. 6 Box and whisker plots for heavy metal concentrations in the surface sediments and SQGs (top), percentage of samples each guideline TEL-PEL (lower left) and ERL-ERM (lower right)

75%, 65%, and 35%, respectively, which indicate occasionally toxic (22%) adverse biological effects. Ni, As, and Cr were the limited metals with concentrations exceeding the PELs by 60%, 10%, and 6%, respectively, and this indicates that Ni has the highest adverse biological effect as frequently toxic (56%). Therefore, adverse biological effects may be formed more frequently due to the concentrations of Ni, As, and Cr. According to the NOAA sediment quality criteria (ERL and ERM), the levels of Zn for all sediments were less than the corresponding ERL which indicates minimal toxic (25%) effect. The concentrations for Pb, Cu, Cr, and Ni are lower than the corresponding ERLs by 94%, 90%, 50%, and

12%, respectively. As, Cr, Ni, Cu, and Pb contents are in the range of the corresponding ERLs and ERMs by 100%, 50%, 41%, 10%, and 6%, respectively, indicating occasional adverse effects. Ni was the only metal with levels exceeding the ERM (by 47%) which indicates frequently toxic (75% to 100%) effects (Fig. 6).

Sediment quality guidelines (SQGs) indicated that most of the identified heavy metals (As, Cr, Cu, Ni, and Pb) in Edremit Gulf ranged between TEL and PEL values, except for average the Ni in at the northern part of the Gulf and around Gömeç in southern coast, which was more than ERM value. Heavy metals contamination was associated with the

evacuation of wastewater from WWTP and fertilizers, along with shipping activities. Khaled et al. (2006) deduce that average concentrations of the investigated heavy metals (Cd, Cr, Cu, Ni, Pb, Zn) in the sediment studied were generally higher than the ERL but lower than the ERM values for most locations, and anthropogenic origin was the proposed major reason for pollution. While Albuquerque et al. (2019) concluded that the concentration of Pb exceeds the TEL reference value could be related to sugarcane cultivation, because the application of fertilizers and pesticides contributes to the increase of toxic metals in the soil and sediments, such as Cr, Zn and Pb; on the other hand De Lima et al. (2022) found that Cd and Cr concentrations between TEL and PEL in their studied areas and proposed that regardless of anthropogenic influence, suggesting that the region has high natural levels of these metals.

Toxicity estimation and risk assessment

To classify the sediments toxicities, several principles have been defined. Mearns et al. (1986) categorized the data of amphipod survival tests nontoxic (average survival: 96–96.5%), marginally toxic (average survival: 76.5–83%), clearly toxic (average survival < 76%), and highly toxic (average survival < 20%). Later Swartz et al. (1995) categorized the results of amphipod survival tests as nontoxic (< 13% mortality), uncertain (13–24% mortality), or toxic (> 24% mortality) and according to Hansen et al. (1996); sediments were categorized as toxic if mortality was higher than 24%, while sediments with $\leq 24\%$ mortality were thought as nontoxic. Like Zhang et al. (2017), this research regarding the uncertainties in the toxicity estimation results of different organisms and end points, a wider toxicity range of 10–30% was characterized as “uncertain”, < 10% was defined as “nontoxic”, while > 30% was “toxic”.

If more than 75% of sediment samples in the marine environment are correctly estimated to be toxic or nontoxic, sediment quality guidelines (SQGs) are considered to supply a reliable basis for evaluating sediment quality (Long et al. 1998; MacDonald et al. 2000). Based on this, the concentrations of 6 trace elements (As, Cr, Cu, Ni, Pb and Zn) determined in the surface sediment samples of the Edremit Gulf were compared with the TEL and PEL values in the previous sections (Table 2, Fig. 6). When sediment quality guidelines (SQGs) are evaluated by considering the number of samples with $Q_{PEL} > 1$, $Q_{PEL} \leq 1 \leq Q_{TEL}$ and $Q_{TEL} < 1$ calculated on the basis of the given TEL and PEL, all Q_{TEL} and Q_{PEL} values vary between 4.9–24.4 and 1.2–6.7, respectively (Fig. S3, top). Based on these data, virtually all of the sampling locations are higher than 1 both for Q_{TEL} and Q_{PEL} and the whole area shows toxic (30%) characteristics as indicated in Fig. S3.

Figure S3 demonstrates the contribution of each trace element to the total toxicological risks, i.e., the ratio of $Q_{i,PEL}/Q_{PEL}$. In the 68 sediment samples, the $Q_{i,PEL}$ of As, Cr, Cu, Ni, Pb, and Zn accounted for 8.45–41.84%, 9.73–25.57%, 2.76–11.38%, 14.82–52.86%, 4.48–21.04, and 4.22–15.17% of Q_{PEL} , respectively. The results proposed that Ni made a dominant contribution to the toxicities of trace element mixtures in the sediments, which was followed by As, Cr, Pb, Zn, and Cu. These results showed that Ni and As were the major metal contaminants in the sediments of Edremit Gulf. In contrast, the risks caused by Cu and Zn were relatively low.

MacDonald (1994), proposed SQGs where the adverse effects of TEL is considered reliable if the minimal effect range is less than 10%, while if the adverse effects exceed 65% of the probable effect range, the PEL is considered consistent. Thus, the TEL of trace metals in sediments would display only limited acute toxic impacts to exposed aquatic organisms. However, if the exposure period is long enough, the possibility of chronic toxic impacts cannot be refused. Hence, a TRI integrating the TEL and PEL was used to evaluate toxic risks in terms of both chronic and acute toxic impacts in aquatic organisms exposed to sediment polluted by trace elements. Overall TRI results, displayed moderate toxic risk ($10 < TRI \leq 15$) at 23 locations, while no toxic risk ($TRI \leq 5$) and considerable toxic risk ($15 < TRI \leq 20$) were observed at three (Stn; 9, 18, 55) and two (Stn; 24, 45) stations, respectively (Fig. 7). The mean TRI_i values of metals showed a decreasing order of $TRI_{As} (2.91) > TRI_{Ni} (2.62) > TRI_{Cr} (1.23) > TRI_{Pb} (0.87) > TRI_{Cu} (0.86) > TRI_{Zn} (0.45)$ with mean contributions of 33.5%, 27.8%, 13.3%, 10.5%, 9.8%, 5.2%, respectively. Calculated TRI results indicate that As accounts for most of the overall sediment toxicity. Compared to the others, the most prominent difference of As and Ni, which provides the most important contribution to TRI, is mainly their relatively low TEL values and their high concentrations measured in surface sediments. This points out the potential toxicity of sediments in Edremit Gulf and stands out As and Ni as two heavy metals deserving more concern. In recent studies related with TRI, Rahman et al. (2022) found highest TRI values of Cr and suggested that it could have happened due to the reasons that Cr concentration in these sampling points was high due to direct dumping leather waste. Lu et al. (2020) suggested that As and Cd contributed higher at TRI compared to other toxicity assessments, and Cd, Cu, Zn contents were mainly affected by anthropogenic activities, while As and Pb were generally controlled by natural and agricultural resources.

Sediment transport patterns

Using the GSTA model, the sediment transport vectors were generated. The transport pattern that was developed

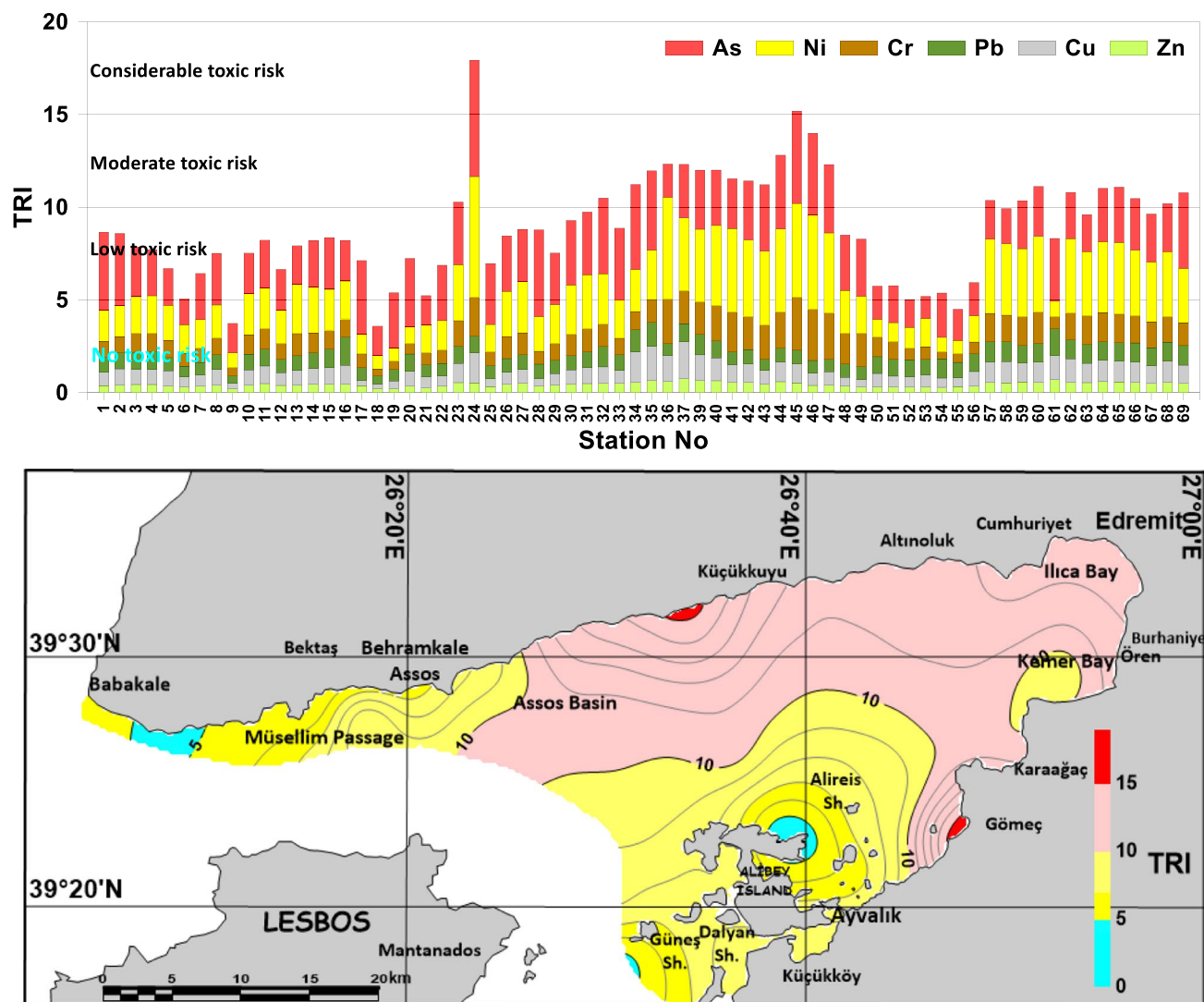


Fig. 7 Stationary (top) and Areal distribution (below) of the Toxic Risk Index (TRI)

is identified and displayed in Fig. 8, which shows the grain size trend and transport directions, denoted by arrows, in the Edremit Gulf. The vectors represented the net transport directions of surface sediments, and the vector length indicates the importance of grain size trend, rather than the scale of transport rate.

The circulation of the coastal marine reflects the directional flow features on the dispersion and regional tendency of the sediments. The primary driving forces for sediment transport in shallow nearshore are wind-induced waves and associated movements. The conclusion of the ADCP measurements in Edremit Gulf showed that currents were primarily driven by winds and the associated fluctuation of sea surface layers. The ADCP data recorded for the shallow (< 50 m) zone demonstrated that the surface currents are generally slightly faster than the bottom currents that form in the opposite direction (TUBITAK 115Y180 2018). The

surface waters that moved toward the east at the southern zone and in the opposite direction on the northern coastal areas displayed complex and small-scale cycles that occur in the southern part of the Gulf because of the coastal structure. Small scale cyclonic cycles in the comparatively shallow eastern part and an anti-cyclonic cycle in the coastal zone around the eastern section of the islands were observed. The general circulation of this region is not only wind-driven but also the general thermohaline circulation of the Aegean Sea, particularly during the summer months, contribute significantly to the deep-water circulation system of Edremit Gulf.

In this study, two distinct sediment transport modes consisting of strong basinward transport sediments consisting of mainly river-born and westerly directed weak transport of basin sediments were recognized in the Edremit Gulf. As tracers for sediment movement elemental, carbonate, carbon content and contamination assessment data were evaluated.

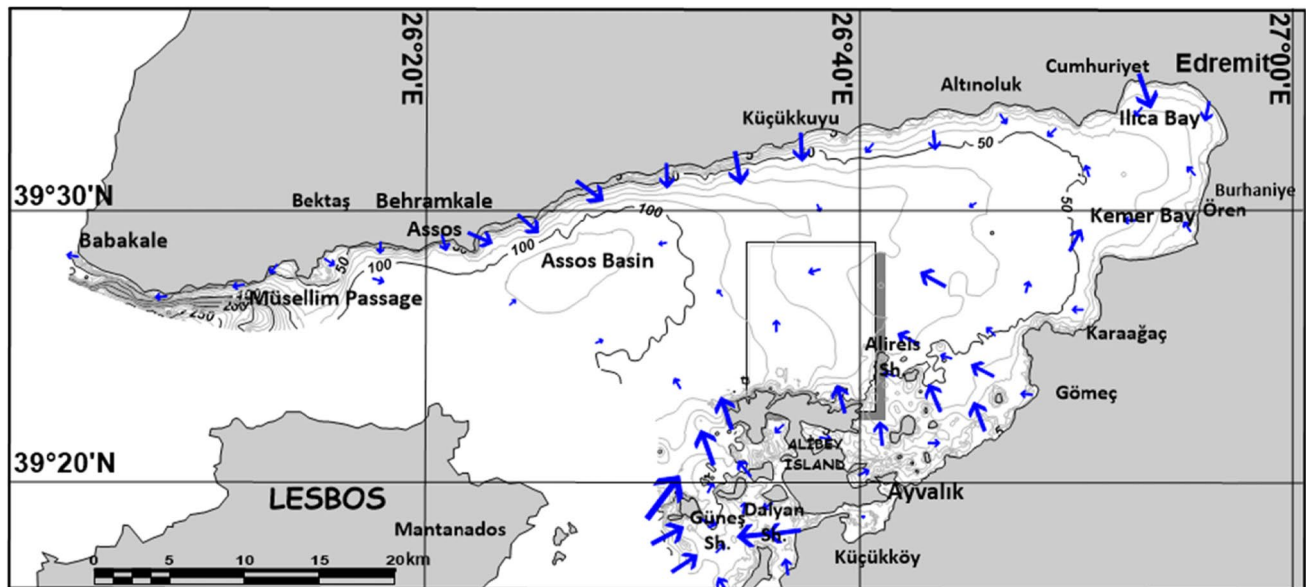


Fig. 8 Map showing net sediment transport pattern derived from GSTA model

Within this system, utilizing tracing and hydrodynamic information of the pattern of sediment transport was also evaluated. Even though the main controls of sediment transport are definitely hydrodynamic, the depositional zones are strongly influenced by the topography where the geomorphological characteristics trapping the sediments. Spatial distribution of the trace elements, PCA components, degree of sediment pollution and several other indices are consistent with the determined residual grain-size trend pattern of Edremit Gulf transport vectors (Fig. 8). This contribution portrays on pollution problems and can provide in-depth understandings of the contaminant transport processes, thus supporting the practices of pollution control and prevention in the Edremit Gulf.

Conclusion

In this present study, we investigated geochemical distribution, controlling factors, possible sources, and pollution indexes of six toxic elements (i.e., As, Cr, Cu, Ni, Pb, and Zn) in the underexplored Edremit Gulf, Eastern Aegean Sea.

- The mean concentrations of six toxic elements showed the descending order of $Cr > Zn > Ni > Pb > As > Cu$ and only As was higher than their corresponding background values.
- Using the grain size trend model, it was detected that the transportation of fine sediment was related to the residual currents, which was consistent with the trace element distribution and calculated pollution patterns.

- The results illustrated that heavy metals exhibit two spatial distribution patterns (As, Cr, Ni and Cu, Pb, Zn). All these elements were also positively and significantly correlated with the fine fraction of the sediment.
- The multivariate statistical methods with element concentrations and correlation analysis indicated that the study area is predominantly under the pressure of anthropogenic source. The enrichments of As and Pb are remarkable in the sediment, with the consistently high EF, I_{geo} and Cf values.
- Ecological risk assessment based on TELs and PELs showed that the contaminations of Ni and As were at relatively high levels and toxicological risk estimations Q_{TEL} and Q_{PEL} indicated that Ni and As were the major trace element contaminants in the sediments of Edremit Gulf. Furthermore, the influencing mechanisms of the changing sediment conditions on transformation of trace elements in the coastal areas of the Edremit Gulf need to be investigated in the future.

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Author contribution Muhammet Duman conceived and designed the study. Muhammet Duman, Atilla Hüsnü Eronat and Ezgi Talas completed marine survey and collect sediment samples. Ezgi Talas, Filiz Kucuksezgin and Tarık İlhan conducted the lab experiment. Muhammet Duman, Filiz Kucuksezgin, Atilla Hüsnü Eronat and Şenol Aydın drafted the initial manuscript.

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Data availability The data sets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Declarations

Ethics approval and consent to participate Not applicable.

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