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***Research Article***

# A Research on Sources of Heavy Metal and Organic Matter Pollution of Recent Sediments in Northwestern Marmara Sea and Precautions to be Taken

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**Abstract**

Tekirdağ and Istanbul cities are residential areas that are situated in the northwestern shores of Marmara Sea. Both cities constitute an important part of Türkiye in terms of population density, agriculture, industry, and marine transportation. Because of the mentioned activities, polluting materials reach the Marmara Sea in the area either directly or indirectly through rivers, thus polluting the Marmara Sea. This study investigates the sources of heavy metal and organic matter pollution in northwestern Marmara aquatic sediments and which precautions should be taken. Sediment samples were taken through "gravity core" technique in the study, and element analysis and organic matter value of these samples were obtained. Arsenic (As) was found to be high generally in all locations in the study. Zinc (Zn) is very high in locations 1-7, 14, and 20. Upon examining the geographic map of the area, these locations which are high in pollution are either next to the residential areas or are discharge areas of polluted rivers. It is interesting that other elements are also higher than normal in these locations. The fact thar As and Zn are found more in aquatic environments sheds light on the irresponsible and excessive agricultural spraying and fertilizer use. In addition, the impact of industrial activities is also visible for this pollution. Location number 12 where Cobalt (Co) levels are very high is around Fevzipaşa residential area, and it can be seen that a river flows into the sea at this point. It can be seen in these analyses that one of the main reasons of pollution in aquatic sediments are the rivers discharging to the sea from the land. It can be seen in the study areas that the organic carbon percentage (%TOC) has high levels particularly in the discharge points of rivers into the sea. The fact that the TOC level is high shows that pollution is an inland plant-based pollution. Nitrogen levels, however, are relatively low (%N: 0.03-0.09) in locations where TOC levels are high. It is thought that eutrophication might be low in areas where nitrogen (N) levels are below 20 g/kg. high carbon concentrations in the sediments, however, show that biological break down of carbon matters is slow.
In order to prevent the pollutions found here, contents of the fertilizers and sprays used in agricultural activities should be analysed. Furthermore, analyses should be made on agricultural lands before the fertilization and spraying. Industrial waste should be discharged to the sea after it is refined in advanced treatment facilities, and ships should be prevented from discharging their waste in the sea in maritime shipping.

**Keywords:** Soil, Pollution, TOC, OC, IC, Marmara Sea, Marmara

# Kuzeybatı Marmara Denizi'ndeki Sedimentlerin Ağır Metal ve Organik Madde Kirliliğinin Kaynakları ve Alınması Gereken Önlemler Üzerine Bir Araştırma

**Öz**

Tekirdağ ve İstanbul şehirleri, Marmara Denizi'nin kuzeybatı kıyılarında yer alan yerleşim alanlarıdır. Her iki şehir de nüfus yoğunluğu, tarım, sanayi ve deniz ulaşımı açısından Türkiye'nin önemli bir bölümünü oluşturmaktadır. Söz konusu faaliyetler nedeniyle bölgedeki kirletici maddeler doğrudan veya dolaylı olarak akarsular aracılığıyla Marmara Denizi'ne ulaşmakta ve böylece Marmara Denizi'ni kirletmektedir. Bu çalışmada kuzeybatı Marmara'daki sucul sedimanlarda ağır metal ve organik madde kirliliğinin kaynakları ve hangi önlemlerin alınması gerektiği araştırılmaktadır. Çalışmada "gravity core" tekniği ile sediment örnekleri alınmış ve bu örneklerin element analizi ve organik madde değerleri elde edilmiştir. Araştırmada genel olarak tüm lokasyonlarda arsenik (As)

yüksek bulunmuştur. Çinko (Zn) 1-7, 14 ve 20 numaralı lokasyonlarda oldukça yüksektir. Bölgenin coğrafi haritası incelendiğinde kirliliğin yüksek olduğu bu lokasyonların ya yerleşim yerlerinin yakınında ya da kirlenmiş nehirlerin deşarj alanları olduğu görülmektedir. Bu lokasyonlarda diğer unsurların da normalden yüksek olması ilginçtir. As ve Zn'nin sucul ortamlarda daha fazla bulunması, sorumsuz ve aşırı tarımsal ilaçlama ve gübre kullanımına ışık tutmaktadır. Ayrıca bu kirlilikte endüstriyel faaliyetlerin etkisi de görülmektedir. Kobalt (Co) seviyelerinin oldukça yüksek olduğu 12 numaralı lokasyon Fevzipaşa yerleşim alanı civarında olup, bu noktada bir nehrin denize aktığı görülmektedir. Bu analizlerde, sucul sedimentlerdeki kirliliğin ana nedenlerinden birinin karadan denize boşalan nehirler olduğu görülmektedir. Çalışma alanlarında organik karbon yüzdesinin (%TOC) özellikle akarsuların denize döküldüğü noktalarda yüksek düzeylerde olduğu görülmektedir. TOC düzeyinin yüksek olması kirliliğin karasal bitki kaynaklı bir kirlilik olduğunu göstermektedir. Bununla birlikte, TOC seviyelerinin yüksek olduğu yerlerde nitrojen seviyeleri nispeten düşüktür (%N: 0,03-0,09). Azot (N) düzeyinin 20 g/kg'ın altında olduğu bölgelerde ötrofikasyonun düşük olabileceği düşünülmektedir. Ancak çökeltilerdeki yüksek karbon konsantrasyonları, karbon maddelerinin biyolojik parçalanmasının yavaş olduğunu göstermektedir.
Burada bulunan kirliliklerin önlenmesi için tarımsal faaliyetlerde kullanılan gübre ve spreylerin içeriklerinin analiz edilmesi gerekmektedir. Ayrıca gübreleme ve ilaçlama öncesinde tarım arazilerinde analizlerin yapılması gerekmektedir. Endüstriyel atıklar ileri arıtma tesislerinde arıtıldıktan sonra denize deşarj edilmeli, deniz taşımacılığında gemilerin atıklarını denize boşaltması engellenmelidir.

**Anahtar Kelimeler:** Toprak, Kirlilik, TOK, OK, IK, Marmara Denizi

# 1. Introduction

The high level of organic matter and inorganic matter concentrations seen in Marmara Sea is a serious pollution problem. The fact that Marmara Sea is an inland sea and there are abundant residential areas around it make it vulnerable to both domestic and industrial pollution (Kam and Yümün, 2021; Pekey et al., 2004). High levels of heavy metal and organic matter concentrations in sea water are the major indicators of pollution. Among the natural sources of heavy metals are earth debris carried by the wind, volcanic eruptions, forest fires, biogenic processes, and sea salt (Muhammed et al., 2011; Blaser et al., 2000). Anthropogenic sources of heavy metals, however, are solid, liquid, and gas waste released from industrial facilities, mining activities, and agricultural activities (Paramasivam et al., 2015; Kam and Önce, 2016; Önce et al., 2021; Dinçer et al., 2019; Sarkar et al., 2018; Srivastava et al., 2018). All heavy metals show a toxic effect above a certain concentration (Alcorlo et al., 2006; Egemen, 2006; Yümün et.al.2016; Bayrak et. al. 2022; Yümün et.al. 2022). Thus, Pb, Zn, Cu, Cr, Cd, and some other metals are considered to be metals that have the potential to show human originated toxic effect in environmental risk evaluations (Ma et al., 2016; Valdes et al., 2014). It is highly important to effectively observe the heavy metal concentrations in aquatic environments (Ahmed et al., 2015; Kahlon et al., 2018).

Since deep sediments in aquatic environments are heavy metal storages, they are used to determine the heavy metal pollution (Yümün, 2016; Yümün, 2017; Yümün and Önce, 2017; Dinçer et al., 2019; Önce et al., 2021; Yümün et.al.2023). Among its most important reasons is that the accumulation in the sediment does not show momentary change. This change occurs based on long processes. Furthermore, this situation represents both the natural and the anthropogenic accumulation. The fact that heavy metals occur as neatly accumulated layers in sediments enables to compare the change between the current situation and past terms. Sediments are reservoirs where elements are stored in aquatic environments, and they form a living and feeding environment for many living beings. Therefore, pollutions that occurs in a sediment can be dangerous for living beings and the environment (Türkmen and Akbulut, 2015; Kam et al., 2021). Aquatic sediments serve as a potential storage; they are separated from the water surrounding it; and they have the potential to store more metals than the water because of the long-term accumulation.

Even the 1% of the contaminants that enter aquatic environments remain in the water; almost all of it collapses and is stored in the sediment (Salomons and Stigliani, 1995). This situation makes the sediment a secondary source that pollutes water (Meng et al., 2016; Zhao et al., 2013). In addition to heavy metals, organic pollution is also an important factor that threatens aquatic environments. Organic matter is found in fine-grained clay or chemical rocks (shale, clay stone, and limestone) in aquatic environments.

Organic matters accumulated in the clay can remain so without degradation. Accumulation of organic matters is closely related to the accumulation processes of heavy metals (Türkmen et al., 2006; Sungur and Özcan, 2015; Zaynab et al., 2022). Organic pollution becomes particularly intense in areas which accommodate domestic population. Detergents, pesticides, proteins, and phenols compose the organic components in domestic wastewater composition. When domestic wastewater flows into the aquatic environment without being refining the high carbon, phosphorus, and nitrogen contents, organic pollution occurs (Metcalf, 2003; Alaş and Çil, 2002). TOC/N ratio determines whether organic matter is aquatic-based or terrestrial-based. Terrestrial organic matter is rich in carbon than nitrogen and shows a high TOC/N ratio. Terrestrial plants are rich in cellulose, and algae contain rich components in terms of nitrogen. TOC/N ratio of terrestrial organics is between 12 and 14. Planktonic with TOC/N ratio between 6 and 8 shows aquatic-based organic matter. TOC/N = 10 value is a characteristic of mixed areas between aquatic and terrestrial sources (Lallier-Verges et al., 1998; Azevedo, 2003; Dinçer et al., 2019). Organic matter gives important information particularly for eutrophication (Bayram et al., 2011; Yümün and Önce Nişancıoğlu, 2023). TOC values are quite changeable; 1 mg / L in subterranean water and sea water, 2-10 mg / L in lakes or river water, and up to 10 g / L in swamps can be seen (Cwqgs, 1996). Organic carbon content in lower surface sediments depends on lithology of the sediment, microbiological degradation level, productivity of water column, and contribution of the environment. Many researchers carried out studies on the subject before. Yümün (2017) investigated the heavy metal concentration and their ratios in the aquatic sediments of west Marmara Sea. As a result of these studies, he developed a correlation coefficient described as Pollution Index (PI) to determine heavy metal results. Thanks to this coefficient (PI), elements such as Pb, Hg, Cu, and Ni were found out to get richer because of anthropogenic sources. Yümün and Önce (2017) separated the area into pollution zones by investigating the heavy metal concentrations in Edremit Bay aquatic sediments. They used foraminiferal as bioindicators in their study to determine the harm that pollutions leaved on the environment. Yümün et al. (2021) investigated the existence of toxic and radioactive elements in Gemlik Bay aquatic sediments in their study. According to the analysis results, they have determined radioactive elements such as Cs-137 that occurs in artificial ways. Moreover, a richness in toxic elements as a result of agricultural and industrial activities was also determined. Dinçer et al. (2019) carried out TOC, TN, TC, and IC analyses in Edremit Bay aquatic sediments in their study. It was determined that organic pollution has occurred in the Bay where agriculture is made intensely. Atmaca and Erdem (2016) determined soil properties in certain river beds in Tekirdağ city in their study. In his study, Sarı (2008) determined the heavy metal sources in the drainage basin of rivers (Biga, Gönen, and Kocasu and its branches) in the south of Marmara Sea and their potential pollution effect through organic carbon and heavy metal analysis of surface sediments (including the selective assimilation). As a result of the study, it was seen that organic matter value is high in samples taken from areas where population is dense.

This study also investigates the heavy metal and organic matter pollution in aquatic sediments in west Marmara (Tekirdağ-İstanbul). Toxic materials that were analysed are Al, As, Cd, Hg, Cr, Cu, Fe, Pb, Mn, Ni, and Zn. In order to determine the organic matter pollution, TOC, TN, TC, and IC analyses and calculations were made in the sediment. As a result of the analyses, it was seen that there is pollution in areas that are risky in terms of water quality.

# 2. Material and Method

## 2.1. Sample Collection

A total of 20 sediment samples were collected from the study area using the gravity coring method. These samples were obtained at an average distance of 1.5-2 km from the coastline and at an approximate water depth of 40-50 meters. The samples were stored in containers to prevent contact with air and were maintained at temperatures below 20°C until analysis (Chen et al., 2022; Yümün et al., 2021). The sampling locations included discharge points of rivers flowing into the Marmara Sea, areas receiving domestic sewage, tourism zones, and regions with a high concentration of industrial facilities. The geographical location of the study area and sample points are illustrated in Figure 1, and the sample coordinates are provided in Table 1.

*Table 1: Sample coordinates of the study area*

| Sample Location | X | Y |
|---|---|---|
| **TK-1** | 543318.49 d D | 4535338.03 m K |
| **TK-2** | 544357.00 d D | 4535921.00 m K |
| **TK-3** | 546021.00 d D | 4535981.00 m K |
| **TK-4** | 547922.00 d D | 4536350.00 m K |
| **TK-5** | 549813.00 d D | 4536638.00 m K |
| **TK-6** | 552210.00 d D | 4537373.00 m K |
| **TK-7** | 555755.00 d D | 4537973.00 m K |
| **TK-8** | 559286.00 d D | 4538290.00 m K |
| **TK-9** | 565984.00 d D | 4538188.00 m K |
| **TK-10** | 570584.00 d D | 4533672.00 m K |
| **TK-11** | 578029.00 d D | 4533258.00 m K |
| **TK-12** | 585386.00 d D | 4541034.00 m K |
| **TK-13** | 590526.00 d D | 4544513.00 m K |
| **TK-14** | 595881.00 d D | 4545990.00 m K |
| **TK-15** | 602825.41 d D | 4545761.19 m K |
| **TK-16** | 608082.12 d D | 4544428.65 m K |
| **TK-17** | 611381.91 d D | 4544291.05 m K |
| **TK-18** | 616390.54 d D | 4543057.22 m K |
| **TK-19** | 624144.19 d D | 4538755.11 m K |
| **TK-20** | 631453.98 d D | 4538541.55 m K |


*Figure 1. Location map of study area and samples*

## 2.2. Laboratory Analyses

### *2.2.1 ICP-OES Analysis*

Approximately 200 grams of samples were taken from the sediment samples for heavy metal analysis. The collected samples were placed in aluminium containers and left to dry in an oven at 50°C for 4 hours. The dried samples were crushed with a mortar and sifted through a 250 micron sieve to separate large particles. The remaining samples were placed in sealed bags and transported to the laboratory for analysis. Prior to analysis, solid samples needed to be made into a solution. Therefore, 12 ml HNO3 and 4 ml HCl were added to the samples placed in combustion tubes for the melting process, and they were burned for 1 hour at 98 oC and 1.5 hours at 200 oC, and readings were taken (Yümün et. al. 2019, Yümün and Kam 2019, Khan et al. 2022).

### *2.2.2. Organic Matter Analyses*

Total Organic Carbon (TOC) consists of the sum of the concentration of all organic carbon atoms covalently bonded to the organic molecules of the water column (Doğanay, 2014). It includes all organic substances, both natural and artificial. Practically, Total Organic Carbon is calculated with the following formula (Fil et. al. 2018).

Total Organic Carbon = Total Carbon – Total Inorganic Carbon

Approximately 250 g of sediment samples were taken for analysis. It was brought to the laboratory environment in protected bags and analyzed. Analyzes were performed with the TOC-L series (Model SSM 5000A) device (Dinçer et. al. 2019).

Nitrogen measurement was made in the laboratory with a Vapodest VAP 20s model device. In TOC measurement, both total carbon (TC) and inorganic carbon (IC) are calculated. The total amount of organic carbon is calculated by subtracting the amount of inorganic carbon from the total carbon content.

# 3. Results and Discussion

## 3.1. Results of Elemental Analysis and Findings

In order to determine the degree of permanent pollution in sediment samples taken from the sea, elemental analysis and organic matter analyses were carried out in the laboratory with the ICP-OES device. Toxic element analysis results are given in Table 2, and organic matter analysis results are given in Table 3. The concentration values obtained from the analyses were also visualised graphically in order to make them more visual and facilitate their correlation (Figs. 2 and 3).

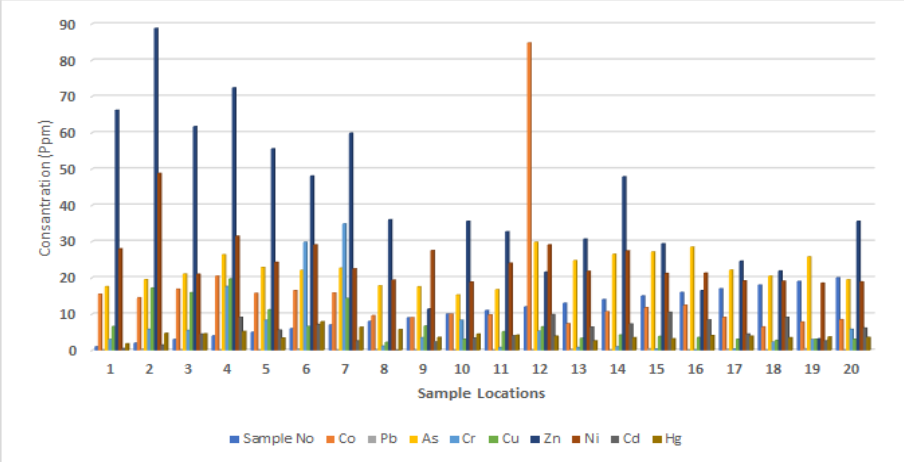


*Figure 2. First Group Elements (Sample Location 1(TK-1))*

*Table 2. Sediment toxic element analysis results of the study area (mg/L)*

| | | | First Group Elements | | | | | | | Second Group Elements | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **SampleNo** | **Co** | **Pb** | **As** | **Cr** | **Cu** | **Zn** | **Ni** | **Cd** | **Hg** | **SampleNo** | **Fe** | **Al** | **Mn** |
| **TK-1** | 15.50 | 0.00 | 17.58 | 3.00 | 6.60 | 66.20 | 28.00 | 0.50 | 1.80 | **TK-1** | 24287.5 | 15409.8 | 590.2 |
| **TK-2** | 14.50 | 0.25 | 19.51 | 5.80 | 17.20 | 88.80 | 48.80 | 1.50 | 4.70 | **TK-2** | 73048.3 | 16369.6 | 169.7 |
| **TK-3** | 16.87 | 0.10 | 21.06 | 5.50 | 15.90 | 61.70 | 21.00 | 4.52 | 4.60 | **TK-3** | 30528.5 | 14632.1 | 561.5 |
| **TK-4** | 20.48 | 0.00 | 26.40 | 17.60 | 19.70 | 72.40 | 31.50 | 9.13 | 5.20 | **TK-4** | 35554.9 | 17375.5 | 685.5 |
| **TK-5** | 15.74 | 0.00 | 22.86 | 8.30 | 11.20 | 55.60 | 24.30 | 5.60 | 3.40 | **TK-5** | 27361.4 | 13992 | 330.7 |
| **TK-6** | 16.48 | 0.37 | 22.06 | 29.80 | 6.60 | 48.10 | 29.10 | 7.13 | 7.90 | **TK-6** | 24840.8 | 12098 | 332.9 |
| **TK-7** | 15.83 | 0.00 | 22.65 | 34.90 | 14.30 | 59.90 | 22.50 | 2.60 | 6.40 | **TK-7** | 29538 | 15088.1 | 361.1 |
| **TK-8** | 9.55 | 0.15 | 17.80 | 1.20 | 2.20 | 36.00 | 19.40 | 0.00 | 5.70 | **TK-8** | 20859.2 | 13240.3 | 281.5 |
| **TK-9** | 9.04 | 0.12 | 17.52 | 3.50 | 6.70 | 11.40 | 27.50 | 2.32 | 3.60 | **TK-9** | 14881 | 91316.4 | 386 |
| **TK-10** | 10.01 | 0,05 | 15.26 | 8.30 | 3.07 | 35.60 | 18.82 | 3.43 | 4.50 | **TK-10** | 11893.4 | 65555.8 | 49.1 |
| **TK-11** | 9.78 | 0.00 | 16.74 | 0.80 | 5.17 | 32.70 | 24.04 | 4.06 | 4.20 | **TK-11** | 15615.8 | 11396.8 | 81.5 |
| **TK-12** | 84.75 | 0.10 | 29.80 | 5.30 | 6.52 | 21.60 | 29.09 | 9.86 | 3.90 | **TK-12** | 18978.9 | 13817.9 | 99.5 |
| **TK-13** | 7.34 | 0.23 | 24.76 | 0.80 | 3.32 | 30.70 | 21.83 | 6.45 | 2.60 | **TK-13** | 14611.9 | 19735.8 | 46.4 |
| **TK-14** | 10.65 | 0.00 | 26.49 | 1.00 | 4.24 | 47.90 | 27.41 | 7.26 | 3.50 | **TK-14** | 15732.1 | 11102.1 | 78.1 |
| **TK-15** | 11.79 | 0.45 | 27.15 | 0.40 | 3.89 | 29.40 | 21.21 | 10.50 | 3.20 | **TK-15** | 14216.6 | 18894.2 | 47.7 |
| **TK-16** | 12.46 | 0.00 | 28.47 | 0.10 | 3.56 | 16.50 | 21.29 | 8.40 | 4.10 | **TK-16** | 13652.3 | 19256.1 | 55.6 |
| **TK-17** | 9.12 | 0.24 | 22.14 | 0.40 | 3.06 | 24.60 | 19.18 | 4.52 | 3.90 | **TK-17** | 13003.6 | 17631.4 | 57.9 |
| **TK-18** | 6.46 | 0.00 | 20.47 | 2.30 | 2.77 | 21.90 | 19.08 | 9.13 | 3.50 | **TK-18** | 12329 | 17349.5 | 54.1 |
| **TK-19** | 7.83 | 0.45 | 25.77 | 3.00 | 3.09 | 3.20 | 18.54 | 2.60 | 3.70 | **TK-19** | 12825 | 17482.8 | 52.2 |
| **TK-20** | 8.46 | 0.00 | 19.46 | 5.80 | 3.07 | 35.60 | 18.89 | 6.13 | 3.60 | **TK-20** | 45698.01 | 11545.6 | 47.2 |

Since the concentration values of Co, Pb, As, Cr, Cu, Zn, Ni, Cd, and Hg elements are less than 1000 ppm, these elements are considered first-group elements and are presented in the graph in figure 2. Since Fe, Al, and Mn concentration values are greater than 1000 ppm, it was decided to evaluate these elements differently from other elements. Figure 2 shows that As is generally high in all locations. It is seen that Zn is very high in locations 1–7, 14, and 20. Looking at the map in Figure 1, it can be seen that these locations are either adjacent to residential areas or places where polluted streams discharge into the sea. It is also noteworthy that other elements are very high in these locations. Location 12, where Co values are very high, is around the Fevzipaşa settlement, where a stream flows into the sea.

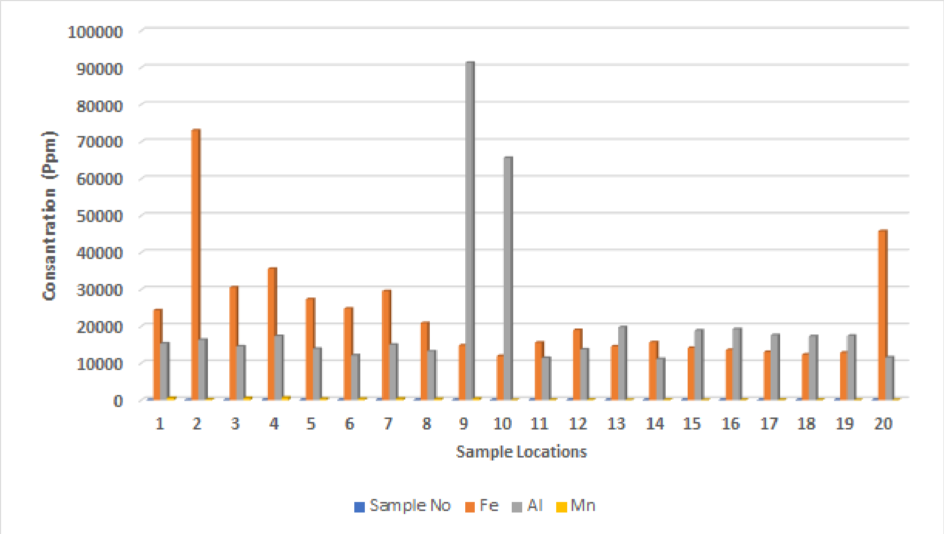


*Figure 3. Second Group Elements*

## 3.2. The Results of Organic Matter Analysis and Findings

Marine sediment analyses were carried out to examine land-based organic matter pollution in the marine environment (Table 3). It has been evaluated that the pollution effect of streams that are discharging into the sea, seen in elemental analyses, can also be seen in organic matter analyses. It is seen that the organic carbon percentages (%TOC) obtained in sediment samples no. 2, 4, 9, 12–19 are greater than 1%. These values again show that the percentage of organic carbon (%TOC) is high, especially at the discharge points of the rivers into the sea. The high TOC value indicates that the pollution is of intra-continental vegetal origin. The low nitrogen values (%N: 0.03-0.09) in these locations indicate that the pollution is caused by organic matter produced by algae. Irizuki et al. (Irizuki et al. (2015) emphasized that eutrophication may be less in regions where the TOC value is below 20 g/kg. High carbon concentrations in sediment indicated that biodegradation of carbonaceous substances was slow. In most locations, the TOC/TN ratio was found to be greater than 4. These values are a sign that intra- continental pollution is active. The low total nitrogen value indicates high biological activity on the seabed.

*Table 3. Sediment organic matter analysis results of the study area*

| Sample No | N% | TC % | %IC | TOC % | TOC/TN |
|---|---|---|---|---|---|
| **TK-1** | 0.07 | 1.82 | 0.9 | 0.94 | 13.428 |
| **TK-2** | 0.09 | 1.80 | 0.74 | 1.07 | 11.89 |
| **TK-3** | 0.05 | 1.86 | 1.11 | 0.78 | 15.6 |
| **TK-4** | 0.13 | 1.74 | 0.48 | 1.21 | 9.31 |
| **TK-5** | 0.07 | 1.72 | 1.01 | 0.74 | 10.58 |
| **TK-6** | 0.03 | 1.65 | 1.32 | 0.12 | 4.0 |
| **TK-7** | 0.02 | 0.49 | 0.01 | 0.4 | 20.0 |
| **TK-8** | 0.06 | 0.98 | 0.3 | 0.42 | 7.0 |
| **TK-9** | 0.06 | 1.20 | 0.11 | 1.71 | 28.5 |
| **TK-10** | 0.07 | 1.56 | 0.86 | 0.9 | 12.8 |
| **TK-11** | 0.05 | 1.46 | 0.46 | 0.5 | 10.0 |
| **TK-12** | 0.03 | 1.34 | 0.54 | 1.5 | 50.0 |
| **TK-13** | 0.07 | 1.56 | 0.89 | 1.7 | 24.2 |
| **TK-14** | 0.09 | 1.40 | 0.64 | 1.6 | 17.8 |
| **TK-15** | 0.04 | 1.65 | 0.89 | 1.9 | 47.5 |
| **TK-16** | 0.06 | 1.02 | 0.67 | 1.45 | 24.17 |
| **TK-17** | 0.04 | 1.34 | 0.66 | 1.78 | 44.5 |
| **TK-18** | 0.03 | 1.98 | 0.34 | 1.65 | 55.0 |
| **TK-19** | 0.08 | 1.24 | 0.98 | 1.67 | 20.88 |
| **TK-20** | 0.09 | 1.64 | 0.44 | 1.031 | 11.4 |

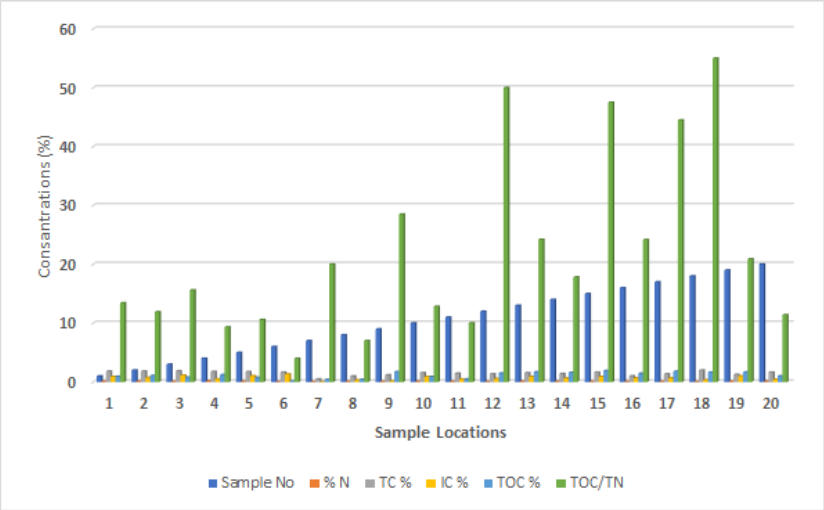


*Figure 4. Organic Matter Concentrations (%)*

## 3.3. Methods to Evaluate Heavy Metal Pollution in Marine Environments

### *3.3.1. Enrichment factor*

Enrichment factor (EF) is one of the indexes used to detect the rate of anthropogenic heavy metal pollution in soil, seas, lakes and streams, and aquatic ecosystem sediments. This index is calculated by using various normalization elements (Fe, Al, Li, etc.) and the background values given in previous studies. Calculation of the normalized enrichment factor is widely used to determine anthropogenic effects (Abrahim & Parker, 2008; Barbieri et. al., 2016). The conservative (reference) element used to normalize metals in the calculations is defined as the element with very low chemical mobility, characterized by its concentration remaining unchanged in soil or marine sediment, the absence of vertical mobility and/or degradation (Barbieri et. al., 2016). There is no definitive rule regarding the selection of the reference element. The concentration of the reference element in the sample is considered to be of crustal (lithogenic) origin. The purpose of using EF values instead of concentrations of elements is to normalize the value of the reference element by eliminating the effects of large differences in grain size, carbonate dilution and mineral content (Bruland et. al., 1974; Herut & Sandler, 2006; Park & Presley, 1997).

In this study, Fe was used as the normalization element when calculating the enrichment factor. Iron element was considered as the reference element because it does not show large variations in the core. In many studies similar to this study, Fe was used as a conservative element in the same way (Gargouri et. al., 2011; Shafie et. al., 2013). Average shale metal concentrations are taken as basis in evaluating the metal contents of marine sediments (Algan et.al. 2004; Pekey et. al., 2004; Aksu et. al., 1997; Yümün and Kam 2019). Normalization according to the iron element gives better results in regions with high mafic and ultramafic input in the area, such as Gemlik Bay, Albania and Iskenderun Bay. Average shale metal concentrations are taken as the basis in evaluating the metal contents of marine sediments (Abrahim & Parker 2008; Aksu et. al., 1997; Algan et.al. 2004). Therefore, when interpreting the sediment heavy metal analysis results, the data obtained for each metal were compared with the shale average values. In this study, the enrichment factor (EF) of metals (Zn, As, Co, Cu, Ni, Pb and Mn) was calculated using heavy metal analysis results. Enrichment Factor is calculated using the following equation (1);

**EF= (Cn/Cref)/(Bn/Bref)** (1)

Description of this formula; EF: Enrichment factor, Cn: Metal value measured in the study, Cref: Value of the reference element measured in the study, Bn: Background value of the measured element, Bref: Represents the shell (background) value of the reference element. Heavy metal contents of some geological reference rocks are given in Table 4.

It is generally accepted that if the calculated EF value result is close to 1 (EF<1), it indicates shell origin, between 1 and 3, it indicates low enrichment, between 3 and 5, it is controversially evidence of shell origin (high enrichment), and at values greater than EF>5, it indicates no shell origin (over-enrichment) (Galuszka, et al. 2014, Halstead, et al., 2000). In the calculation and evaluation of this factor, the difference compared to other pollution indexes is the normalization by selecting the reference element. The most striking result in the enrichment factor analysis is that the Arsenic Enrichment Factor ($EF_{As}$) is very high in all locations. It is thought that this result is due to intensive agricultural activities in the region.

*Table 4. Heavy metal contents of some geological reference rocks [159, 160].*

| | Fe % | Mn ppm | Co ppm | Ni ppm | Cu ppm | Zn ppm | Pb ppm | As ppm |
|---|---|---|---|---|---|---|---|---|
| Earth crust[154] | 5 | 950 | 25 | 75 | 55 | 70 | 13 | 1,8 |
| Shale [155,156] | 4,7 | 850 | 19 | 70 | 45 | 95 | 20 | 13 |
| Sandstone [155] | 0,9 | 50 | <1 | 2 | 5 | 16 | 7 | 1 |
| Limestone [155, 157] | 0,4 | 1100 | <1 | 20 | 4 | 20 | 9 | 1 |
| Ultrabasics [158] | 5,7 | 700-2600 | 75-101 | 1700-2900 | 46-62 | | | |
| Basalt [156,157] | 8,6 | 1500 | 48 | | | | | |

*Table 5. Enrichment Factors of sediment samples taken from the study area*

| **EF** | **COR-1** | **COR-2** | **COR-3** | **COR-4** | **COR-5** | **COR-6** | **COR-7** | **COR-8** | **COR-9** | **COR-10** | **COR-11** |
|---|---|---|---|---|---|---|---|---|---|---|---|
| $EF_{Zn}$ | 1,94 | 0,86 | 1,44 | 1,45 | 1,45 | 1,38 | 1,44 | 1,23 | 0,55 | 2,14 | 1,50 |
| $EF_{Mn}$ | 1,28 | 0,122 | 0,97 | 1,01 | 0,63 | 0,7 | 0,64 | 0,71 | 1,37 | 0,22 | 0,27 |
| $EF_{As}$ | 20,1 | 7,41 | 19,2 | 20,6 | 23,2 | 24,6 | 21,2 | 23,69 | 32,7 | 35,64 | 29,78 |
| $EF_{Co}$ | 1,27 | 0,39 | 1,1 | 1,15 | 1,15 | 1,32 | 1,08 | 0,92 | 1,21 | 1,69 | 1,25 |
| $EF_{Cu}$ | 0,25 | 0,21 | 0,47 | 0,5 | 0,37 | 0,24 | 0,44 | 0,10 | 0,41 | 0,23 | 0,30 |
| $EF_{Ni}$ | 0,77 | 0,44 | 0,46 | 0,59 | 0,59 | 0,78 | 0,5 | 0,62 | 1,23 | 1,05 | 1,03 |

| **EF** | **COR-12** | **COR-13** | **COR-14** | **COR-15** | **COR-16** | **COR-17** | **COR- 18** | **COR-19** | **COR-20** |
|---|---|---|---|---|---|---|---|---|---|
| $EF_{Zn}$ | 0,81 | 1,50 | 2,17 | 1,48 | 0,86 | 1,35 | 1,27 | 0,17 | 0,56 |
| $EF_{Mn}$ | 0,28 | 0,17 | 0,26 | 0,18 | 0,21 | 0,23 | 0,23 | 0,21 | 0,054 |
| $EF_{As}$ | 43,60 | 47,06 | 46,76 | 53,06 | 57,93 | 47,29 | 46,12 | 55,7 | 11,8 |
| $EF_{Co}$ | 0,89 | 1,0 | 1,35 | 1,66 | 1,83 | 1,40 | 1,05 | 1,22 | 0,37 |
| $EF_{Cu}$ | 0,31 | 0,21 | 0,25 | 0,25 | 0,24 | 0,21 | 0,20 | 0,22 | 0,06 |
| $EF_{Ni}$ | 1,01 | 1,0 | 1,16 | 0,99 | 1,04 | 0,98 | 1,03 | 0,96 | 0,27 |

### 3.3.2. Contamination Factor ($Cfi$) (Hakanson 1980)

Contamination factor (Cf) is a method that is most frequently used in studies investigating heavy metals in sediment and provides important data about the current situation. The pollution factor classification used in interpreting the calculation results is given in Table 6. The average values of some elements used as background values are given in Table 7.

$$C_f^{\,i} = C^{\,i} / C_n^{\,i} \qquad (2)$$

C i : metal value measured in sediment

: is the pre-industrial reference value of the metal.

The contamination factor ($Cfi$) of the element results of the sediment samples taken from the study area was calculated (Table 6). In Table 6, it is understood that the Arsenic

*Table 6. Contamination Factor Classification (Hakanson, 1980)*

| $C_f$ | **Sediment Quality** |
|---|---|
| **$C_f$ <1** | Low contamination |
| **1 < $C_f$ < 3** | Medium contamination |
| **3 < $C_f$ < 6** | Significant contamination |
| **$C_f$>6** | Very high contamination |

Contamination Factor (**$C_f$ As: 7,2-14,7**) is very high in sample locations 1-18 and clearly indicates contamination. It is understood that the Contamination Factor for other elements in all locations indicates low contamination (Cf <1). This situation is also seen in the Enrichment Factor.

*Table 7. World average values of some elements used as background values*

| **Toxic Elements** | **Zn** | **As** | **Fe** | **Cd** | **Co** | **Cr** | **Cu** | **Ni** | **Pb** | **Mn** |
|---|---|---|---|---|---|---|---|---|---|---|
| | **ppm** | **ppm** | **Ppm** | **ppm** | **ppm** | **ppm** | **ppm** | **ppm** | **ppm** | **ppm** |
| **Earth Crust [154]** | 70 | 1,8 | 5000 | - | 25 | - | 55 | 75 | 13 | 950 |
| ***Shale [155, 156]** | 95 | 13 | 47000 | 0,3 | 19 | 90 | 45 | 70 | 20 | 850 |
| **Sandstone [155]** | 16 | 1 | 900 | - | <1 | - | 5 | 2 | 7 | 50 |
| **Limestone [155, 157]** | 20 | 1 | 400 | - | <1 | - | 4 | 20 | 9 | 1100 |
| **Ultrabasics [158]** | - | - | 5700 | - | 75-101 | - | 46-62 | 1700-2900 | - | 700-2600 |
| **Basalt [156, 157]** | - | - | 8600 | - | 48 | - | - | - | - | 1500 |

*Table 8. Contamination factor of elemental analysis results of sediments taken from the study area ($C_{fi}$)*

| **Sample Location** | **$C_f$ Mn** | **$C_f$ Fe** | **$C_f$ Ni** | **$C_f$ Cu** | **$C_f$ Zn** | **$C_f$ Co** | **$C_f$ As** |
|---|---|---|---|---|---|---|---|
| **TK-1** | 0,62 | 0,49 | 0,37 | 0,12 | 0,94 | 0,62 | 9,8 |
| **TK-2** | 0,18 | 1,5 | 0,65 | 0,3 | 1,3 | 0,58 | 10,8 |
| **TK-3** | 0,59 | 0,60 | 0,28 | 0,29 | 0,88 | 0,67 | 11,7 |
| **TK-4** | 0,72 | 0,71 | 0,42 | 0,36 | 1,0 | 0,82 | 14,7 |
| **TK-5** | 0,35 | 0,55 | 0,32 | 0,2 | 0,79 | 0,63 | 12,7 |
| **TK-6** | 0,35 | 0,5 | 0,39 | 0,12 | 0,69 | 0,66 | 12,3 |
| **TK-7** | 0,38 | 0,59 | 0,3 | 0,26 | 0,86 | 0,63 | 12,6 |
| **TK-8** | 0,29 | 0,42 | 0,25 | 0,04 | 0,51 | 0,38 | 9,9 |
| **TK-9** | 0,4 | 0,29 | 0,37 | 0,12 | 0,16 | 0,36 | 9,7 |
| **TK-10** | 0,05 | 0,24 | 0,25 | 0,06 | 0,51 | 0,4 | 8,5 |
| **TK-11** | 0,09 | 0,3 | 0,32 | 0,09 | 0,47 | 0,39 | 9,3 |
| **TK-12** | 0,1 | 0,38 | 0,39 | 0,12 | 0,3 | 0,34 | 7,2 |
| **TK-13** | 0,05 | 0,29 | 0,29 | 0,06 | 0,44 | 0,29 | 13,75 |
| **TK-14** | 0,08 | 0,31 | 0,37 | 0,01 | 0,68 | 0,29 | 14,7 |
| **TK-15** | 0,05 | 0,28 | 0,28 | 0,07 | 0,42 | 0,47 | 15,1 |
| **TK-16** | 0,06 | 0,27 | 0,28 | 0,06 | 0,24 | 0,5 | 15,8 |
| **TK-17** | 0,06 | 0,26 | 0,26 | 0,06 | 0,35 | 0,36 | 12,3 |
| **TK-18** | 0,056 | 0,25 | 0,25 | 0,05 | 0,31 | 0,26 | 11,37 |
| **TK-19** | 0,055 | 0,26 | 0,25 | 0,06 | 0,05 | 0,31 | 0,05 |
| **TK-20** | 0,05 | 0,91 | 0,25 | 0,06 | 0,5 | 0,34 | 0,8 |

#### ***3.3.3. Pollution Index (PI (Yümün, 2017))***

A correlation was developed by Yümün (2017) to determine the pollution status of marine sediments and to ensure their interpretability. The Pollution Index (PI) value is obtained by dividing the sum of the ratios of heavy metal measurement values to the average value by the number of measurements. The empirical value obtained as Pollution Index (PI) was calculated with the following equation (Table 9).

$$PI = [(MV_1/MV_{avg}) + (MV_2/MV_{avg}) + \ldots\ldots + (MV_n/MV_{avg})] / n \quad (3)$$

Descriptions of the parameters used in the correlation are given below and the ranges given in Table 10 are suggested for the evaluation of the obtained parameters.

**PI:** Pollution Index

**$MV_1$**: Heavy Metal Measurement Value (ppm)

**$MV_{avg}$:** Average Heavy Metal Measurement Value (ppm)

**n:** Number of heavy metals measured

This correlation provides a more realistic approach compared to other pollution analyses, as it uses local data as "*background values*". For this purpose, the geochemical analysis results obtained from all previous studies were compiled and their average values were taken. The class ranges given in Table 9 were used in the pollution analyses.

*Table 9. Pollution index calculations for samples*

| Sample No | Al | As | Co | Pb | Cd | Cr | Cu | Zn | Ni | Mn | Hg | Fe | Pollution Indeks (PI) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| **TK-1** | 15409.80 | 17.58 | 15.50 | 0.00 | 0.50 | 3.00 | 6.60 | 66.20 | 28.00 | 590.20 | 1.80 | 24287.50 | 0.82 |
| **TK-2** | **16369.60** | **19.51** | **14.50** | **0.25** | **1.50** | **5.80** | **17.20** | **88.80** | **48.80** | **169.70** | **4.70** | **73048.30** | **1.14** |
| **TK-3** | 14632.10 | 21.06 | 16.87 | 0.10 | 4.52 | 5.50 | 15.90 | 61.70 | 21.00 | 561.50 | 4.60 | 30528.50 | 1.00 |
| **TK-4** | **17375.50** | **26.40** | **20.48** | **0.00** | **9.13** | **17.60** | **19.70** | **72.40** | **31.50** | **685.50** | **5.20** | **35554.90** | **1.33** |
| **TK-5** | 13992.00 | 22.86 | 15.74 | 0.00 | 5.60 | 8.30 | 11.20 | 55.60 | 24.30 | 330.70 | 3.40 | 27361.40 | 0.87 |
| **TK-6** | 12098.00 | 22.06 | 16.48 | 0.37 | 7.13 | 29.80 | 6.60 | 48.10 | 29.10 | 332.90 | 7.90 | 24840.80 | 1.03 |
| **TK-7** | 15088.10 | 22.65 | 15.83 | 0.00 | 2.60 | 34.90 | 14.30 | 59.90 | 22.50 | 361.10 | 6.40 | 29538.00 | 1.07 |
| **TK-8** | 13240.30 | 17.80 | 9.55 | 0.15 | 0.00 | 1.20 | 2.20 | 36.00 | 19.40 | 281.50 | 5.70 | 20859.20 | 0.53 |
| **TK-9** | 91316.40 | 17.52 | 9.04 | 0.12 | 2.32 | 3.50 | 6.70 | 11.40 | 27.50 | 386.00 | 3.60 | 14881.00 | 0.91 |
| **TK-10** | 65555.80 | 15.26 | 10.01 | 0.05 | 3.43 | 8.30 | 3.07 | 35.60 | 18.82 | 49.10 | 4.50 | 11893.40 | 0.69 |
| **TK-11** | 11396.80 | 16.74 | 9.78 | 0.00 | 4.06 | 0.80 | 5.17 | 32.70 | 24.04 | 81.50 | 4.20 | 15615.80 | 0.49 |
| **TK-12** | **13817.90** | **29.80** | **84.75** | **0.10** | **9.86** | **5.30** | **6.52** | **21.60** | **29.09** | **99.50** | **3.90** | **18978.90** | **1.07** |
| **TK-13** | 19735.80 | 24.76 | 7.34 | 0.23 | 6.45 | 0.80 | 3.32 | 30.70 | 21.83 | 46.40 | 2.60 | 14611.90 | 0.56 |
| **TK-14** | 11102.10 | 26.49 | 10.65 | 0.00 | 7.26 | 1.00 | 4.24 | 47.90 | 27.41 | 78.10 | 3.50 | 15732.10 | 0.60 |
| **TK-15** | 18894.20 | 27.15 | 11.79 | 0.45 | 10.50 | 0.40 | 3.89 | 29.40 | 21.21 | 47.70 | 3.20 | 14216.60 | 0.68 |
| **TK-16** | 19256.10 | 28.47 | 12.46 | 0.00 | 8.40 | 0.10 | 3.56 | 16.50 | 21.29 | 55.60 | 4.10 | 13652.30 | 0.55 |
| **TK-17** | 17631.40 | 22.14 | 9.12 | 0.24 | 4.52 | 0.40 | 3.06 | 24.60 | 19.18 | 57.90 | 3.90 | 13003.60 | 0.50 |
| **TK-18** | 17349.50 | 20.47 | 6.46 | 0.00 | 9.13 | 2.30 | 2.77 | 21.90 | 19.08 | 54.10 | 3.50 | 12329.00 | 0.50 |
| **TK-19** | 17482.80 | 25.77 | 7.83 | 0.45 | 2.60 | 3.00 | 3.09 | 3.20 | 18.54 | 52.20 | 3.70 | 12825.00 | 0.49 |
| **TK-20** | 11545.60 | 19.46 | 8.46 | 0.00 | 6.13 | 5.80 | 3.07 | 35.60 | 18.89 | 47.20 | 3.60 | 45698.01 | 0.61 |
| **Average Value Of Marmara Sea** | 17067.90 | 23.44 | 34.60 | 6.90 | 38.20 | 126.46 | 15.69 | 81.11 | 107.95 | 404.45 | 1191.10 | 29833.36 | 3.84 |
| **Main Value** | 21445.60 | 22.26 | 16.54 | 0.45 | 6.85 | 12.58 | 7.52 | 41.95 | 28.54 | 227.28 | 60.72 | 23775.69 | 0.92 |

Table 10. Recommended Pollution Index Ranges (Yümün 2017).

| Parameter Range | Evaluation |
|---|---|
| **PI>1** | Polluted Environment |
| **PI<1** | Clean Environment |
| **PI=1** | Critical Environment |

# 4. Conclusions and Recommendations

Tekirdağ and Istanbul are important regions of Turkey in terms of population density, agriculture, industrialization and maritime transportation. Uncontrolled discharge of both domestic waste and agricultural and industrial waste in the region can cause pollution of rivers in the terrestrial area. The Marmara Sea is also affected by these pollutions directly and/or indirectly (due to the discharge of polluted rivers). Pollutions are divided into two classes: 1. Pollution of sea water, 2. Pollution of bottom sediments. It was considered as temporary pollution because sea water can be renewed due to waves and water currents. However, the pollution in bottom sediments is permanent and cannot be cleaned. For this reason, even if the water in the sea is clear from time to time, it does not mean that the sea is clean. For this reason, it was decided to conduct pollution analysis of sediments in the part of the Marmara Sea between Tekirdağ and Istanbul.

In the study, the origin of heavy metal and organic matter pollution in current marine sediments in the study area and the precautions to be taken were investigated. When the sediments in the study area were evaluated with the Enrichment Factor, Arsenic (As) was calculated to be higher than 5 in all locations, which is an indicator of anthropogenic pollution. In addition, the Enrichment Factor values of Zn and Co heavy metals are 1 and above 1. Values between 1 and 3 are accepted as the beginning of pollution. It is understood that the abundance of Zn and As in the marine environment is a result of agricultural pesticide and fertilizer use and industrial activities in the region. Looking at the pollution index calculations, it was seen that the index was 1 and above in locations 2, 4, 6, 7 and 12. An index of 1 or above is defined as a dirty area. Likewise, when the Contamination Factor calculations are examined, all locations except locations 19 and 20 were determined to have very high contamination. The highest values were determined at locations TK-15 and TK-16 (16 and 15,8).  Location 12, where cobalt (Co) values are very high, is around the Fevzipaşa settlement and it is seen that the stream flows into the sea at this point. All sediment evaluation factors calculated in the study gave common results, which shows us the accuracy of the calculations and analyzes made.

The percentage of organic carbon (%TOC) is high at the points where rivers discharge into the sea. The high TOC value in the study indicates that the pollution is of intra-continental vegetal origin. However, Nitrogen concentrations are low in these locations (%N: 0.03-0.09). It shows that eutrophication may be low in regions where nitrogen (N) values are below 20 g/kg. High carbon concentrations in the sediment showed that carbonaceous substances biodegraded slowly.

In order to prevent the pollution detected here, it is recommended to control the fertilizers and pesticides used in agricultural activities. Industrial wastes should be discharged into the sea after being treated in advanced treatment facilities, and ship wastes should be prevented from being discharged into the sea during maritime transportation. These measures are vital for the protection of the Marmara Sea and a sustainable environmental policy.

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