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Element-based ecological and human health risk assessment in a lagoon system in a densely populated basin

by

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Abstract

This study analysed the ecological deterioration and health risk in sediment samples taken from Dalyan and Poyraz Lagoons in the Karacabey floodplain of Turkey, which is under pressure from agriculture, industry and settlement activities. Multi-element analyses were performed with ICP-MS on the surface and core sediment samples from the lagoons. Total organic carbon, chlorophyll degradation products and carbonate analyses were performed to determine the transport and illuviation dynamics of the elements. While Pb and Zn showed moderate enrichment at some sampling points, no enrichment of the other elements was detected. According to ecological risk analysis data, Cd and Hg posed a moderate ecological risk at some sampling points. The modified hazard quotient data indicated very high contamination of Ni, a high level of As contamination and significant Cr contamination. A carcinogenic health risk was detected from Ni, Cr and As due to the lithological characteristics of the basin. It was concluded that the lithological characteristics, the agricultural and mining activities carried out in the Susurluk Basin – which is drained by Koca Stream – and domestic and industrial waste contributed to the higher element concentrations in the Karacabey floodplain.

Key words: Element contamination, Dalyan and Poyraz lagoons, sediment, geographic information systems

1. Introduction

Problems regarding ecological degradation have emerged along with the continuous increase in anthropogenic effects in wetlands (Jahan & Strezov 2018, Kükrer et al. 2015, Loska & Wiechuła 2003, Ozkan & Buyukisik 2012, Töre et al. 2021). One of the main causes of ecological degradation is toxic element contamination (Di Beneditto et al. 2019, Fural et al. 2020, Islam et al. 2021, Kowalska et al. 2020, Wu et al. 2022). Elements with high toxicity can remain intact in the ecosystem for a long time and cannot be purified by treatment systems (Al-Solaimani et al. 2022, Kumar et al. 2022, Ustaoğlu et al. 2020). Element contamination can spread far in aquatic ecosystems. The most striking example of this is the detection of ecological risks even on the ocean floor (Sanei et al. 2021). Elements stored in the sediment can be released back into the water when suitable ecological conditions occur (Yuan et al. 2014). This makes the elements stored in sediment from anthropogenic sources a secondary source in terms of ecological risk. Elemental contamination in wetlands affects people through the food chain and triggers health problems (Alamri et al. 2021, Ali et al. 2022, Jeong et al. 2020, Mutlu & Aydin Uncumusaoğlu 2018, Ustaoğlu et al. 2022, Zhang et al. 2016).

Floodplains are sensitive ecosystems with rich biodiversity (Al-Solaimani et al. 2022, Kowalska et al. 2020, Thoms 2003). Since floodplains are located at the junction of freshwater and saltwater sources and remain underwater in some seasons, they provide a large amount of nutrients to the organisms living in the habitat. This allows woody plants in the floodplain ecosystem to reach 20–30 meters in height and provides a suitable breeding and living area for migratory birds and native fauna (Wakeley et al. 2007). The water sources feeding the floodplains have been polluted by anthropogenic effects in recent years (Matella & Merenlender 2015). This way, the floodplains may lose their ecological characteristics (Howie et al. 2018). Therefore, in addition to the floodplains, the water resources that feed the floodplain forests must be protected from all anthropogenic effects.

Karacabey floodplain, located at the mouth of Kocasu Stream on the coast of the Marmara Sea in western Turkey, is an important wetland area facing the aforementioned problems (Figure 1). The floodplains and their surroundings are very rich in terms of biodiversity. Nearly 250 bird species, many reptiles and dominant floodplain vegetation have been detected in the floodplain area and around the lagoon (Akay et al. 2018). Being a frequent destination for migratory birds and containing a rich floral and faunal environment, the floodplain is important to this area. In recent years, some ecological degradation has been observed in the floodplain, which covers an area of 22,399 km^2. This study hypothesises that anthropogenic activities (especially agriculture) in the Susurluk basin and around the floodplain may cause ecological degradation in the floodplain and may create potential health problems for the people who live in the area.

To this end, the research problem consisted of the following four items:

- Kocasu Stream – which drains the Susurluk basin, an agricultural and industrial centre home to more than three million people – discharges into the sea near the floodplain.
- Since the floodplain floor is located at a lower elevation than the river bed and sea level, water discharges from both aquatic environments into the floodplain.
- Intensive agricultural activities continue around the floodplain, and there is a direct discharge of waste water into the floodplain.
- The floodplain does not have adequate national or international protection status.

Considering all these research problems, this aims of the study were as follows:

- Determine the ecological and health risk in the sediments of Dalyan Lagoon and Poyraz Lagoon, which are an important component of the floodplain ecosystem.
- Present the pollution records by examining the spatial and temporal changes of ecological indices.
- Identify the possible sources and transport processes of pollutants using multivariate statistical analysis.
- Calculate carcinogenic and non-carcinogenic health risks from toxic metals.

2. Materials and methods

2.1. Sediment sampling and analytical procedure

Since they can stably store PTEs (Potential Toxic Elements) for a long time, sediments are used as indicators in ecological risk studies (Cüce et al. 2022,

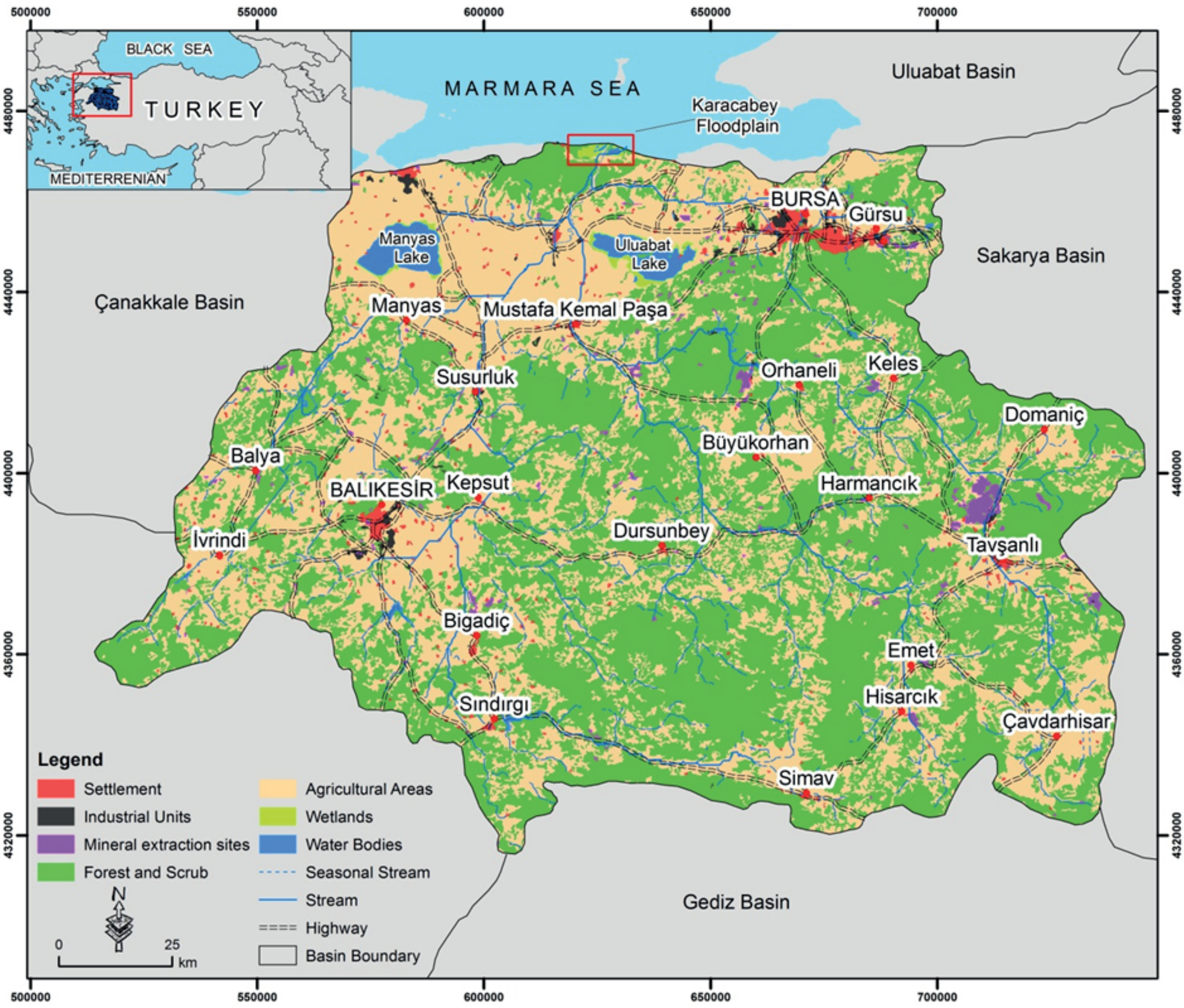

Figure 1

Location and land use map of the study area

Özkan et al. 2022, Sojka et al. 2018). Surface sediments were taken with a Van Veen grab from 18 sampling points in the Dalyan Lagoon; a core 40 cm in length was taken with a Kajak gravity core sampler from one sampling point in the Poyraz Lagoon (Figure 2). The sediments were then dried in an oven at 60°C for 24 hours, and then pulverised with the help of a porcelain mortar. Elemental analyses were performed with ICP-MS by Bureau Veritas Analytical Labs in Canada. Reference material, duplicate and blank sample measurements were taken to test the quality control and validity of the analyses. The recovery rates of the elements were found to range between 96% and 124%. Total organic carbon (TOC) analysis was done using the Walkley Black titration method (Gaudette et al. 1974) and carbonate (CO_3^{-2}) analysis was conducted with a Scheibler calcimeter (Schlichting & Blume 1966). Analysis of the chlorophyll degradation products (CDP) was performed by acetone extraction and spectrophotometry (Lorenzen 1974).

Enrichment factor (EF) (Brady et al. 2015; Sutherland 2000), toxic risk index (TRI), (Zhang et al. 2016), modified ecological risk index (mRI), modified potential ecological risk index (mPER) (Brady et al. 2015, Hakanson 1980), modified hazard quotient (mHQ) (Benson et al. 2018), ecological contamination index (ECI) (Benson et al. 2018) and contamination severity index (CSI) (Pejman et al. 2015) were used for ecological risk analysis.

Since fishing takes place in Dalyan Lagoon and Poyraz Lagoon, there is a risk of elemental

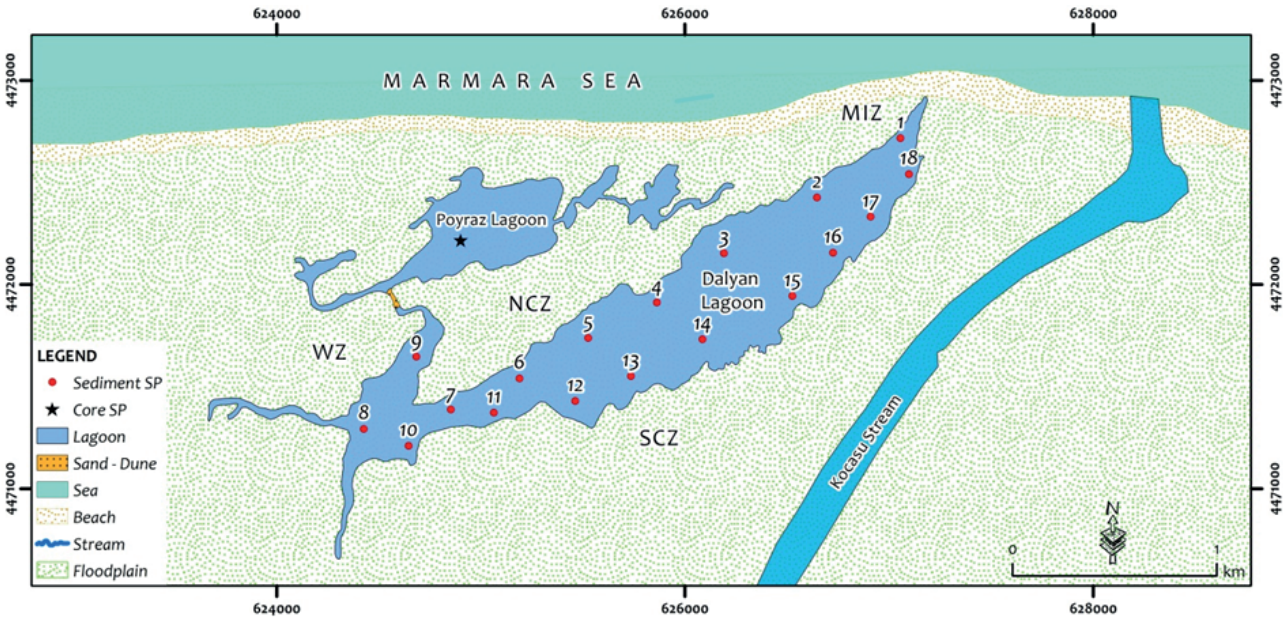

Figure 2

Sediment sampling points

contamination affecting people through the food chain. Therefore, the ecological degradation and health risks were explored together. To determine the non-carcinogenic health risk, exposure through ingestion (Ex_{ping}), exposure through contact (via skin) (Exp_{derm}) and hazard indices (HI) were calculated (Sun et al. 2018, USEPA 2005). The lifetime cancer risk index (LCR) was calculated to detect carcinogenic health risks (Iqbal et al. 2013, USEPA 2005).

The EF index was used to identify the natural and anthropogenic sources of the elements. EF was calculated with Formula 1.

$$EF = \frac{\left(\frac{C_i}{C_{ref}}\right) sample}{\left(\frac{B_i}{B_{ref}}\right) background} \qquad (1)$$

The results of the analysis were evaluated according to the following criteria: EF < 2, no enrichment; EF = 2–5, moderate enrichment; EF = 5–20, significant enrichment; EF = 20–40, very high enrichment; and EF > 40, extraordinary enrichment (Sutherland 2000).

The toxic risk index (TRI_i) was used to determine the toxic risk level (Zhang et al. 2016). was calculated with Formula 2.

$$TRI_i = \sqrt{\frac{\left(\frac{C_i}{TEL}\right)^2 + \left(\frac{C_i}{PEL}\right)^2}{2}} \qquad (2)$$

In the formula, C_i indicates element concentration, TEL is the 'threshold effect level' and PEL the 'probable effect level' (MacDonald et al. 1996).

The integrated TRI was calculated according to Formula 3.

$$TRI = \sum_{i=1}^{n} TRI_i \qquad (3)$$

TRI data were evaluated as follows: TRI ≤ 5, no toxic risk; 5 < TRI ≤ 10, low toxic risk; 10 < TRI ≤ 15, moderate toxic risk; 15 < TRI ≤ 20, considerable toxic risk; and TRI > 20, very high toxic risk.

mER was used to determine the ecological risk level, and mPER was used to determine the potential ecological risk level. mER was calculated according to Formula 4.

$$mER = EF \times Tr^i \qquad (4)$$

In the formula, EF is the enrichment factor and Tr^i is the toxic risk coefficient of the elements (Hakanson, 1980; Brady et al., 2015). The toxic risk coefficients of the elements were as follows: Hg = 40, Cd = 30, As = 10, Cu, Pb and Ni=5, Cr = 2, Zn = 1, Mn = 1, Co = 5 and Tl = 10. The results were evaluated as follows: mER < 40, low potential ecological risk; 40 ≤ mER < 80, moderate potential ecological risk; 80 ≤ mER < 160, significant potential ecological risk; 160 ≤ mER < 320, high potential ecological risk; and mER ≥ 320, very high potential ecological risk (Hakanson, 1980).

mPER was calculated with Formula 5.

$$ {}_{m}PERI = \sum mRI \tag{5} $$

The results were evaluated as follows: mPER < 150, low ecological risk; 150 ≤ mPER < 300, moderate ecological risk; 300 ≤ mPER < 600, considerable ecological risk; and mPER ≥ 600, very high ecological risk (Hakanson, 1980).

MHQ, which is based on the method of comparing the ecological and toxicological effects of the elements with different threshold levels, was calculated with Formula 6 (Benson et al., 2018; MacDonald et al., 2000).

$$ {}_{m}HQ = \left[C_i \left(\frac{1}{TEL} + \frac{1}{PEL} + \frac{1}{SEL} \right) \right]^{\frac{1}{2}} \tag{6} $$

In the formula, C_i represents element concentration and TEL, PEL and SEL indicate the threshold impact level, probable impact level and severe impact level, respectively. The mHQ findings were evaluated as follows: mHQ < 0.5, insignificant; 0.5 ≤ mHQ <1, very low risk; 1 ≤ mHQ <1.5, low risk; 1.5 ≤ mHQ < 2, moderate risk; 2 ≤ mHQ <2.5, considerable risk; 2.5 ≤ mHQ < 3, high risk; 3 ≤ mHQ <3.5, very high risk; and mHQ > 3.5, extreme risk (Benson et al. 2018) chromium (Cr).

ECI was used to determine the integrated total ecological risk level of the elements. ECI enables resource-specific ecological risk assessment using principal component analysis/factor analysis data, and is calculated with Formula 7.

$$ ECI = B_n \sum_{i=1}^{n} mHQ_i \tag{7} $$

In the formula, B_n is the reciprocal of the eigenvalue obtained from the analysis of principal components for elements. The ECI findings were evaluated as follows: ECI < 2, uncontaminated; 2 ≤ ECI < 3, uncontaminated to slightly contaminated; 3 ≤ ECI < 4, slightly to moderately contaminated; 4 ≤ ECI < 5, moderately to considerably contaminated; 5 ≤ ECI < 6, considerably to highly contaminated; 6 ≤ ECI < 7, highly contaminated; and ECI > 7, extremely contaminated (Benson et al. 2018).

CSI was developed with the purpose of analysing elemental contamination (Pejman et al. 2015). The index is based on a source-specific approach and comparison with 'effects range low' and 'effects range median' threshold values (Long et al. 1998). CSI was calculated according to Formula 8 and Formula 9.

$$ CSI = \sum_{i=1}^{n} W_i \left[\left(\frac{C_i}{ERL_i} \right)^{\frac{1}{2}} + \left(\frac{C_i}{ERM_i} \right)^{2} \right] \tag{8} $$

In the formula, W_i is the contamination weight of PTEs, C_i is the PTE concentration and *n* is the number of PTEs used in the analysis.

$$ W_i = \frac{(loading\ value_i\ x\ eigen\ value)}{\sum_i^n (loading\ value_i\ x\ eigen\ value)} \tag{9} $$

The eigenvalue and load values of the factor/component determined to be anthropogenic in the principal components/factor analysis were used in the calculation of W_i. The CSI findings were evaluated as follows: CSI < 0.5, uncontaminated; 0.5 ≤ CSI < 1, very low contamination; 1 ≤ CSI < 1.5, low contamination; 1.5 ≤ CSI < 2, low to moderate contamination; 2 ≤ CSI < 2.5, moderate contamination; 2.5 ≤ CSI < 3, moderate to high contamination; 3 ≤ CSI < 4, high contamination; 4 ≤ CSI < 5, very high contamination; and CSI ≥ 5, ultra-high contamination.

Exposure through ingestion (Exp_{ing}), an important component of health risk indices, was calculated with Formula 10.

$$ Exp_{ing} = \frac{C_{sed} \times IR \times CF \times EF \times ED}{BW \times AT} \tag{10} $$

In the formula, C_{sed} indicates element concentration, IR is the absorption rate (114 mg day^{-1}), CF is the unit conversion (10^{-6} kg mg^{-1}), EF is the exposure frequency (350 days $year^{-1}$), ED is the exposure duration (30 years), BW is the mean body weight (70 kg) and AT is the average number of days exposed (30 years × 365 = 10.950 days). According to this formula, a person weighing 70 kg is assumed to be exposed to 114 mg of sediment per day for the duration of 30 years (Song et al. 2019).

Exposure through skin/contact (Exp_{derm}) was calculated with Formula 11.

$$ Exp_{derm} = \frac{C_{sed} \times CF \times SA \times AF \times ABS \times EF \times ED}{BW \times AT} \tag{11} $$

In the formula, SA refers to the skin surface area exposed (5700 cm^2), AF is the *soil-to-skin* adherence factor of the sediment (0.07 mg cm^{-2}) and ABS is the absorption factor of the sediment (0.001) (Iqbal et al. 2013).

HQ and HI are used to detect non-carcinogenic health risks from ingestion and dermal exposure of metals (USEPA, 2005). HQ (hazard quotient) was calculated with Formula 12.

$$ HQ_{ing} / HQ_{derm} = \frac{Exp_{ing} / Exp_{derm}}{RfD0} \tag{12} $$

In the formula, Exp_{ing} represents exposure through ingestion, Exp_{derm} represents exposure through contact/skin and RfDO represents the reference dose. The HI (hazard index) was calculated with Formula 13.

$$HI = \frac{\sum_{i=1}^{n} HQ_{ing}}{HQ_{derm}} \tag{13}$$

In the formula, HQ represents exposure through ingestion and contact, HI represents total non-carcinogenic health risks and RfD0 represents the reference dose. Based on analyses, HI values above 1 point to the existence of non-carcinogenic health risks in the wetland, while a value of less than 1 indicates no health risks (Iqbal et al. 2013, Mohammadi et al. 2019). Elements with high toxic effects, such as As, Pb, Cd, Cr or Hg, may cause cancer risk in case the exposure is prolonged. Cancer slope factors (CSF) for PTEs are provided as follows: As (1.5 mg kg^{-1} day^{-1}), Pb (0.042 mg kg^{-1} day^{-1}), Cd (6.30 mg kg^{-1} day^{-1}) and Cr (8.50 mg kg^{-1} day^{-1}) (EPA 2009, Mohammadi et al. 2019).

LCR (lifetime cancer risk) was calculated with Formula 14 and Formula 15.

$$LCR = Exp_{(ing-derm)} \times CSF \tag{14}$$

$$\sum LCR = (LCR_{ing} + LCR_{derm}) \tag{15}$$

In the formulas, Exp_{ing} represents exposure through ingestion, Exp_{derm} represents exposure through contact/skin and CSF represents the cancer slope factors of PTEs (Fallahzadeh et al. 2017). The acceptable carcinogen risk level is between 10^{-6} and 10^{-4}. Values over 10^{-4} indicate cancer risk, while values below 10^{-6} indicate no cancer risk (Tepanosyan et al. 2017, USEPA 2005).

Spatial analysis of the findings obtained from ecological risk indices was performed by kriging interpolation using Arc-Map 10.5 software. Multivariate statistical analysis was performed with Statgraphics 18 software to identify possible sources and transport processes of the elements.

3. Results and discussion

3.1. Spatial distribution of elements

3.1.1. Sediment characteristics

Elements are listed in ppm according to average concentration: Fe (29,727) > Al (19,872) > Mn (697) > P (637) > Ni (123.2) > Cr (122) > Zn (109) > Pb (53.12) > Cu (32.51) > As (27.30) > Co (18.50) > Mo (0.98) > Tl (0.36) > Cd (0.27) > Hg (0.081).

The spatial distribution of the elements was evaluated by dividing the floodplain into four different regions. The marine impact zone (MIZ) is the part of the floodplain with water exchange with the sea and which is open to marine influence. The western zone (WZ) is the area under the influence of the Poyraz Lagoon in the north, which is connected to the main body of the floodplain by a narrow strait. The northern coastal zone (NCZ) is the unit associated with the floodplain forest and the coast. The southern coastal zone (SCZ) is the area under the influence of floodplain forest and agricultural lands. According to the spatial analysis, it was noted that the maximum element concentrations were more extensive in WZ and MIZ (Figure 3), suggesting that significant amounts of elements were transported to the floodplain from the Marmara Sea and Poyraz Lagoon. Table 1 presents the minimum, maximum and mean values of the elements and the other variables.

The data obtained from this study were compared with the concentrations of randomly selected lakes located in different parts of Turkey and around the world. The Pb, Zn, Ni, Co, Mn, As and Cr concentrations of Dalyan Lagoon were found to be higher than those of other wetlands. The Cu and Cd concentrations of Dalyan Lagoon were lower than those of Nador Lagoon. The Fe concentration was lower than that of Chilka Lake and higher than that of other wetlands. The Al and Hg concentrations of Dalyan Lagoon were found to be lower than all the compared wetlands (Table 2).

3.2. Spatial distribution of TOC, CDP and CO_3^{-2}

TOC is associated with the transport and sedimentation processes of elements (Hefni et al. 2020). The minimum TOC concentration (0.16%) was in ST 12 and the maximum (5.71%) in ST 11. The average TOC was found to be 2.69%. It is noteworthy that the TOC concentration was high in the SCZ, which is close to the agricultural lands of Dalyan Lagoon, and in the WZ, where it is connected to Poyraz Lagoon (Figure 3). This shows that the agricultural lands in the southeast of Dalyan Lagoon and the discharge from Poyraz Lagoon are effective on the spatial distribution of TOC.

Plants store the elements they take from sediment and water inside the cell. With the collapse of the dead plants to the bottom, the elements taken from the ecosystem through the roots are carried to the sediment. Because of this plant characteristic, CDP represents the role of primary producers in element transport processes. The highest CDP concentration (121.25 μg/gr) was in the WZ and the lowest (9.06 μg/gr) in the MIZ. The mean CDP concentration was 42.91 *μg/gr*. The fact that the CDP concentration reached the maximum value at the junction of Dalyan Lagoon and Poyraz Lagoon showed that the primary production

Table 1

Minimum, maximum and mean concentrations of the variables

(ppm)	Mo	Cu	Pb	Zn	Ni	Co	Mn	Fe	As	Cd	Cr	Al	Hg	Tl	TOC (%)	CDP ($\mu g\ gr^{-1}$)	CO_3^{-2} (%)
Min	0.54	16.83	34.74	43.80	89.10	13.35	363	17500	21.20	0.10	78.40	10700	0.052	0.22	0.16	10.00	2.00
Max	2.99	46.56	136.42	230.60	146.80	23.00	911	35000	54.10	0.40	238.50	26100	0.115	0.42	5.71	121.25	23.19
Mean	1.04	32.55	80.74	108.00	122.81	18.40	804	29989	30.00	0.28	120.50	19626	0.085	0.35	2.69	42.10	7.09

Table 2

Comparing different wetlands based on selected PTE concentrations

Location	Cu	Pb	Zn	Ni	Co	Mn	Fe	As	Cd	Cr	Al	Hg	Reference
Dalyan L. (Turkey)	32.51	53.12	109.02	123.18	18.49	697	29727	27.27	0.27	121.97	19872	0.081	This study
S.Moussa L.(Morocco)	30.40		49.80	29.10			28000			96.90	67800		Maanan et al. 2004
Homa L. (Turkey)	18.51	10.49	71	85.10		562	23272		0.10	103	27081	0.33	Uluturhan et al. 2011
Chilka L. (India)	50	41	42	106		314	42000			37			Panda et al. 1995
Berre L (France)	31	41.70	120						0.23	58.70			Arienzo et al. 2013
Nador L. (Morocco)	39.80	37.11	95.80	25.80	14.00	577		11.40	0.30	57			González et al. 2007

processes took place faster and more intensively in Poyraz Lagoon (Figure 3). CO_3^{-2} concentration, another variable that controls the processes of transportation to the sediment by forming a complex with the elements, varied between 1.74% and 23.19%, with an average of 7.09%. The CO_3^{-2} concentration was determined to be lowest in the MIZ and highest in the SZC. The spatial analysis of CO_3^{-2} indicates point density (Figure 3). The area surrounding Dalyan Lagoon is covered with quaternary-aged alluvium and unweathered terrestrial fragments. Alluvium and terrestrial fragments are interrupted in the south by a complex lithological series containing Upper Paleozoic/Triassic schist, marble, metabasite, etc. (MTA, 2021). The spatial analysis revealed that the high CO_3^{-2} concentration around the SCZ may be due to the aforementioned complex lithological series.

3.3. Anthropogenic impact assessment with EF

Pb (1.33) > Zn (1.28) > Cd (1.12) > P (1.03) > Cu (1.00) > Tl (0.99) > Cr (0.86) > Hg (0.79) > Co (0.77) = Mo (0.77) > Ni (0.71) > Fe (0.56) > As (0.31) > Mn (0.30), according to the mean EF values from surface sediment. According to this data, there is no problem of metal enrichment in Dalyan Lagoon. However, examination of the spatial distribution indicates point enrichment. Moderate enrichment was detected for Pb (3.81) and Zn (2.10) in the MIZ. The EF value of Cd reached 1.74 in ST 10. An EF value of > 1.5 indicates the onset of anthropogenic accumulation (Zhang & Liu 2002). It is noteworthy that the EF value of P approached the moderate enrichment limit in the SCZ (1.41), which is under the influence of agricultural lands.

The Marmara Sea is located in the north of Dalyan Lagoon, and there are agricultural lands in the south of the lagoon (Figure 2). Water is discharged into the lagoon by waves from the sea and by streams from the land, making it difficult to identify the source of the elements. Based on the spatial analysis, the likely source of moderate Pb enrichment in the MIZ is the discharge of the sea because MIZ is the region most open to the influence of the waves (Figure 2). In a study conducted on the surface sediments of the Marmara Sea on the northern shores of the Karacabey floodplain, Pb enrichment was found to be at low/moderate level due to the anthropogenic activities carried out in the Susurluk basin and its lithological characteristics (Pehlivan, 2017). The literature on the subject and spatial analysis strengthen the possibility that the source of Pb enrichment is seawater intrusion (Pehlivan et al., 2021). Zn, which was found to be moderately enriched around the MIZ, is an important plant nutrient. Most of the Susurluk basin is cultivated (Figure 1). Onion and peach cultivation is carried out intensively in the Karacabey floodplain, which is adjacent to the floodplain. Onions need Zn-containing fertilisers during development, while peaches need Zn-containing fertilisers especially during flowering (GUBRETAS, 2021). The area around the MIZ is the sampling point most exposed to the influence of Kocasu Stream (Figure 1). Therefore, the likely source of moderate Zn enrichment is the agricultural activities. The probable cause of Cd and P approaching the moderate enrichment limit is agricultural activities, as in the case of Zn, because Cd and P are important plant nutrition elements used in agriculture (Gomes & Soares, 2013; Raghothama, 2005). A previous study conducted in the floodplains concluded that the Cr, Ni, Co, Cu, Zn, As, Al, Fe, Mg, Mn and P concentrations

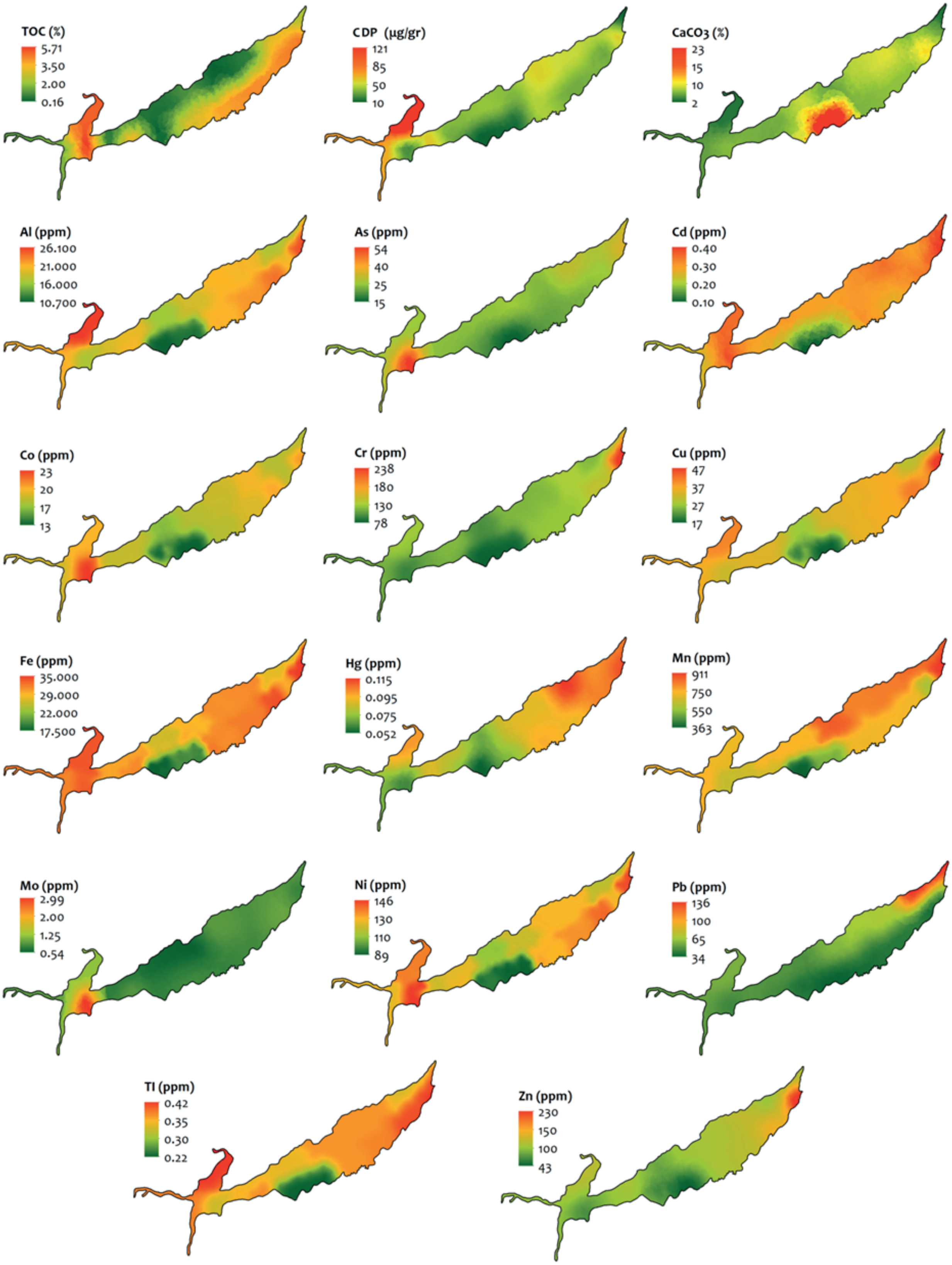

Figure 3

Spatial analysis of TOC, Chl-*a*, CO_3^{-2} and metal content in the sediment were close to the elemental concentrations of the rocks in the basin (Turan, 1999). In this case, it shows that the anthropogenic effect has increased in the last 22 years in terms of Pb and Zn, which now show moderate enrichment (Figure 4).

The mean EF values in the core taken from Poyraz Lagoon were as follows: Cd (1.96) > Mn (1.52) > Mo (1.46) > Cu (1.14) > Hg (1.11) > Pb (1.10) > P (1.06) > Hg (0.79) > Co (0.91) > As (0.88) = Tl (0.88) > Ni (0.86) = Cr (0.86) > Fe (0.82). Accordingly, no enrichment

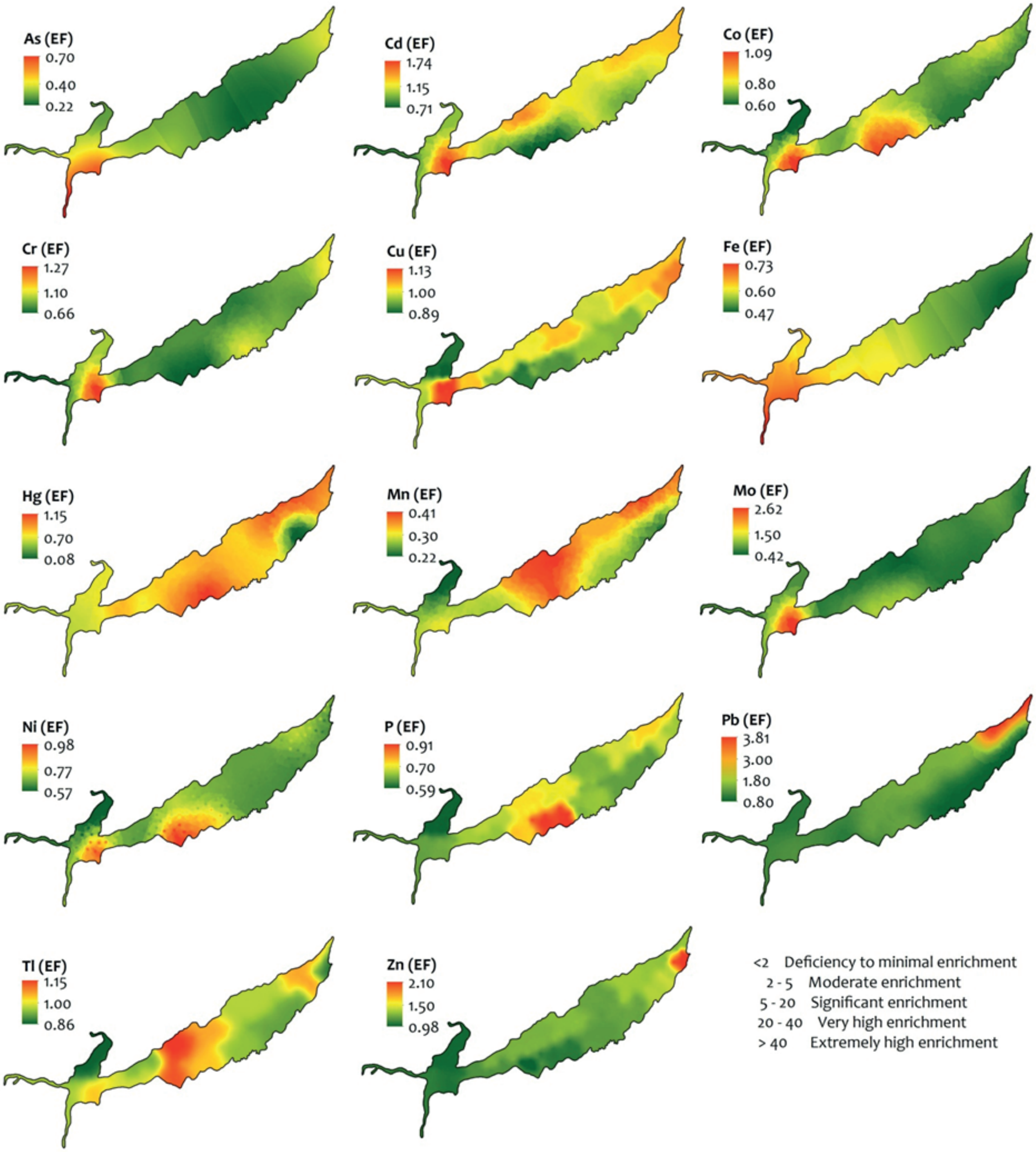

Figure 4

Spatial analysis of EF

of any element was detected. However, some point enrichments in the core were remarkable. Mo was moderately enriched in the base of the core, while Cd, Mn and Mo were moderately enriched in the surface and middle sections.

Moderate enrichment of Mo and Mn may be linked to mining activities, because there are large Mo and Mn mineral deposits and enterprises in the Susurluk basin, where Koca Stream is located (MTA, 2021). Cd enrichment shows a continuous increased from past to present in the core. The probable reason for this increase is the settlement and industrial and agricultural activities in the Susurluk basin. Domestic and industrial waste is among the anthropogenic sources of Cd (Liu et al., 2016). The statistical analysis suggests that Cd, among other nutrients, is of agricultural origin. According to the temporal changes of EF levels, Cd, Cu, Mn and Zn show a tendency to increase today. Zn and Cu are important nutrients used in agriculture, as with Cd. In the multivariate statistical analysis, Cd, Cu and Zn were found to be in the same cluster. In this case, the main source of these elements is likely the agricultural activities carried out in the basin. The enrichment level of As, Co, Cr, Fe, Hg, Mo, Ni and Pb is decreasing from past to present (Figure 5).

3.4. Potential ecological risk (mER and mPER) assessment

Cd (33.72) > Hg (31.74) > Tl (9.93) > Pb (6.65) > Cu (5.00) > Co (3.83) > Ni (3.56) > As (3.11) > Cr (1.71) > Zn (1.28) > Mn (0.30), according to the mean mER values from surface sediment. According to these data, no element was found to create an ecological risk in Dalyan Lagoon. However, the mean mER level of Cd and Hg approached the moderate ecological risk limit. Examination of the spatial distribution of mER helped identify point risk zones. A moderate ecological risk was determined at ST 10 (54) for Cd and at ST 13 (46) for Hg (Figure 6). Cd is an important element used in agriculture. Hg is caused by different anthropogenic activities, especially domestic and industrial waste (Jiang et al., 2019).

The mean mPER in Dalyan Lagoon was 101. Based on this value, there is a low potential ecological risk throughout the lagoon. mPER was highest (132) in MIZ, approaching the level of moderate ecological risk (Figure 6). This situation may be due to the effect of sea intrusion. The percentage of responsibility of the elements for the total ecological risk is as follows: Cd (33.38%) > Hg (31.42%) > Tl (9.83%) > Pb (6.58%)

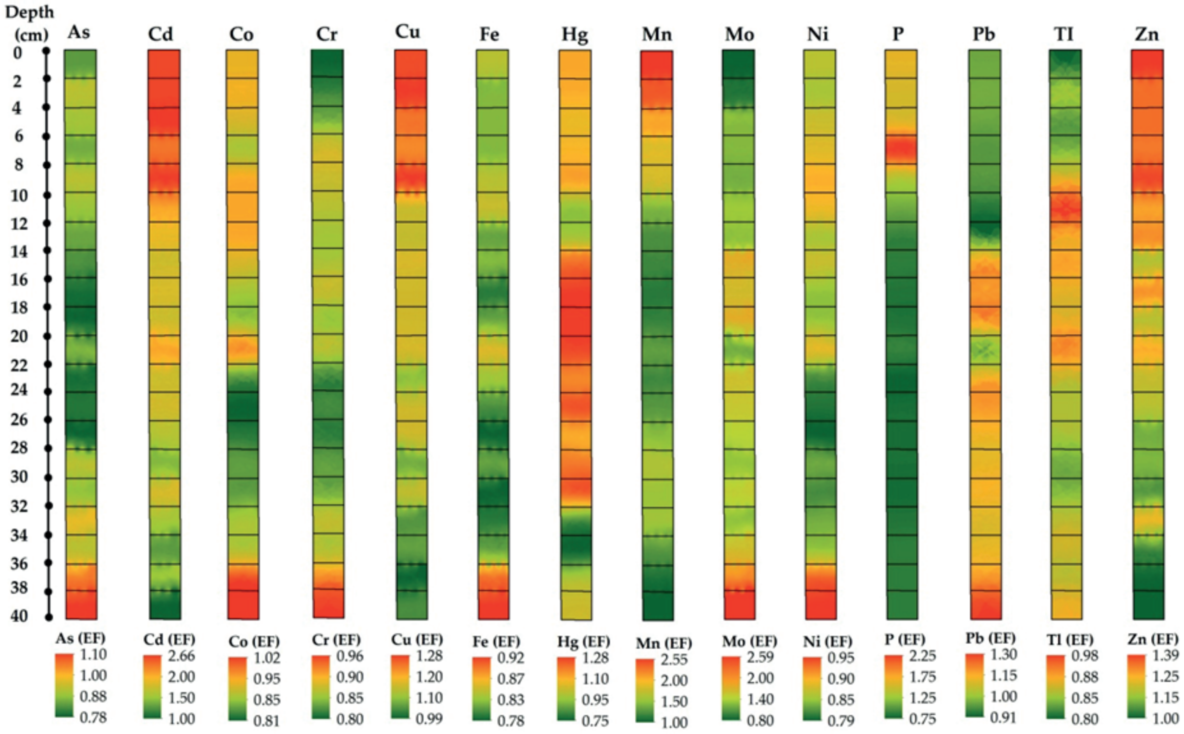

Figure 5

Temporal variation of EF levels of elements in Poyraz Lagoon

> Cu (4.95%) > Co (3.79%) > Ni (3.52%) > As (3.07%) > Cr (1.69%) > Zn (1.26%) > Mn (0.29%). The findings show that 64.80% of the ecological risk was caused by Cd and Hg. These data show that agriculture and domestic and industrial waste are causing ecological pressure in the floodplain.

The mean mER values in the core samples were as follows: Cd (58.92) > Hg (44.20) > As (8.82) > Tl (8.81) > Cu (5.71) > Pb (5.49) > Co (4.54) > Ni (4.29) > As (3.11) > Cr (1.72) > Mn (1.52) > Zn (1.21). According to the average, a moderate ecological risk was determined for Cd and Hg, but no ecological risk was found for other elements (Figure 7). According to the average mPER (145), there is no potential ecological risk in Poyraz Lagoon. However, moderate potential ecological risk was detected in the 0–10 cm, 16–18 cm, 20–22 cm and 30–32 cm slices. It is noteworthy that mPER shows an increasing trend from past to present. The findings reveal that the ecological risk levels of Cd, Cu, Mn and Zn tend to increase. Cd, which is estimated to be caused by agricultural activities, is responsible for 40.63% of the potential ecological risk detected in the Poyraz Lagoon. Hg, likely caused by industrial and domestic waste, is responsible for 30.48% of the potential ecological risk detected in Poyraz Lagoon.

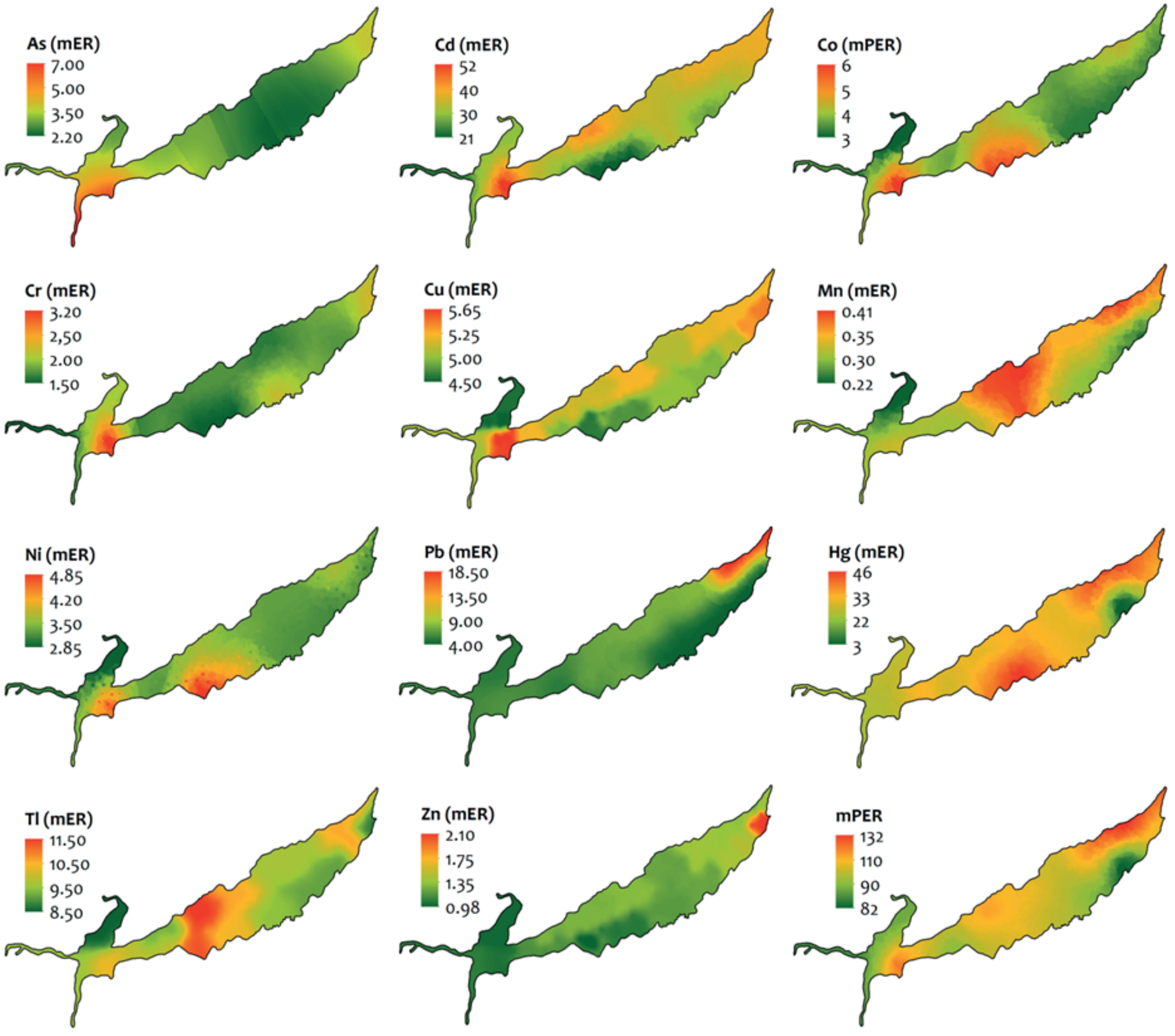

Figure 6

Spatial analysis of mER and mPER

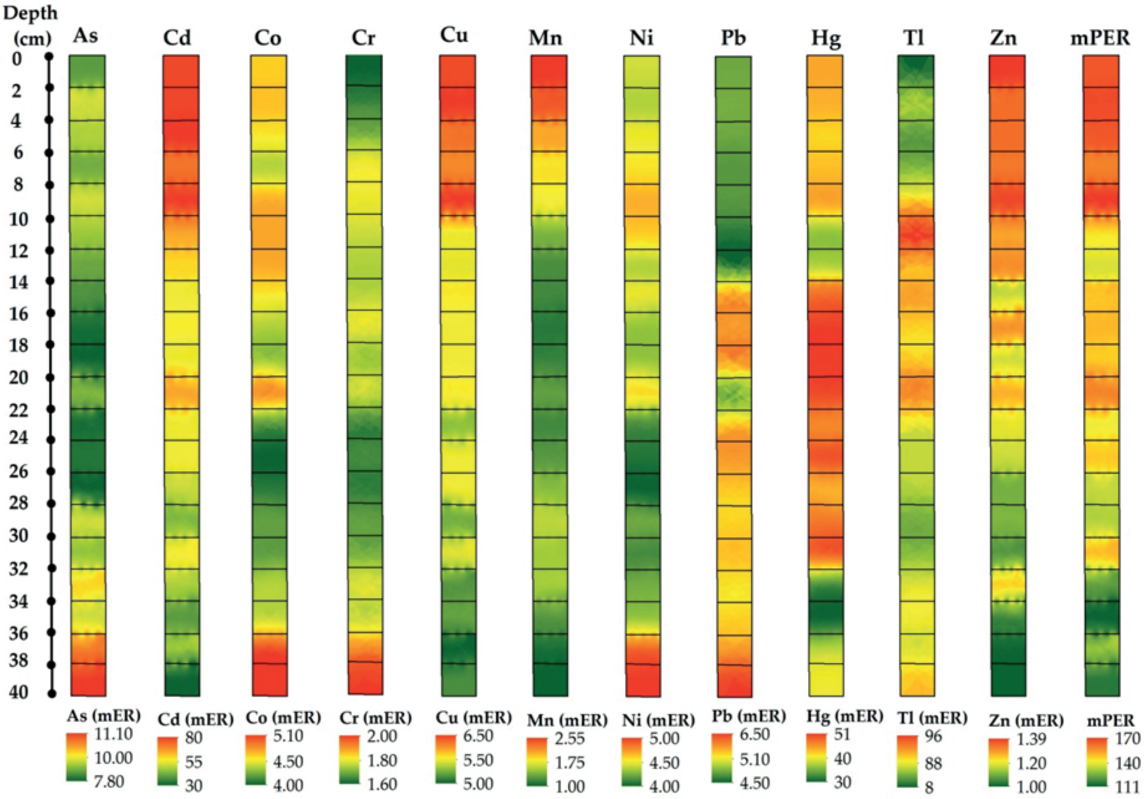

Figure 7

Temporal variation of mER and mPER in Poyraz Lagoon

3.5. Ecological and toxic risk assessment with mHQ, ECI, CSI and TRI

Unlike mPER – which is based on the enrichment level of elements – mHQ, ECI, CSI and TRI results are based on various threshold values obtained from laboratory experiments. The aim of this study is to make a detailed and multifaceted evaluation by using indices working with a combination of two different mechanisms. According to the mean mHQ values, Ni (3.44) > As (2.63) > Cr (2.38) > Pb > (1.50) > Cu (1.16) > Zn > (1.15) > Hg (0.84) > Cd (0.73). According to the average data, very high levels of Ni pollution, high levels of As pollution, significant levels of Cr pollution, low levels of Pb, Cu, Zn pollution and very low levels of Hg and Cd pollution were detected in Dalyan Lagoon. Based on the spatial distribution of mHQ, As pollution was highest (3.74) at ST 10 near the WZ, creating pollution with extreme severity. Cr caused a very high level of pollution (3.35) at ST 18. Ni caused pollution with extreme severity (3.75) at ST 10 near the WZ. Pb caused significant pollution (2.44) in the MIZ (Figure 8). The contamination levels of other elements were found to be at acceptable levels. The spatial analysis showed that high levels of metal contamination were concentrated in the MIZ and the WZ. In a previous study conducted on the coasts of Kocasu Delta, it was determined that Ni, Pb and Cr caused low to moderate pollution. The possible sources of Ni, Pb and Cr were identified as lithological characteristics in the Susurluk Basin and discharge partly from anthropogenic activities (Pehlivan et al., 2021). Ni, As and Cr are predicted to have common, but naturally and anthropogenically complex sources. The cluster analysis and Spearman correlation analysis support this prediction (Figure 8 and Table 6).

The ECI ranged from 2.14 to 3.23, with an average value of 2.74. According to the average value, the level of pollution was uncontaminated to slightly contaminated in Dalyan Lagoon at the time of the study. However, when the spatial analysis is examined, it can be observed that some point inputs increase the pollution level of the lake. ECI, the maximum level of which (3.23) was at ST 18 in the MIZ, indicates a level of slight to moderate contamination at this point (Figure 8). This may be the result of water discharge from the sea.

CSI ranged from 0.88 to 2.62. The average CSI pointed to low to moderate severity of contamination (1.58). Examination of the spatial distribution of

CSI shows that the most polluted point of Dalyan Lagoon was ST 18 (2.62), located in the MIZ. There was moderately severe contamination at ST 18.

TRI was lowest (7.33) at ST 13, located in the SCZ, and highest (15.94) at ST 15, located in the MIZ. In Dalyan Lagoon, the mean TRI was found to be 11.43 (moderate toxic risk). The toxic risk liability ratio of PTEs were as follows: Ni (37%) > Cr (18.63%) > As (17.93%) > Pb (9.53%) > Cu (6.47%) > Zn (5.77%) > Hg (2.79%) > Cd (1.74%). Ni, Cr and As were found to be responsible for 73.56% of the toxic risk. These ratios show that the toxic risk level is affected by natural factors.

The lagoon generally has a moderate toxic risk. However, a significant (15.94) toxic risk was detected at ST 18 (Figure 8). The combination of indices based on element enrichment and those based on toxicological thresholds brought a multifaceted perspective to the research. The findings show that there may be different levels of toxic and ecological risks without enrichment.

3.6. Health risk indices

According to HI mean values, As (0.1425) > Fe (0.0666) > Cr (0.0637) > Al (0.0311) > Pb (0.0238) > Ni (0.0175) > Mn (0.0078) > Cu (0.0013) > Zn (0.0006) > Hg (0.0004) = Cd (0.0004). The HI data showed no non-carcinogenic health risk hazard in Dalyan Lagoon. The riskiest area of the lagoon in terms of HI was ST 10, located in the WZ, due to As (0.2826). However, the

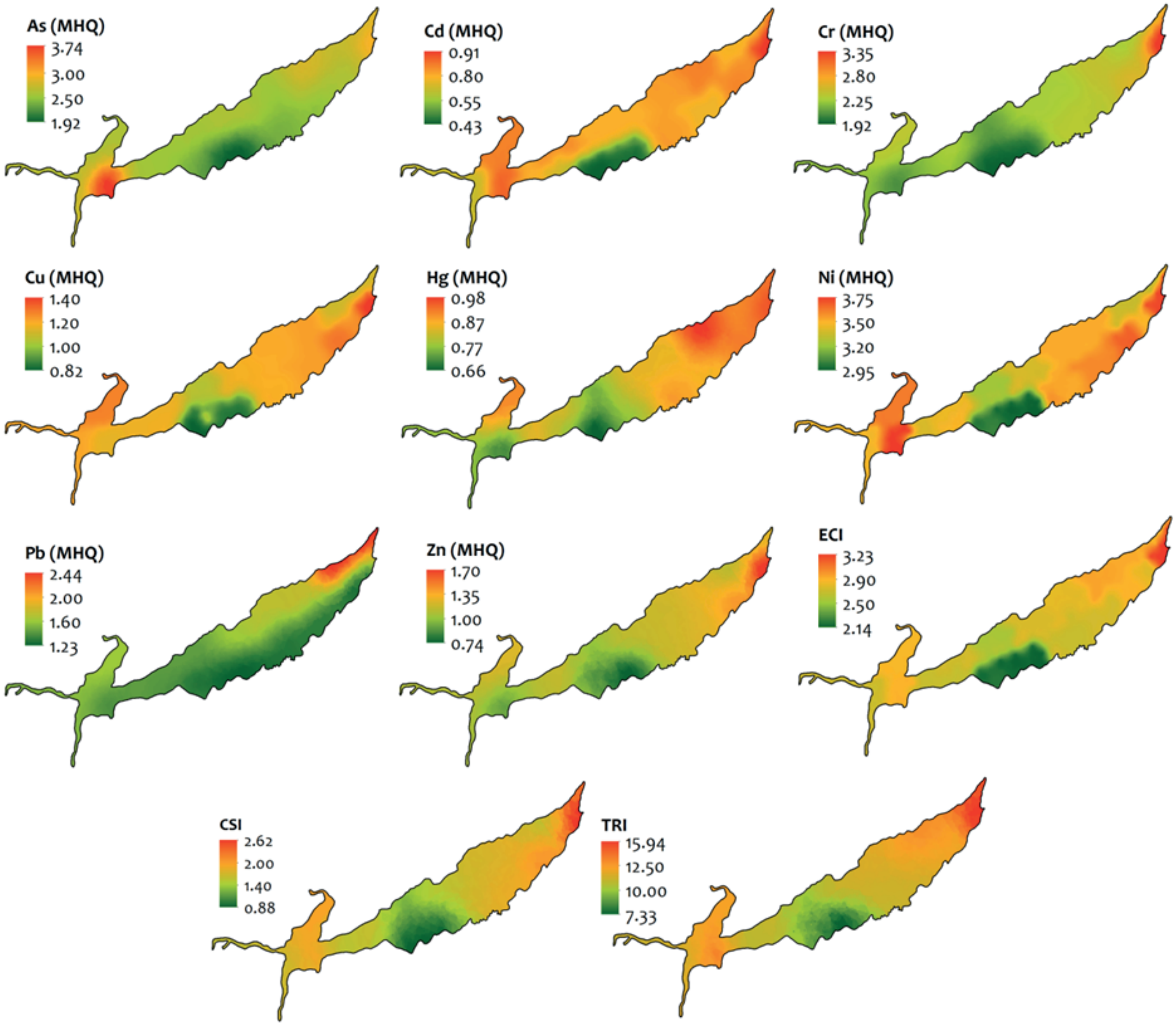

Figure 8

Spatial analysis of MHQ, ECI, CSI and TRI maximum value at this point was found to be about four times lower than the risk level. Table 3 presents the HI data of the lagoon.

Table 3

Distribution of HI according to sampling points

ST.	Cu	Pb	Zn	Ni	Mn	Fe	As	Cd	Cr	Al	Hg
1	0.0011	0.0611	0.0005	0.0165	0.0093	0.0622	0.1504	0.0004	0.0614	0.0268	0.0005
2	0.0014	0.0316	0.0006	0.0180	0.0091	0.0698	0.1635	0.0005	0.0692	0.0328	0.0006
3	0.0013	0.0303	0.0006	0.0180	0.0090	0.0694	0.1301	0.0005	0.0617	0.0331	0.0005
4	0.0013	0.0271	0.0006	0.0169	0.0096	0.0654	0.1390	0.0004	0.0617	0.0301	0.0005
5	0.0010	0.0217	0.0005	0.0154	0.0080	0.0596	0.1332	0.0004	0.0517	0.0254	0.0004
6	0.0013	0.0220	0.0006	0.0173	0.0081	0.0669	0.1306	0.0005	0.0621	0.0313	0.0004
7	0.0013	0.0220	0.0005	0.0169	0.0072	0.0667	0.1332	0.0004	0.0574	0.0304	0.0005
8	0.0014	0.0239	0.0005	0.0177	0.0082	0.0710	0.1437	0.0003	0.0589	0.0338	0.0004
9	0.0015	0.0262	0.0006	0.0193	0.0078	0.0759	0.1384	0.0005	0.0665	0.0417	0.0005
10	0.0012	0.0202	0.0004	0.0207	0.0070	0.0750	0.2826	0.0005	0.0481	0.0265	0.0003
11	0.0013	0.0203	0.0005	0.0176	0.0073	0.0696	0.1301	0.0003	0.0603	0.0331	0.0004
12	0.0006	0.0156	0.0002	0.0134	0.0041	0.0392	0.1107	0.0001	0.0410	0.0168	0.0003
13	0.0007	0.0155	0.0003	0.0128	0.0057	0.0439	0.0742	0.0002	0.0414	0.0182	0.0004
14	0.0013	0.0173	0.0006	0.0181	0.0078	0.0698	0.1139	0.0005	0.0656	0.0335	0.0005
15	0.0014	0.0184	0.0006	0.0189	0.0080	0.0716	0.1228	0.0004	0.0659	0.0354	0.0005
16	0.0015	0.0191	0.0008	0.0198	0.0079	0.0759	0.1358	0.0005	0.0761	0.0381	0.0001
17	0.0014	0.0163	0.0007	0.0181	0.0063	0.0676	0.1431	0.0005	0.0732	0.0328	0.0005
18	0.0018	0.0196	0.0012	0.0205	0.0102	0.0784	0.1891	0.0006	0.1246	0.0409	0.0006

According to LCR data, Pb and Cd did not pose a carcinogenic health risk at any point in Dalyan Lagoon. Ni was determined to cause a carcinogenic health risk at all sampling points; As caused a carcinogenic health risk in the WZ by exceeding the tolerable limit in the sea facing-parts of the SCZ (at ST 2, ST 16, ST 17 and ST 18); and Cr caused a carcinogenic health risk (Table 4). Another study determined that the volcanic rock formations and regional anthropogenic effects in the Susurluk basin, where the Karacabey floodplain is located, increased the Ni and Cr concentrations (Pehlivan et al., 2021). Statistical analysis placed As in the same element group. The reasons for the carcinogenic health risk in Dalyan Lagoon were found to be the anthropogenic activities carried out in the basin and the lithological characteristics of the basin.

3.7. Multivariate statistical analysis

Four components with an eigenvalue of > 1 were calculated in the principal component analysis (PCA). These four components were responsible for 84.8% of the total changes. PC 1 represented 47.92% of the total variance and consisted of negatively charged Cu, Zn, Ni, Co, Cd, Mn, Tl, Hg and P. PC 2 represented 17.50% of the total variance and consisted of positively charged Mo, Co, As, TOC and negatively charged Hg and P. PC 3 accounted for 11.25% of the total variance and consisted of positively charged Pb, Mn and negatively charged Al and TOC. PC 4 represented 8.09% of the variance and consisted of positively charged Fe and CDP and negatively charged Cr, TOC and CO$_3^{-2}$. PTEs of natural and anthropogenic origin were combined in all components. According to the PCA, Co, Hg and P in PC 1 and PC 2; Mn in PC 1 and PC 3; and TOC in PC 2, PC 3 and PC 4 had close values (Table 5). The complexity of PTEs within clusters may indicate multiple sources.

Table 4

Distribution of LCR by sampling points

Station	Pb	Ni	As	Cd	Cr
1	9.E-06	2.E-04	7.E-05	6.E-06	9.E-05
2	5.E-06	2.E-04	7.E-05	8.E-06	1.E-04
3	4.E-06	2.E-04	6.E-05	7.E--06	9.E-05
4	4.E-06	2.E-04	6.E-05	6.E-06	9.E-05
5	3.E-06	2.E-04	6.E-05	6.E-06	8.E-05
6	3.E-06	2.E-04	6.E-05	7.E-06	9.E-05
7	3.E-06	2.E-04	6.E-05	6.E-06	9.E-05
8	4.E-06	2.E-04	6.E-05	5.E-06	9.E-05
9	4.E-06	2.E-04	6.E-05	8.E-06	1.E-04
10	3.E-06	2.E-04	1.E-04	8.E-06	7.E-05
11	3.E-06	2.E-04	6.E-05	5.E-06	9.E-05
12	2.E-06	1.E-04	5.E-05	2.E-06	6.E-05
13	2.E-06	1.E-04	3.E-05	2.E-06	6.E-05
14	3.E-06	2.E-04	5.E-05	7.E-06	1.E-04
15	3.E-06	2.E-04	6.E-05	6.E--06	1.E-04
16	3.E-06	2.E-04	6.E-05	7.E-06	1.E-04
17	2.E-06	2.E-04	6.E-05	7.E-06	1.E-04
18	3.E-06	2.E-04	9.E-05	9.E-06	2.E-04

The fact that the Karacabey floodplain is open to the effects of rivers and seas and that many types of anthropogenic activities continue in the river basin strengthens this possibility.

Table 5

Principal component analysis

PTEs	Comp. 1	Comp. 2	Comp. 3	Comp. 4
Mo	-0.042	**0.542**	0.047	-0.090
Cu	**-0.331**	-0.037	-0.119	0.026
Pb	-0.061	-0.081	**0.456**	0.282
Zn	**-0.297**	-0.185	-0.087	-0.245
Ni	**-0.307**	0.213	-0.102	0.006
Co	**-0.289**	0.271	0.009	0.041
Mn	**-0.258**	-0.176	**0.236**	0.098
Fe	-0.061	0.330	-0.366	**0.460**
As	-0.167	**0.391**	0.246	-0.231
Cd	**-0.312**	0.080	0.092	-0.089
Cr	-0.279	-0.198	-0.056	**-0.310**
Al	0.0834	-0.079	**-0.568**	0.221
Tl	**-0.308**	-0.054	-0.138	0.165
Hg	**-0.252**	**-0.253**	-0.109	0.031
P	**-0.293**	**-0.246**	-0.117	-0.047
CO_3^{-2}	0.188	-0.075	-0.217	**-0.347**
Chl-*a*	-0.185	-0.049	0.127	**0.448**
TOC	-0.146	**0.249**	-0.248	**-0.260**

A high positive correlation was found between Mn and Pb, which was found to be moderately enriched by anthropogenic effects at some sampling points in Dalyan Lagoon. It is estimated that these two elements have lithological and anthropogenic origins. A strong positive correlation was detected between Zn, which was moderately enriched in local areas in Dalyan Lagoon, and Ni, Co, Mn, Cd, Cr, Tl, Hg and P. In this cluster, Ni showed a very high level of contamination, causing carcinogenic health risks; Cd created a moderate ecological risk in local areas of Dalyan Lagoon; and there was significant Cr contamination, creating a carcinogenic health risk at some sampling points. There is a strong positive correlation between Hg, which created a moderate ecological risk in local areas due to its high toxic effect, and Cu, Zn, Mn, Cd, Cr and Tl. Other than Tl, these elements caused different levels of ecological and health risks. A high positive correlation was found between highly contaminated As and Mo and between Ni and Co. In this correlation set, PTEs other than Co created different levels of ecological and health risks. Dalyan Lagoon is open to the influence of Kocasu Stream and the Marmara Sea (Figure 2). Therefore, PTE resources are multiple and complex (Table 6).

According to cluster analysis, Cu, Tl, Zn, Cr, P, Ni, Co, Hg and Cd formed a cluster with high similarity. Among these elements, Zn was moderately enriched, Cr was at a significant level, Ni was highly contaminated and Cd and Hg had moderate ecological risk. No risk factors were identified for Tl and Co. Mo and As appeared to be closely related. On the other hand, CDP, TOC, Fe, Pb, Al and CO_3^{-2} formed a separate cluster more closely related to each other. Cluster analysis data showed the complexity of the elements' sources.

4. Conclusion

The following basic results were obtained in the investigation of the element accumulations in the core taken from Poyraz Lagoon and the surface sediments

Table 6

Spearman correlation analysis

	Mo	Cu	Pb	Zn	Ni	Co	Mn	Fe	As	Cd	Cr	Al	Tl	Hg	P	TOC	Chl-a	CO_3^{-2}
Mo																		
Cu	0.03																	
Pb	-0.02	0.04																
Zn	-0.18	**0.89**	0.00															
Ni	**0.46**	**0.88**	0.00	**0.69**														
Co	**0.57**	**0.80**	0.12	**0.56**	**0.95**													
Mn	-0.18	**0.69**	**0.51**	**0.67**	**0.52**	**0.57**												
Fe	**0.52**	0.22	-0.14	-0.15	0.45	0.47	-0.12											
As	**0.79**	0.38	0.12	0.25	**0.66**	**0.77**	0.27	0.16										
Cd	0.27	**0.86**	0.15	**0.74**	**0.87**	**0.86**	**0.69**	0.14	**0.64**									
Cr	-0.19	**0.83**	0.01	**0.99**	**0.63**	0.49	**0.62**	-0.24	0.24	**0.69**								
Al	-0.23	-0.08	-0.33	-0.13	-0.12	-0.22	-0.29	**0.44**	**-0.49**	-0.39	-0.18							
Tl	-0.03	**0.95**	0.06	**0.78**	**0.80**	**0.71**	**0.64**	0.29	0.25	**0.77**	**0.69**	-0.04						
Hg	-0.30	**0.74**	0.30	**0.77**	0.50	0.41	**0.64**	0.03	0.00	**0.63**	**0.74**	0.04	**0.69**					
P	-0.31	**0.89**	0.12	**0.92**	**0.62**	**0.53**	**0.76**	-0.03	0.08	**0.72**	**0.87**	-0.03	**0.86**	**0.87**				
TOC	**0.50**	0.40	-0.14	-0.32	**0.57**	0.48	-0.01	0.36	0.35	0.34	0.36	-0.09	0.36	0.22	0.27			
Chl-*a*	-0.05	**0.53**	0.26	0.34	0.40	0.39	0.43	0.25	0.04	0.41	0.31	**-0.34**	**0.58**	0.40	**0.43**	0.09		
CO_3^{-2}	-0.08	**-0.52**	-0.25	0.35	**-0.57**	**-0.53**	-0.43	-0.14	-0.37	**-0.49**	-0.29	**0.26**	**-0.56**	-0.16	-0.29	-0.01	**-0.50**	

of Dalyan Lagoon, located in the Karacabey floodplain, which hosts a high degree of biodiversity:

- According to EF values, anthropogenic accumulation was not detected for any element. However, there are point enrichments for Pb and Zn. There is a risk of enrichment for Cd.
- The part of the lagoon that is in contact with the sea – the MIZ and the WZ, which is connected to the neighbouring Poyraz Lagoon – stand out as the points where the element accumulations in the lagoon are intense. It is thought that the Marmara Sea and Poyraz Lagoon, with a stronger connection with the sea, have important effects on element accumulations in the floodplain.
- It was determined that the agricultural areas along the southern coasts of the lagoon also play a role in element accumulation. The use of fertilisers in agricultural areas should be controlled. Precautions should be taken for fertiliser residues that mix into rivers and lakes through irrigation waters.
- Although the element accumulations detected by EF and the mER and mPER indices detecting enrichment did not indicate an accumulation of anthropogenic origin and a potential ecological risk, the mHQ index working with the threshold values obtained from eco-toxicological tests pointed to very high levels of Ni pollution, high levels of As pollution and significant levels of Cr pollution. This important difference between the results is due to the working principle of the indices. This shows that making calculations in ecological risk studies by using indices with different advantages can yield more comprehensive results.
- While no element-based non-carcinogenic risk was detected according to the health risk indices, a carcinogenic risk was determined for Ni, Cr and As.
- It was determined that the human impact on the accumulation of Pb and Zn has risen since previous studies were conducted in the region, as mentioned in the Discussion section.
- The core sediments studied to gain an understanding of the temporal trends of the elements showed that the ecological risk levels of Cd, Cu, Mn and Zn tended to increase over time.

The study hypothesised that human-induced element accumulations would be detected in Dalyan Lagoon, which has agricultural areas around it and is affected by the sea. However, the results showed that human accumulation was limited, although natural element concentrations were at levels that suggest a certain risk. It should be noted that water discharge from the sea has a significant effect on the enrichment of these elements. All ecological risk indices used in this study are calculated based on the total element concentration. It is thought that determining element fractions in future studies in the region will make important contributions to the understanding of the bio-availability of these elements.

Statements & Declarations

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Competing interests

The authors have no relevant financial or non-financial interests to disclose.

Availability of data and materials

The datasets generated during and/or analysed during the current study are available from the corresponding author on reasonable request.

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