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MAPPING OF SELECTED TRACE METALS AND ASSOCIATED RISK IN COASTAL SEDIMENTS ALONG THE NORTHWEST ANATOLIA COASTS OF TURKEY

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Abstract

This study investigates the distribution of selected trace metals (As, Cd, Cr, Cu, Ni, Pb, and Zn) and associated risk in coastal sediment samples taken from the Northwest Anatolia coastline of Turkey. The 451 km long coastal area contains many industrial areas and tourist sites. Trace metal concentrations in sediment samples collected from 100 stations were determined by inductively coupled plasma mass spectrometry (ICP-MS). The ecological risk was evaluated according to the Sediment Quality Guidelines (SQG) of the United States Environmental Protection Agency (EPA) and by calculating and examining pollution indices including the degree of contamination (*Cd*), modified degree of contamination (*mCd*), potential ecological risk index (*Ri*), and enrichment factors (*EF*). Trace metal concentrations were visualized and spatially described utilizing bubble maps. Based on the risk analysis, arsenic was found to be the most dominant pollutant in the study area and the other metals did not indicate a serious anthropogenic pollution.

Key words: coastal sediments, contamination factor, enrichment factor, sediment quality guidelines, trace metals

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1. Introduction

An important problem facing the developing world is increasing environmental pollution. Industrialization, population growth, and poorly planned urban wastewater disposal cause increasing aquatic ecosystem pollution (Gavrilescu, 2022; Topçuoğlu, 2005). Outdated discharge systems such as those installed in oil refineries, thermal power plants, and chemical manufacturing plants introduce dyes, disinfectants, drugs, and other industrial by-products into the ecosystem.

Environmental interactions, which are complex and intense activities in which physical, chemical, biological, social, cultural, economic, and ecological processes interact, often cause contaminants to spread amongst the air, water, and soil. Therefore, uncontrolled environmental pollution affects life not only in the area where it originates but also in places many kilometres away.

Contaminants are transported via precipitation and erosion towards the sea and further introduced into coastal areas by wind, waves and sea currents (Karageorgis et al., 2002). Sediments demonstrate a great capacity to accumulate trace metals and organic pollutants even from low concentrations (Chen et al., 2019; Christophoridis et al., 2009; Deng et al., 2020; Janaliyeva et al., 2020; Maher and Aislabie, 1992; Slukovskii et al., 2020; Tam and Wong, 2000). Metals accumulate in the sediment in the form of organic and inorganic chemical compounds with various complex structures and can remain there for many years. Once saturated with metal content, the sediment layer then tends to release ions back into the water and becomes a potential source of pollutants for the aquatic system (Gavrilescu, 2021; Tokatlı et al., 2017). In this manner, coastal sediments act as environmental repositories for pollutants. Therefore, the monitoring of coastal sediments for trace metal contamination is of great importance for assessing ecological risk since the sediment includes pollutants from both terrestrial and aquatic environments. However, the degree of contamination in coastal areas also depends on if the area is a closed inner sea, an open sea or flowing water.

Pollution levels in coastal areas and inner seas are higher than in open seas (Karageorgis et al., 2002; Luo et al., 2010; Pekey et al., 2004; Zhang et al., 2007). So, trace metal content in coastal sediments can be delicate indicators for environmental risk in the Marmara Sea which is one of the rarely closed seas in the world, in the Dardanelles Strait which is flowing water combining the Marmara and the Aegean Sea to each other, and in the Aegean Sea which is a semi-closed sea.

Due to potential increased environmental risk from large tourist and industrial development, a systematic ecological survey was conducted in the coastal sediments from Northwest Anatolia including the Marmara Sea, Dardanelles Strait, Aegean Sea and also Bozcaada Island. This study is the first systematic and comprehensive monitoring survey in the region as part of the research project on ecological risk assessment of Northwest Anatolia coastal sediments. This study aims to (1) determine the concentration levels of Arsenic (As), Cadmium (Cd), Chromium (Cr), Copper (Cu), Nickel (Ni), Lead (Pb) and Zinc (Zn) in coastal sediment samples, (2) map the spatial distributions of the selected trace metals using Geographic Information Systems, and (3) evaluate the results according to the sediment quality parameters.

2. Materials and methods

2.1. Study area and sediment sampling

This study collected sediment samples from the coastal areas from Bandırma to Assos, and Bozcaada Island depicted in Fig. 1. Bandırma and Erdek Bays constitute the two sides of the Kapıdağ Peninsula in the Marmara Sea which is one of the rare inland seas of the world. There are large industrial enterprises and a port located in Bandırma Bay.Due to a favorable climate and many sandy beaches, the population of Erdek Bay increase significantly and swells in the summer months due to the tourist boom. The 61 km long Dardanelles Strait connects the Marmara Sea to the Aegean Sea.

Çanakkale is one of Turkey's tourist and cultural centers with an ample amount of beaches, thermal springs and historic sites such as Troia and Assos (Canbaz Öztürk et al., 2013). There are rivers, public beaches, and industrial establishments along the Çanakkale coastline from Güzelyalı to Assos. Bozcaada Island constitutes the Bozcaada district of Çanakkale province and is the third-largest island of Turkey.

The coastal sediment samples were collected during 2017 from 100 georeferenced locations systematically at 3 kilometer intervals, topography permitting, along the coast of Northwest Anatolia (~ 413 km) and coast of Bozcaada Island (~ 38 km) including public beaches.

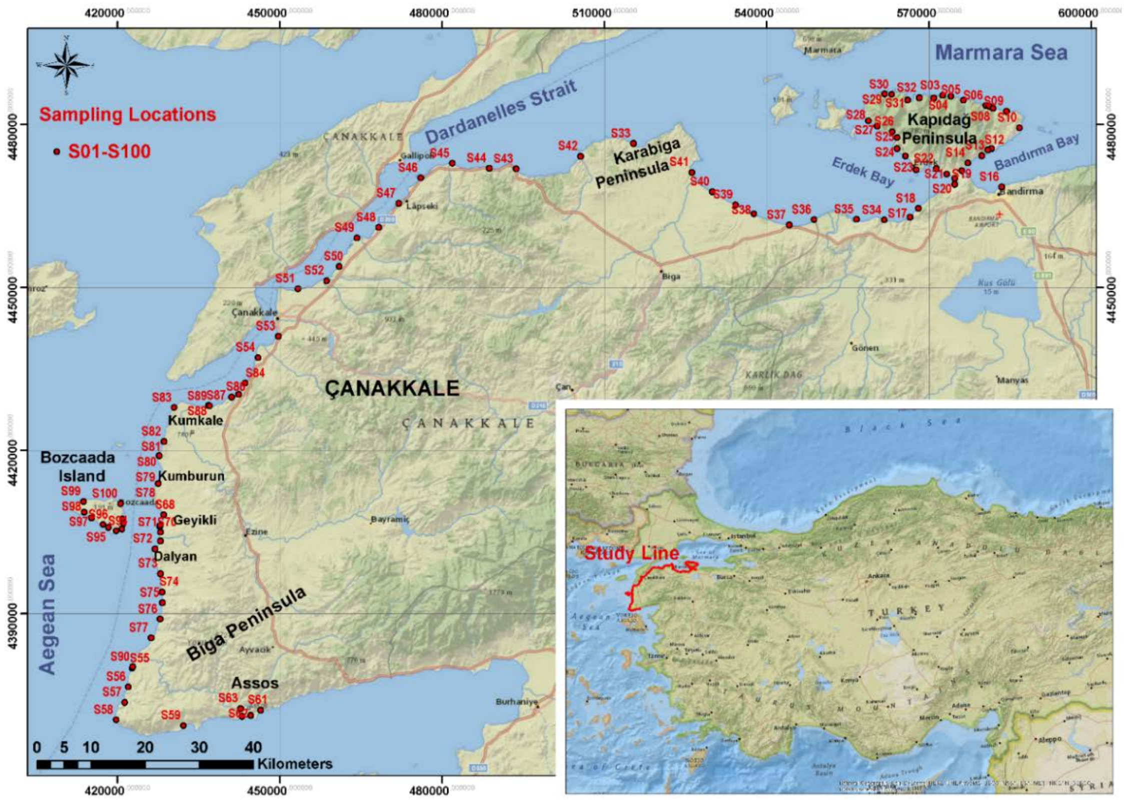


Fig. 1. Sampling locations of coastal sediments from Northwest Anatolia

Sediment samples were taken from a depth of up to 10 cm with a plastic shovel close to the edge of the water and then sealed in polyethylene plastic bags (IAEA, 2009; Luo et al., 2010; Yalcin and Ilhan, 2008; Yalcin et al., 2016; Wang et al., 2019). In each sampling region, the surface sample was obtained from five sub-samples at the corners and center of a square 5m wide. The sub-samples were homogenized in situ.

2.2. Sample preparation and trace element measurements

All coastal sediment samples were air-dried in a controlled clean laboratory and then oven-dried at 60°C until they reached a constant weight. The dried coastal sediment samples were ground with a tungsten ring time-controlled grinder of Retsch (RS 100) and 5 g samples were prepared. Trace element analyses in coastal sediment samples were carried out following aqua regia digestion using inductively coupled plasma mass spectrometry (ICP-MS) in ACME Analytical Laboratories (ISO9001:2018 quality standards), Ltd. In Canada. Sediment samples were digested with a modified aqua regia solution of equal parts concentrated ACS grade HCl and HNO_3 and demineralized H_2O in a hot-water bath (~95°C) for 1 hour. After cooling each sample was made up to a final volume with 5% HCl.

Quality assurance and quality control were assessed using duplicate analyses, procedural blank, and standard reference materials (STD DS11 and STD OREAS45EA). Elemental concentrations of standard reference materials were determined with triplicate ICP-MS analysis and measured results were compared with certified values in Table 1. Recoveries were ranged from 99.9% (Cr and Cu) to 106.9% (Pb). The minimum detection levels were 0.1 ppm for As, Ni and Zn; 0.01 ppm for Cd, Cu and Pb; and 0.5 ppm for Cr.

2.3. Ecological risk assessment methods

2.3.1. Sediment quality guidelines by USEPA

Sediments are classified into three classes as non-polluted, moderately polluted, and heavily polluted on the basis of the Sediment Quality Guidelines (SQGs) proposed by USEPA (Giesy and Hoke 1990). These classification values for the metals studied were given in Table 3. Ecological risk assessment according to SQGs was performed by comparing the mean or median values of trace metal concentration in sediment samples with SQGs.

2.3.2. The contamination factor, contamination degree, the modified degree of contamination and the potential ecological risk index

The contamination factor is an assessment method suggested by Hakanson (1980) used to estimate the level of pollution caused by the metal in the environment and is calculated according to the (Eq. 1) (Hakanson, 1980):

$$C_f^i = \frac{C_e^i}{C_b^i} \quad (1)$$

where C_f^i , C_e^i and C_b are the contamination factor, concentration and background value, respectively, of element i in a sediment sample.

This work uses crustal average values given by Taylor as background values and are listed in Table 2 (Taylor, 1964). Hakanson classified contamination factors in to four groups: C_f^i<1, $1 \leq C_f^i < 3$, $3 \leq C_f^i < 6$ and $C_f^i \geq 6$ denote low, moderate, considerable, and very high contamination factors, respectively (Hakanson, 1980).

Contamination degree (*C_d*) is defined as the sum of all contamination factors (C_f^i) for an area and calculated with (Eq. 2).

$$C_d = \sum_{i=1}^{7} C_f^i \quad (2)$$

The intervals C_d<7, $7 \leq C_d < 14$, $14 \leq C_d < 28$, and $C_d \geq 28$ denote low, moderate, considerable, and high degrees of contamination, respectively.

Abrahim and Parker (2008) proposed a modified and generalized form of the Contamination degree defined by Hakanson (1980), arguing that Hakanson's formula has some limitations for the analysed factors and the average concentration of a polluting element should be based on analysis of at least three samples of affected sediments (from the uppermost layers of a core or related surface sediments). The modified degree of contamination (*mCd*) is calculated by dividing the total value of analysed C_f^i by the total number of metals analysed (Eq. 3).

$$\mathrm{mCd} = \frac{\sum_{i=1}^{i=7} C_f^i}{n} \quad (3)$$

For the classification and description of the modified degree of contamination *(mCd*) in sediments the following gradations are proposed by Abrahim and Parker (2008): *mCd* < 1.5: Zero to very low degree of contamination; 1.5 ≤ *mCd* < 2: Low degree of contamination; 2 ≤ *mCd* < 4: Moderate degree of contamination; 4 ≤ *mCd* < 8: High degree of contamination; 8 ≤ *mCd* < 16: Very high degree of contamination; 16 ≤ *mCd* < 32: Extremely high degree of contamination and *mCd* ≥ 32: Ultra high degree of contamination. Another method developed by Hakanson (1980) to evaluate the potential impact of heavy metals in sediments on organisms in the marine ecosystem is the potential ecological risk index (PERI). This ecological risk assessment (E_r^i) uses the contamination factor (C_f^i) and toxicity coefficient (T_r^i) for each sampling point (Eq. 4). Hakanson (1980) determined the toxicity coefficients for all the metal elements: As =10, Cd = 30, Cr = 2, Cu = 5, Ni = 5, Pb = 5, and Zn =1 (Hakanson, 1980).

$$E_r^i = T_r^i * C_f^i \quad (4)$$

Table 1. The measured and certified values of elemental concentrations in reference materials.

Metals	*STD DS11*			*STD OREAS45EA*		
	Measured ±SD (ppm)	*Certified (ppm)*	*% Recovery*	*Observed±SD (ppm)*	*Certified (ppm)*	*% Recovery*
As	45.0±1.5	42.8	105.1	10.9±0.3	10.3	105.8
Cd	2.49±0.03	2.37	105.1	0.03±0.01	0.03	100.0
Cr	57.7±3.4	61.5	99.9	843.1±15.9	849.0	99.9
Cu	146.49±4.79	149.00	99.9	719.03±9.98	709.00	100.0
Ni	77.9±3.2	77.7	100.3	385.7±12.7	381.0	101.2
Pb	144.4±5.33	138.0	104.6	15.3±0.2	14.3	106.9
Zn	349.9±0.9	345.0	101.4	31.4±1.2	31.4	100.0

Table 2. Descriptive statistical analysis results for selected trace metals in sediment samples, Sediment Quality Guidelines of US EPA (1992) and background concentrations given by Taylor (1964)

		As (ppm)	*Cd (ppm)*	*Cr (ppm)*	*Cu (ppm)*	*Ni (ppm)*	*Pb (ppm)*	*Zn (ppm)*
N		100	100	100	100	100	100	100
Mean		10.5±1.0	0.07±0.03	29.8±4.5	10.33±1.47	16.3±2.0	15.05±2.32	35.3±10.8
Minimum		0.7	0.01	1.2	0.60	1.2	1.56	4.3
Percentiles 25		3.1	0.01	6.3	2.36	4.1	4.15	11.3
Median		8.4	0.03	15.0	5.81	10.2	8.34	19.4
Percentiles 75		15.1	0.04	32.4	12.38	21.6	16.40	33.8
Maximum		65.2	3.15	250.5	107.55	147.3	162.35	1090.3
Skewness		2.4	9.65	3.2	4.15	3.7	5.01	9.6
Kurtosis		9.3	95.09	11.7	22.23	20.0	29.04	94.0
K_S p		0.000	0.000	0.000	0.000	0.000	0.000	0.000
SQGs	**Non-polluted**	<3	_	<25	<25	<20	<40	<90
	Moderately polluted	3- 8	_	25-75	25-50	20-50	40-60	90-200
	Heavily polluted	>8	>6	>75	>50	>50	>60	>200
Background concentrations		1.8	0.2	100	55	75	12.5	70

The potential ecological risk index denoted Ri is a complex pollution index that sums the ecological risk factors (E_r^i) of heavy metals under consideration as in (Eq. 5) (Hakanson, 1980).

$$Ri = \sum_{i=1}^{7} E_r^i \tag{5}$$

The following terminology is used for the interpreting Ri value: An Ri under 150 is considered low ecological risk, $150 \leq Ri < 300$ is considered moderate ecological risk, $300 \leq Ri < 600$ is considerable ecological risk and an $Ri \geq 600$ is very high ecological risk.

2.3.3. Enrichment factor

Enrichment factors (*EF*) are calculated to determine whether the selected metals in the sediment samples are anthropogenic in origin. The EF is an index that normalizes the measured metals according to a reference metal such as Fe or Al. Since Fe and Al are naturally abundant in the earth's crust and remain stationary and immobile during the sediment transport process, they are used as a reference element (Yarahmadi and Ansari, 2018). In this research study, Al was used as the reference element for geochemical normalization and the metal enrichment factor suggested by Simex and Helz (1981) was calculated with (Eq. 6):

$$EF = \frac{(C_x/C_{nl})_{sample}}{(C_x/C_{nl})_{Background}} \tag{6}$$

Here, $C_{x,sample}$, $C_{nl,sample}$, $C_{x,Background}$ *and* $C_{nl,Background}$ denotes the metal concentration in the sediment sample, normalizing element concentration in the sediment sample, metal concentration in the earth's crust, and the normalizing element concentration in the earth's crust. Table 3 gives the terminologies used to describe the sediment quality according to metal enrichment factors (Anbuselvan et al., 2018; Hasan et al., 2013; Talas et al., 2015).

Table 3. Classification of enrichment factors

EF value	*Designation of sediment quality*
EF < 1	No enrichment
1 < EF <3	Minor enrichment
3 ≤ EF <5	Moderate enrichment
5 ≤ EF <10	Moderately severe enrichment
10 ≤ EF <25	Severe enrichment
25 ≤ EF <50	Very severe enrichment
50	Extremely severe enrichment

2.4. Data treatment, statistical analysis, and mapping distributions

Basic statistical parameters of trace metal concentrations for 100 coastal sediment samples were determined with IBM SPSS 20.00 software (IBM Corp. in Armonk, NY). The spatial variability of trace metals is a critical part of environmental observation and ecosystem evaluation (Yang et al., 2009). To aid in data visualization, the spatial data obtained for ecological risk assessment was stored and visualized with the help of Geographical Information Systems (GIS). Bubble maps for trace metal concentration and localized pie charts for ecological risk indices were created with ESRI ArcGIS 10.3 for Windows.

3. Results and discussion

3.1. Distributions of trace metals

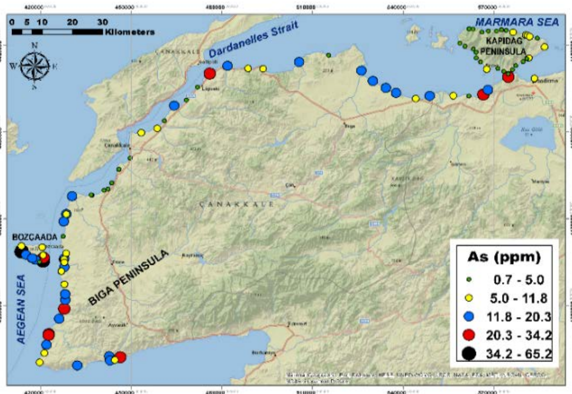
Statistics parameters such as the mean, median, minimum, and maximum for As, Cd, Cr, Cu, Ni, Pb and Zn concentrations for the coastal sediment samples are depicted in Table 2. The Kolmogorov-Smirnov normality test results indicate trace metal distributions do not follow the normal distribution. Due to this non-normality, this study represents the central weight by the median value instead of the arithmetic mean (Canbaz Öztürk et al., 2013). Furthermore, in order to visualize and spatially describe the trace metal concentration distributions, the bubble maps were created and shown in Figs. 2 and 3.

Arsenic concentrations ranged from 0.7 ppm to 65.2 ppm with a mean value of 10.5 ± 1.0 ppm. Three extremely high values in the As distribution were measured in the samples S76 (34.2 ppm) from Biga Peninsula, and S98 (44.0 ppm) and S93 (65.2 ppm) from the Bozcaada coast (Fig. 2a). Cadmium concentrations varied between 0.01 ppm and 3.15 ppm. The average Cd value was 0.07 ± 0.03 ppm. An extremely high value was detected at a single point in the S49 (3.15 ppm) sample taken from the Dardanelles coast. The other high values are the S17 (0.24 ppm), S18 (0.37 ppm) and S19 (0.14 ppm) samples from Kapıdağ Peninsula; S76 (0.18 ppm) from Biga coast; and S100 (0.08 ppm) and S94 (0.16 ppm) from Bozcaada (Fig. 2b). Chromium concentrations ranged from 1.2 ppm to 250.5 ppm with an average of 29.8 ± 4.5 ppm. Higher values come from samples S62 (99.4 ppm), S67 (246.7 ppm), S70 (105.2 ppm), S73 (89.0 ppm), S81 (141.0 ppm), S82 (250.5 ppm) and S83 (164.4 ppm) from the Biga Peninsula coasts and S91 (78.1 ppm) and S100 (173.3 ppm) from Bozcaada Island (Fig. 2c).

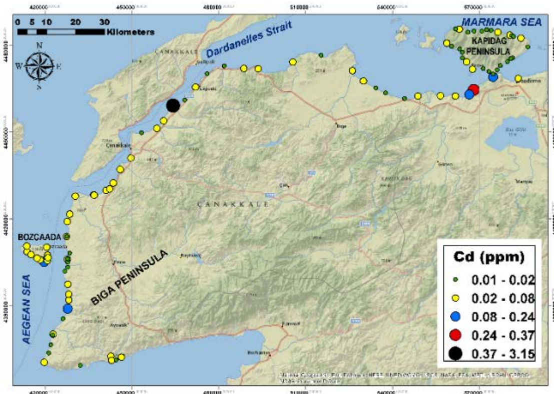
Copper concentrations varied from 0.60 ppm to 107.55 ppm with an average of 10.33 ± 1.47 ppm. The maximum value was detected in S58 from Biga Peninsula and other higher values were found in S18 (74.39 ppm) from Kapıdağ Peninsula and S76 (51.08 ppm) from Biga Peninsula (Fig. 2d). Nickel concentrations ranged from 1.2 ppm to 147.3 ppm with a mean value of 16.3 ± 2.0 ppm. The highest Ni concentration was seen in Bozcaada Island in sample S100 (147.3 ppm) (Fig. 3a). Pb concentrations in the Northwest Anatolia varied between 1.56 ppm and 162.35 ppm (Fig. 3b), while Zn concentrations ranged from 4.3 ppm to 1090.3 ppm (Fig. 3c). The average values for Pb and Zn were 15.05 ± 2.32 ppm and 35.3 ± 10.8 ppm, respectively. The highest Zn and Pb concentrations were found in the sediment coded S49 taken from the Dardanelles.

For a better global visibility, the very long and detailed study area was evaluated by dividing it into 4 regions: (1) Kapıdağ Peninsula and Karabiga Peninsula coasts in the Marmara Sea, 44 samplings from S16 to S44; (2) The Dardanelles Strait coasts in the Marmara Sea, 16 samplings from S45 to S89; (3) Biga Peninsula coasts in the Aegean Sea, 30 samplings from S83 to S61; and (4) Bozcaada Island coasts in the Aegean Sea, 10 samplings from S91 to S100 as seen from Fig. 1. The min, max, and mean values of measured trace metals were given in Table 4 according to these regions and compared with the other studies in literature conducted on trace metal distribution in coastal sediment samples.

While the average As concentration decreased from the Marmara Sea to the Aegean Sea for this study, it was lower than the values given by Yalcin and Ilhan (2008) in Kizkalesi Coast (Mersin), Turkey; Pekey et al. (2004) in Izmit Bay Northeastern Marmara Sea, Turkey; Tunca et al. (2018) in the Aegean Sea Turkey; and it was higher than those of Yalcin et al. (2016) in Antalya, Turkey; Luo et al. (2010) in Northern Bohai and Yellow Seas, China; Borghesi et al. (2016) in the Adriatic Sea, Pialassa della Baiona, Italy; El-Sorogy et al. (2021) in Red Sea coast, Saudi Arabia.



(a)



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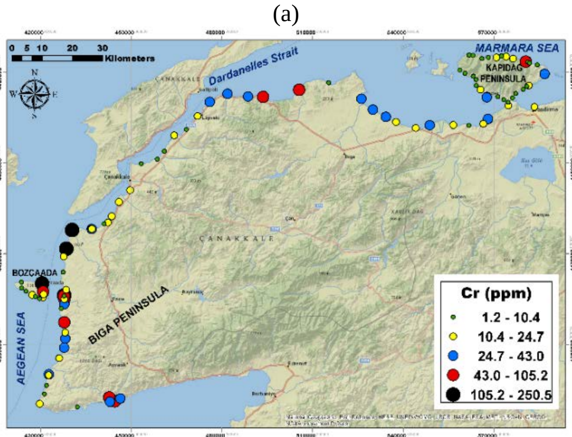

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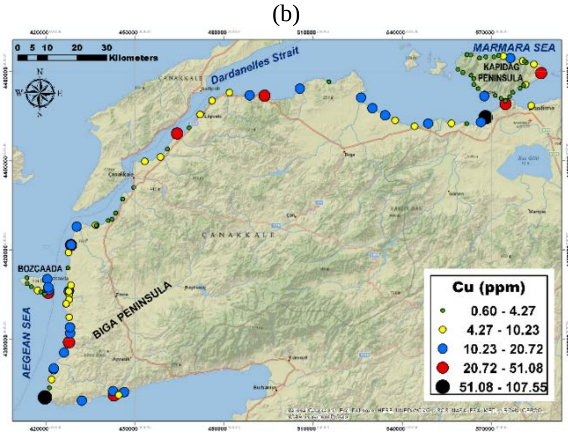
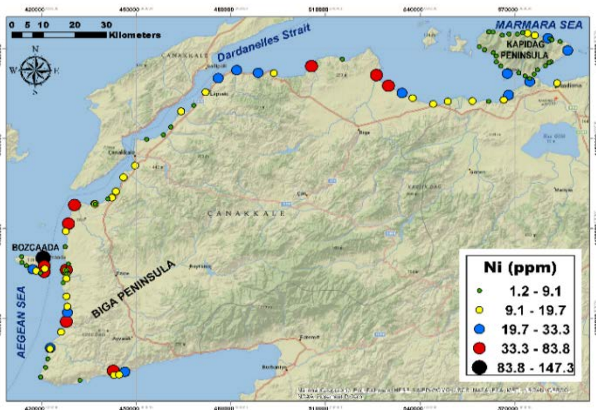


(d)

Fig. 2. Distributions of (a) As, (b) Cd, (c) Cr, (d) Cu in coastal sediments from Northwest Anatolia



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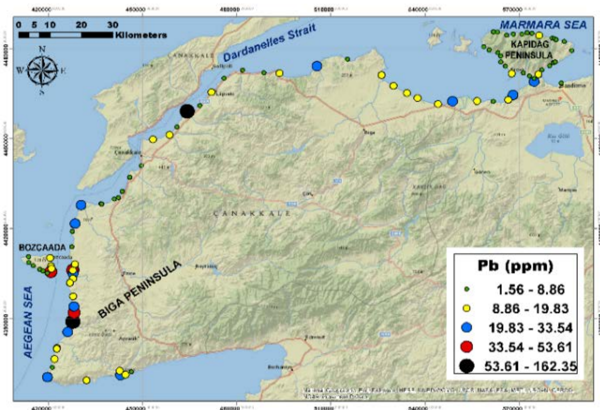

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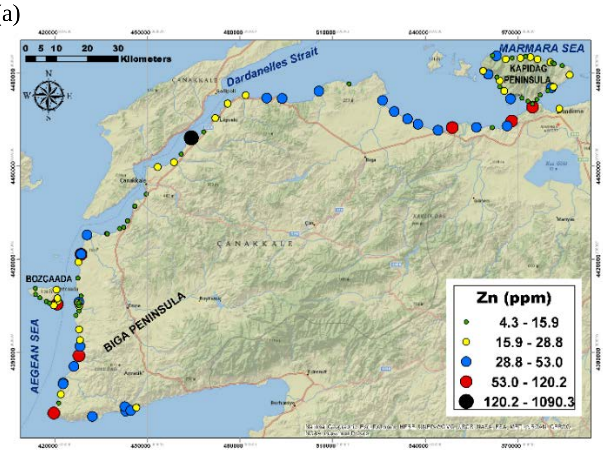


(c)

Fig. 3. Distributions of (a) Ni, (b) Pb and (c) Zn in coastal sediments from Northwest Anatolia

Table 4. The comparison with the other studies in literature conducted on trace metal distribution in coastal sediment samples

Location	*As (ppm)*	*Cd (ppm)*	*Cr (ppm)*	*Cu (ppm)*	*Ni (ppm)*	*Pb (ppm)*	*Zn (ppm)*	*References*
Red Sea, northwest Saudi Arabia	-	0.10-0.26 (0.18)	7.5-33.8 (20.2)	5.6-33.0 (18.7)	4.6-24.0 (13.7)	1.5-5.7 (3.5)	7.7-27.1 (16.7)	Kahal et al. (2018)
Red Sea coast, Saudi Arabia	2.8-12.4 (6.8)	0.14-0.55 (0.30)	16.8-42.2 (27.1)	20.8-54.4 (35.9)	16.4-28.6 (23.5)	4.4-10.8 (7.7)	38.0-118.0 (80.4)	El-Sorogy et al. (2021)
Adriatic Sea, Pialassa della Baiona, Italy	5.90-7.80 (6.71)	0.11-0.68 (0.35)	47.0-70.6 (56.2)	22.43-84.08 (47.19)	46.30-56.50 (51.09)	11.99-27.92 (21.41)	58.2-306.6 (140.7)	Borghesi et al. (2016)
Northern Bohai and Yellow Seas, China	5.6-13.0 (8.5)	0.05-0.83 (0.15)	4.2-94.0 (47.0)	0.53-35 (13.00)	-	9.5-49.0 (25.0)	9.8-170.0 (60.0)	Luo et al. (2010)
Coastal Bohai Bay, China	-	0.12-0.66 (0.22)	60.1-224.5 (101.4)	20.1-62.9 (38.5)	23.4-52.7 (40.7)	20.9-66.4 (34.7)	55.3-457.3 (131.1)	Gao and Chen (2012)
Antalya, Turkey	5.0-36.0 (10.6)	0.1-0.3 (0.14)	41.0-202.0 (112.9)	3.3-22.1 (10.1)	5.8-35.0 (18.5)	3.9-17.6 (7.4)	8.0-55.0 (21.6)	Yalcin et al. (2016)
Kizkalesi Coast (Mersin), Turkey	20.1-32.5 (24.7)	3.7-4.7 (4.2)	325.0-889.0 (553.8)	6.2-14.5 (10.1)	117.3-374.5 (186.8)	3.0-5.9 (4.5)	12.9-30.4 (19.7)	Yalcin and Ilhan (2008)
Izmit Bay Northeastern Marmara Sea, Turkey	20.0-26.8	3.3-8.9	57.9-116.1	60.6-139.0	38.4-70.7	23.8-178.0	500.0-1190.0	Pekey et al. (2004)
Northern Coast of the Marmara Sea, Turkey	-	<0.02-0.50	27.21-61.54	12.7-30.6	20.53-53.88	21.6-31.9	34.1-50.9	Topçuoğlu et al. (2004)
Yeniköy, Edremit, Ayvalık, Dikili, Aliağa, Aegean Sea Turkey	14.01 ± 5.86 35.50 ± 7.92 36.73 ± 15.25 16.72 ± 6.87 13.41 ± 4.43	0.15 ± 0.09 0.23 ± 0.14 0.47 ± 0.20 0.15 ± 0.19 0.27 ± 0.11	24.52 ± 2.67 29.10 ± 19.87 23.24 ± 10.07 17.73 ± 2.09 15.11 ± 2.10	4.49 ± 1.71 5.05 ± 3.00 8.92 ± 1.38 7.40 ± 5.89 8.36 ± 2.45	36.38 ± 3.67 56.09 ± 36.54 41.42 ± 10.64 21.19 ± 15.48 19.84 ± 5.41	13.68 ± 10.77 9.36 ± 2.15 14.78 ± 9.73 2.23 ± 1.20 3.96 ± 1.43	32.50 ± 6.96 43.04 ± 29.27 60.73 ± 3.78 42.38 ± 31.95 72.39 ± 18.44	Tunca et al. (2018)
Kapıdağ-Karabiga Peninsula	0.7-26.5 (6.9±1.0)	0.01-0.37 (0.04±0.01)	1.2-66.2 (17.7±2.7)	0.60-74.39 (8.30±1.85)	1.2-57.1 (12.4±1.9)	1.56-29.36 (8.59±1.13)	5.8-66.7 (25.1±2.3)	This study
Dardanelles Strait	2.0-29.4 (7.4±1.8)	0.01-3.15 (0.23±0.10)	4.0-30.1 (15.7±2.3)	2.00-28.64 (5.43±1.64)	3.3-29.7 (11.9±1.8)	3.58-162.35 (16.51±9.75)	7.7-1090.3 (81.9-67.2)	This study
Biga Peninsula coasts	2.0-34.2 (14.0±1.3)	0.01-0.18 (0.03±0.01)	4.2-250.5 (53.6±12.3)	3.23-107.55 (16.41±3.62)	3.0-55.3 (18.2±2.7)	6.22-156.02 (25.10±5.02)	5.9-120.2 (30.5±4.4)	This study
Bozcaada Island	3.5-65.2 (20.8±6.1)	0.03-0.16 (0.06±0.01)	2.6-173.3 (34.3±16.9)	1.35-37.30 (8.85±3.51)	2.1-147.3 (34.5±14.8)	2.99-40.82 (10.99±3.70)	4.3-71.2 (19.7±6.3)	This study

The mean Cd and Zn values for Dardanelles Strait were higher than the other regions in this study. Cr, Cu, and Pb average values for Biga Peninsula coasts were higher than the other studied parts of this study. The mean Ni value was higher in Bozcaada. The mean Cd values were lower than those of Yalcin and Ilhan (2008); Pekey et al. (2004) and were close to the other references in Table 4. The mean Cr values were similar to the results of Kahal et al. (2018), El-Sorogy et al. (2021), Topçuoğlu et al. (2004), and Tunca et al. (2018); were lower than Gao and Chen (2012), Yalcin et al. (2016), and Yalcin and Ilhan (2008). Even the highest Cu mean was lower than Kahal et al. (2018), El-Sorogy et al. (2021), Borghesi et al. (2016), and Gao and Chen (2012). The study conducted in the Marmara Sea by Pekey et al. (2004) reported higher values as compared to the other regions.

3.2. Assessment according to SQGs by USEPA

The ecological risks arising from the accumulation of the trace metals investigated in Northwest Anatolia coastal sediments were evaluated according to the Sediment Quality Guidelines (SQG) proposed by the US EPA (Giesy and Hoke, 1990; USEPA, 1992). Table 2 depicts the studied trace metals categorized into the three classes specified by the SGQs of the US EPA. When Table 2 is evaluated together with Table 4, Biga Peninsula and Bozcaada Island coasts are classified as heavily polluted in terms of arsenic; Kapidağ-Karabiga Peninsula and Dardanelles Strait are classified as moderately polluted. Even the highest Cd value in the distribution did not exceed 6 ppm proposed by the SQGs of the US EPA. Accordingly, the Northwest Anatolia and Bozcaada Island coasts were classified as non-polluted in terms of Cd. Nine sampling stations in Northwest Anatolia coasts produced samples with Cr concentrations exceeding the 75 ppm heavily polluted specification. These stations are located in the coast off of Biga Peninsula at S62 (99.4 ppm), S67 (246.7 ppm), S70 (105.2 ppm), S73 (89.0 ppm), S81 (141.0 ppm), S82 (250.5 ppm) and S83 (164.4 ppm) and off the coast of Bozcaada Island S91 (78.1 ppm) and S100 (173.3 ppm) (Fig. 4c).

In the distribution of Cr concentrations, 50% of the sediment samples have values of 15 ppm and below. Thus, if it is evaluated according to the 4 sub-fields in the study area, Kapıdağ-Karabiga Peninsula and Dardanelles Strait are classified as non-polluted; Biga Peninsula and Bozcaada Island coasts are classified as moderately polluted in terms of Cr. The sediment samples S18 (74.39 ppm) from Kapıdağ Peninsula, and S76 (51.08 ppm) and S58 (107.55 ppm) from Biga Peninsula are heavily polluted with Cu. However, the overall median value for Cu is 5.81 ppm with 75% percent of the distribution well below the non-polluted specification of 12.38 ppm. A few samples located in the Karabiga Peninsula S42 (57.1 ppm), Biga Peninsula S63 (55.3 ppm), Bozcaada Peninsula S91 (83.8 ppm) and S100 (147.3 ppm) were heavily polluted with respect to Ni. Kapıdağ-Karabiga Peninsulas, Dardanelles Strait and Biga Peninsula are classified as non-polluted; and Bozcaada coast is classified as moderately polluted in terms of Ni (Table 4). Further, the Northwest Anatolia and Bozcaada coasts are found to be overall non-polluted with respect to Zn and Pb.

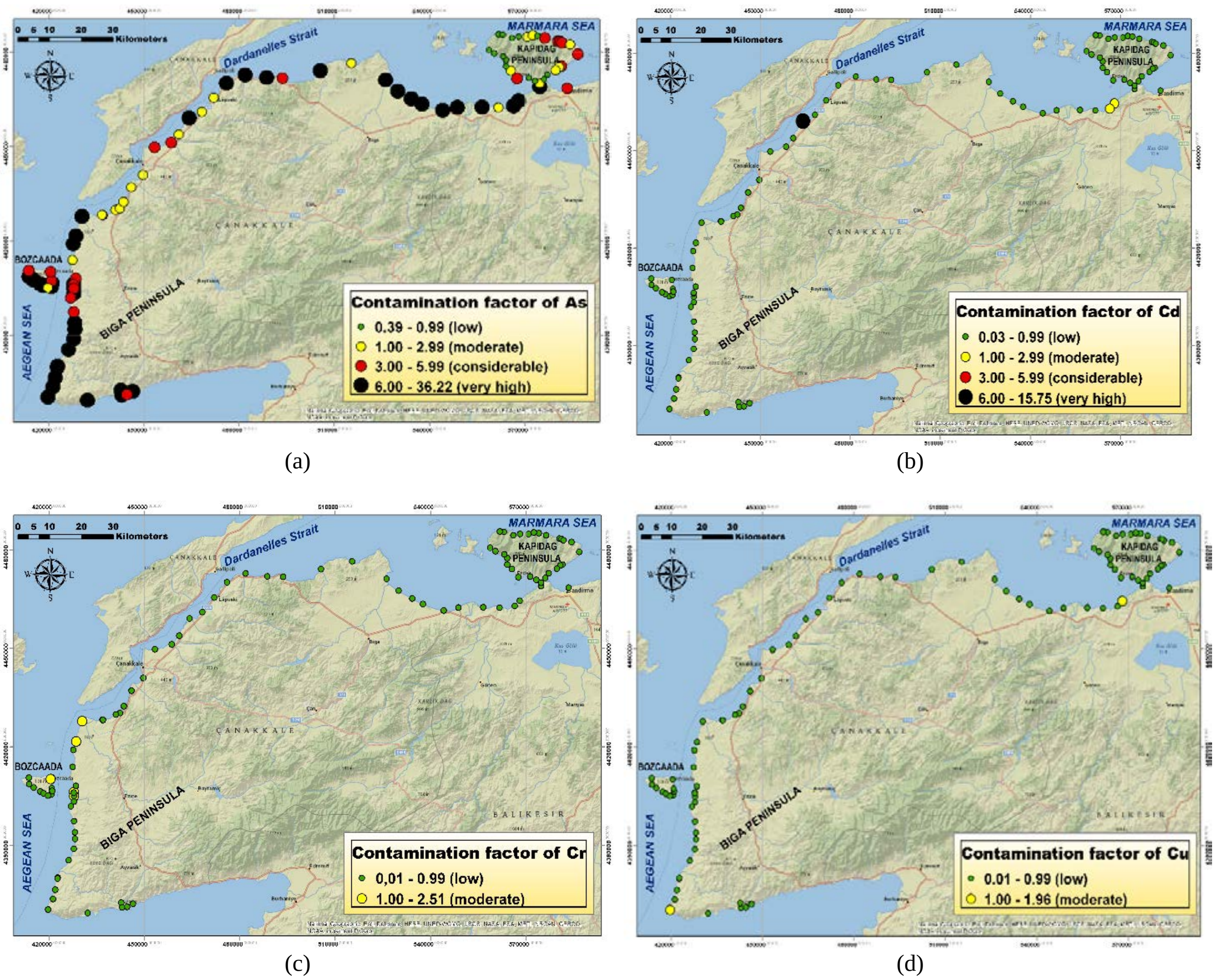


Fig. 4. Mapping of (a) As, (b) Cd, (c) Cr, (d) Cu according to contamination factor

3.3. Classification according to Cd, mCd, Ri

The contamination factors were calculated and mapped for each examined trace metal in Northwest Anatolia (Figs. 4 and 5). The contamination factors for As range from 0.4 to 14.7 with a mean value of 3.8 for Kapıdağ-Karabiga Peninsula; from 1.1 to 16.3 with a mean value of 4.1 for Dardanelles Strait; from 1.1 to 19.0 with a mean value of 7.8 for Biga Peninsula; from 1.9 to 36.2 with a mean value of 11.6 for Bozcaada Island, indicating that considerable contamination factors for Kapıdağ-Karabiga Peninsula and Dardanelles Strait, and very high contamination factors for Biga Peninsula and Bozcaada Island (Fig. 4a). In the Kapıdağ-Karabiga region, the contamination factors for Cd vary between 0.03-1.85 with a mean value of 0.20 and show low contamination factors except for the sediment samples S17 (1.20) and S18 (1.80) having moderate contamination for Cd (Fig. 4b). Dardanelles Strait indicates moderate contamination factor, varying between 0.03-15.75 with a mean value of 1.14. The maximum value (15.75) indicates a very high contamination factor in sample S49. The mean contamination factors of 0.16 (0.03-0.90) for Biga Peninsula and 0.30 (0.15-0.80) for Bozcaada show a low contamination factor.

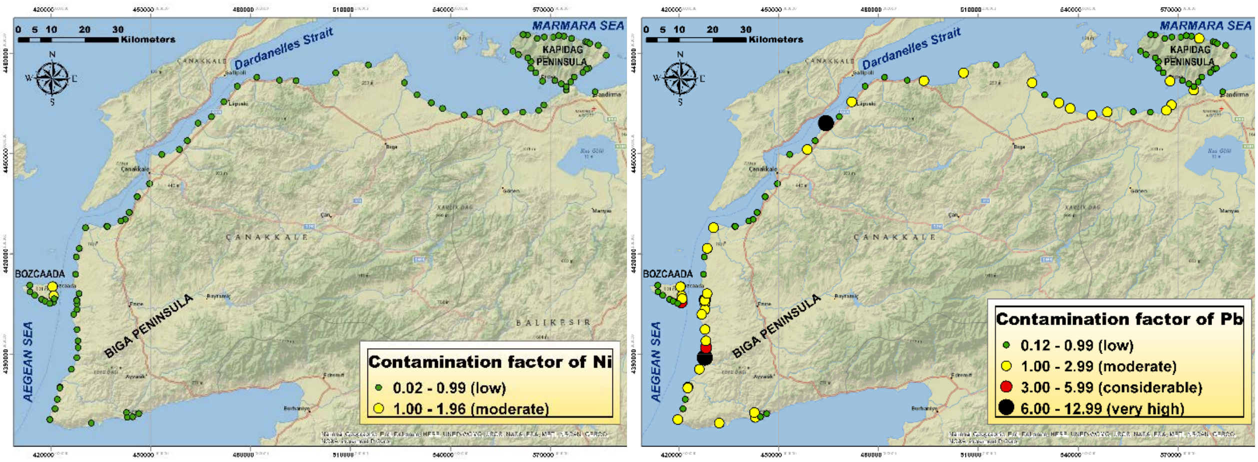
Low contamination factors for Cr have observed overall the study area with a few exceptions having moderate contamination only in six sampling sites S67 (2.47), S70 (1.05), S81 (1.41), S82 (2.51), S83 (1.64) in Biga Peninsula and S100 (1.73) in Bozcaada (Fig. 4c). Only samples S18 (1.35) and S58 (1.96) show moderate contamination for Cu (Fig. 4d). The all-study area shows a low contamination factor for Cu.

The low contamination factors for Ni are noticed throughout the entire study area, just samples S91 (1.12) and S100 (1.96) show moderate Ni contamination (Fig. 5a). Kapıdağ-Karabiga Peninsulas and Bozcaada Island have low contamination factors for Pb. Biga Peninsula and Dardanelles Strait indicate moderate contamination factor of Pb. Also, samples S49 (12.99) and S76 (12.48) show very high contamination factors; samples S67 (4.29), S75 (4.15), and S93 (3.27) show considerable contamination, locally (Fig. 5b).

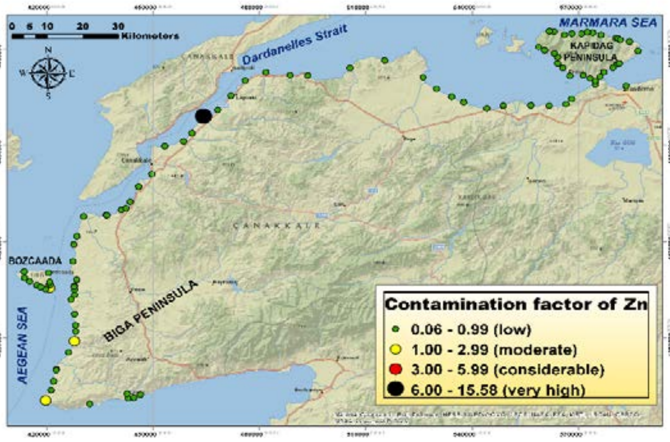
A very high contamination factor for Zn (15.58) was detected at a single point in station S49 (Fig. 5c). Furthermore, samples S58 (1.72), S76 (1.21), and S93 (1.02) show moderate contamination. Apart from these localized measured contaminations, the whole study area has very low contamination factors for Zn. The contamination degree, the modified degree of contamination, and the potential ecological risk index were investigated. The min, max, and mean values for these risk parameters according to four different sub-regions of the Northwest Anatolia coasts were tabulated (Table 5) and mapped for each sampling location (Fig. 6).

From Table 5, Kapıdağ-Karabiga Peninsula and Dardanelles Strait demonstrate a low degree of contamination, Biga Peninsula and Bozcaada Island show a moderate degree of contamination. As to the classification based on the modified degree of contamination, Kapıdağ-Karabiga Peninsula and Dardanelles Strait have a very low degrees of contamination, Biga Peninsula has a low degree of contamination, and Bozcaada Island has a moderate degree of contamination.

With respect to the potential ecological risk index, the entire study area has low ecological risk. According to the map in Fig. 6a, S49 (54.00), S76 (35.43) and S93 (42.32) indicate a very high degree of contamination. Whereas these sampling locations show high degree contamination according to mCd (Fig. 6b) because mCd takes into consideration the effect of all metals measured at one point. Similarly, the potential ecological risk index considers the toxicity coefficient of metals by multiplying with the contamination factor and represents the sum of the risks for all elements at each sampling point.



(a) (b)

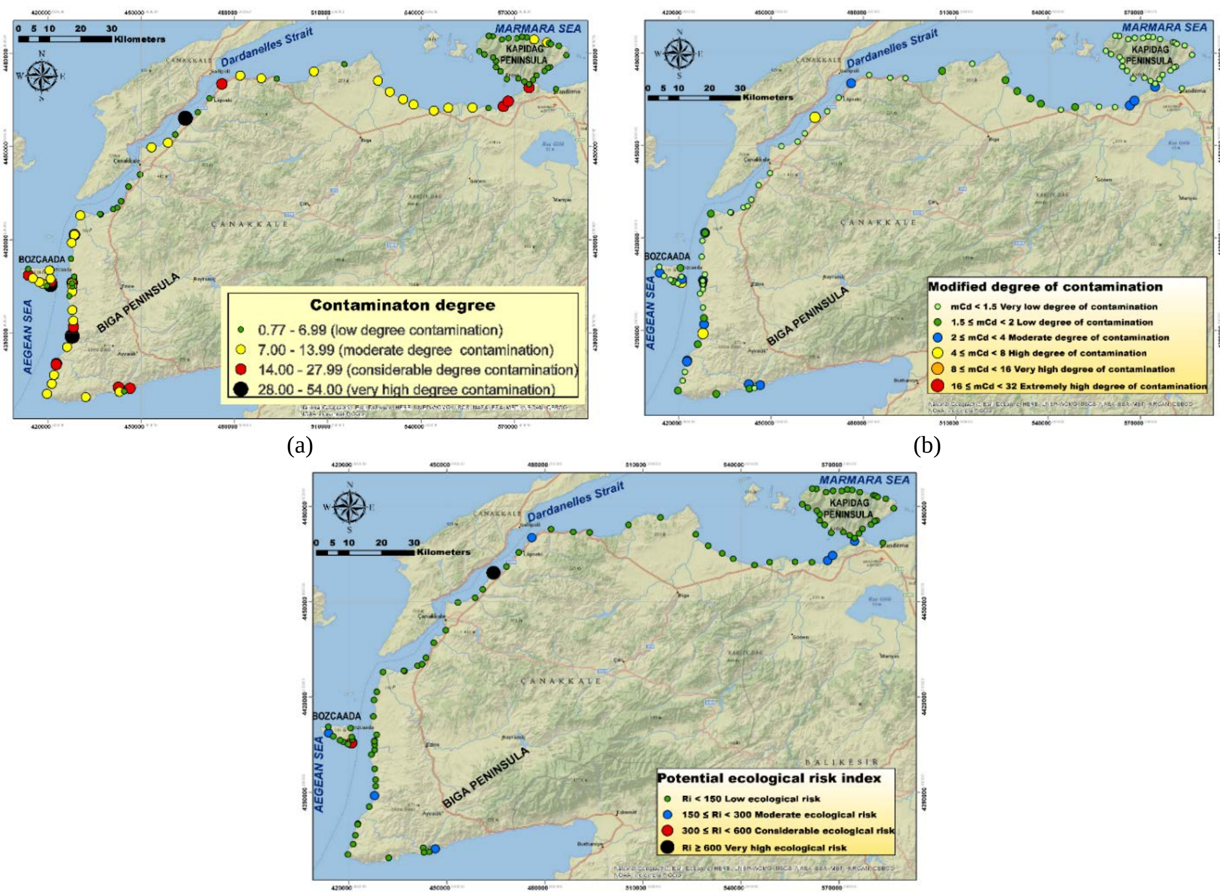


(c)

Fig. 5. Mapping of (a) Ni, (b) Pb and (c) Zn according to contamination factor

Table 5. The min, max and mean values of contamination degree, modified degree of contamination and potential ecological risk index according to four different sub-regions of the Northwest Anatolia coasts

Pollution indices	***Value***	***Kapıdağ-Karabiga Peninsula***	***Dardanelles Strait***	***Biga Peninsula coasts***	***Bozcaada Island***
Contamination degree (*Cd*)	Min-Max	0.8-18.5	2.1-54.0	2.0-35.4	3.3-42.3
	Mean	5.6	8.2	11.5	14.0
Modified degree of contamination (*mCd*)	Min-Max	0.1-2.6	0.3-7.7	0.3-5.1	0.5-6.0
	Mean	0.8	1.2	1.6	2.0
Potential ecological risk index (*Ri*)	Min-Max	5.8-193.3	20.3-645.6	17.4-288.7	42.0-396.7
	Mean	50.2	84.9	96.8	133.1



(a) (b)

(c)

Fig. 6. Mapping of the trace metals according to (a) contamination degree, (b) modified degree of contamination, (c) potential ecological risk index

3.4. Classification according to enrichment factor

The enrichment factors of the selected metals in Northwest Anatolia coastal sediment samples are depicted with a pie chart to show *EF* < 10 and *EF* > 10 metals in Fig. 7. For Kapıdağ-Karabiga Peninsula As indicates very severe enrichment, Cd, Cr, Cu, and Ni show Minor enrichment, Pb is moderately severe enrichment, and Zn moderate enrichment. Thus, the metal enrichment ranking for Kapıdağ-Karabiga Peninsula is as: As> Pb> Zn> Cd> Cr> Ni> Cu. In Dardanelles Strait, As is extremely severe enrichment, Pb is severe enrichment, Cd and Zn are moderately severe enrichment, Cr and Ni are moderate enrichment, Cu is minor enrichment.

The metal ranking is As> Pb> Zn> Cd> Cr> Ni> Cu. Arsenic is extremely severe enrichment, Pb is severe enrichment, Cr is severe enrichment, Zn is moderately severe enrichment, Cu and Ni are moderate enrichment, Cd is minor enrichment in Biga Peninsula with ranking As> Pb> Cr > Zn > Cu > Ni> Cd. Arsenic is extremely severe enrichment, Pb is severe enrichment, Cr is severe enrichment, Zn is moderately severe enrichment, Cu and Ni are moderate enrichment, Cd is minor enrichment in Biga Peninsula with ranking As> Pb> Cr > Zn > Cu > Ni> Cd. Bozcaada Island indicates extremely severe enrichment of As, severe enrichments of Cd and Pb, moderately severe enrichments of Cr, Ni, and Zn, moderate enrichment of Cu. The sequence is as follows: As> Pb> Cd > Ni> Cr >Zn > Cu.

The ecological risk parameters, such as Cd, *mCd*, *Ri*, and *EF*, use the ratio of the metal concentration to the background value of the aforementioned metal in the study area. If the regional concentrations are lower than the crustal averages, it is difficult to exhibit the existence of an anthropogenic impact and vice versa. Thus, this may be the reason that the enrichment factor results showed higher metal enrichment compared to the above cited pollution indices in the study.

There may be a higher arsenic background in this region or Al concentrations may be lower than the crustal background. From the former studies assessing the ecological risk, it is seen that some studies use crustal average values determined by Krauskopf (1979), Martin and Whitfield (1983), Taylor (1964), or Turekian and Wedepohl (1961) as background values, some measure their own background value in their study area (Li et al., 2016), and some accept the values of other studies conducted in the so-called region (Luo et al., 2010).

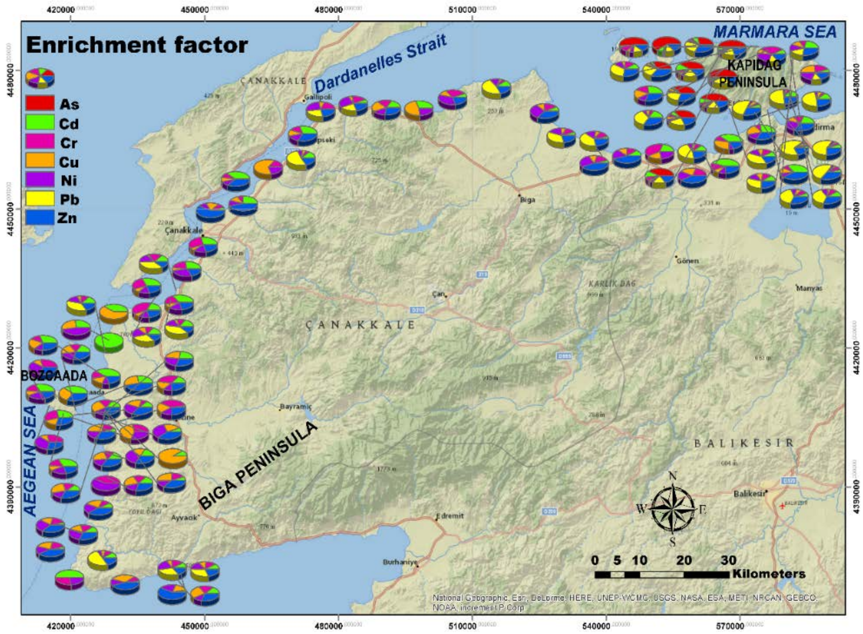
Because there are no measured background values in this study, the crustal average values given by Taylor (1964) were used. Aluminum concentrations were slightly lower than crustal average values for Al. The Spearman correlation analysis was performed to understand whether the enrichment was due to high As background or low Al concentration. Therefore, correlation coefficients were calculated among Al and seven metals in sediment samples of 4 sub-areas of Northwest Anatolia.

The correlation coefficients ($p<0.05$) between Al-As was ($r=0.141$) for Kapıdağ-Karabiga Peninsula, ($r=0.494$) for Dardanelles Strait, ($r=0.732$) for Biga Peninsula, and ($r=0.722$) for Bozcaada Island. These correlation results between Al-As indicate that the As enrichment in Kapıdağ-Karabiga Peninsula and Dardanelles Strait comes from different sources and is of anthropogenic origin, whereas for Biga Peninsula and Bozcaada Island, the As enrichment depends not only on anthropogenic inputs but also on lithogenic origin. For Kapıdağ-Karabiga Peninsula, the other significant correlation As-Pb ($r=0.793$) indicates that these metals are affected by the same source or the same processes.

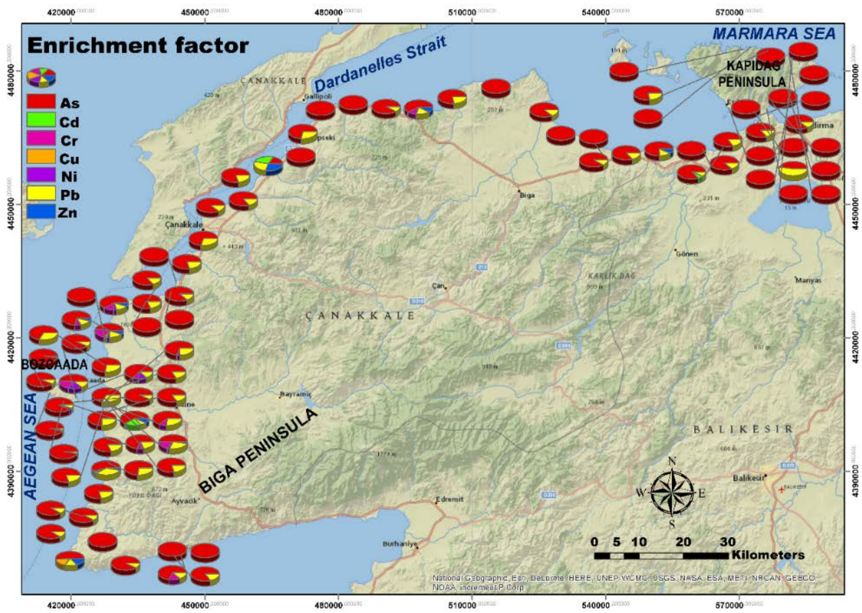
The source of enrichment in Kapıdağ-Karabiga Peninsula can be attributed to the fertilizer factory, ship dismantling plant, sulfuric acid factory, gasoline plant, leather processing facilities, and sewage input. The highest correlation coefficients for Dardanelles Strait were those for Al-Cu ($r=0.757$) and Al-Zn (0.722). The correlation between Al-Cu indicates that Cu is not affected by human activities and is of lithological origin. On the other hand, the result of moderately severe enrichment of Zn above and the correlation between Al-Zn shows that the lithogenic sources are also effective in Zn enrichment. Anthropogenic sources of the metal enrichment in Dardanelles Strait can be agricultural pesticides, fertilizers, ceramic production facilities. Dardanelles Strait as one of The Turkish Straits System is affected by oil production and maritime transportation. Furthermore, many large and small streams reaching the sea from the Dardanelles Strait can lead to reaching of the metal pollution to the sea (Fig. 1). The most important correlation for the Biga Peninsula was Al-As ($r=0.732$), which indicates that the As background is high in this region. In Bozcaada, the high positive correlations Al-Cr ($r=0.891$), Al-Cu (0.842), Al-Ni (0.879), and Al-Zn ($r=0.742$) show that these metals are of terrestrial origin. As can be seen in Fig. 6, the pollution in Bozcaada is in the east of the island, where the harbour is located and where human activities are intense.

4. Conclusions

Concentrations of selected trace metals (As, Cd, Cr, Cu, Ni, Pb, and Zn) in coastal sediments from 100 sites in Northwest Anatolia including Bozcaada Island were studied in order to understand current metal distribution. Ecological risk is investigated according to the sediment quality parameters. The Sediment Quality Guidelines (SQG) of the US EPA do not take into account the crustal background values of metals that may occur naturally in high concentrations in some parts of the world. Therefore, it is suggested that multiple criteria should be used together instead of a single criterion in the evaluation of sediment quality in terms of contamination and environment.



(a)



(b)

Fig. 7. The numerical proportion of the metal enrichment for (a) $EF < 10$ and (b) $EF > 10$ in Northwest Anatolia coastal sediments

Therefore, in this study, the contamination factor, the contamination degree, the modified degree of contamination, the potential ecological risk index, and the enrichment factor metrics were taken into consideration together with the Sediment Quality Guidelines (SQG) of the US EPA in determining the sediment quality parameters. Also, the spatial distribution maps were created to provide efficient estimates for trace metal monitoring in coastal sediments and valuable information for regional sediment quality management.

Although stations S49, S76 and S93 have considerable or very high potential ecological risk locally, the Northwest Anatolia coastal sediments are of low ecological risk. Enrichment of arsenic, the foremost metal in the study, is of anthropogenic origin in Kapıdağ Peninsula, Karabiga Peninsula and Dardanelles Strait in the Marmara Sea coast of the study and is of lithogenic origin in addition to anthropogenic in Biga Peninsula and Bozcaada Island in the Aegean Sea.

The study provides data on the concentrations and distributions of selected trace metals in the coastal sediments of Northwest Anatolia. The obtained data are expected to be useful in the assessment of the current and future environmental risk in the Northwest Anatolia coast which will aid in sustainable environmental planning and coastal management.

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