

Heavy metal pollution in the Black Sea shore and offshore of Turkey

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Abstract Land-locked seas were polluted mainly by land-based pollutants. The Black Sea is the largest enclosed sea in the world and widely perceived to be heavily polluted. In order to determine the impact of marine activities on heavy metal pollution, shore (500 m distance from edge)–offshore (5,555 m distance from edge) samples of various stations through the Sinop, Samsun and Ordu cities located in the Middle Black Sea region between May 2000 and October 2001 were taken. In addition, samples were taken between April–May 2000 in order to designate the heavy metal pollution of certain rivers, streams, harbor and shores in Samsun’s boundary and 32 sampling stations for the routine pollution monitoring studies are selected including rivers, streams, industrial and domestic discharge points along the Black Sea coast of Turkey in the year 1996. Heavy metal concentrations of whole samples were measured and compared with “Quality Criteria of General Marine and Continental Inside Water Sources” currently effective in Turkey so as to bring up the levels of pollution in marine, rivers and streams.

Keywords The Black Sea · Heavy metals · Pollution monitoring · Water quality

Introduction

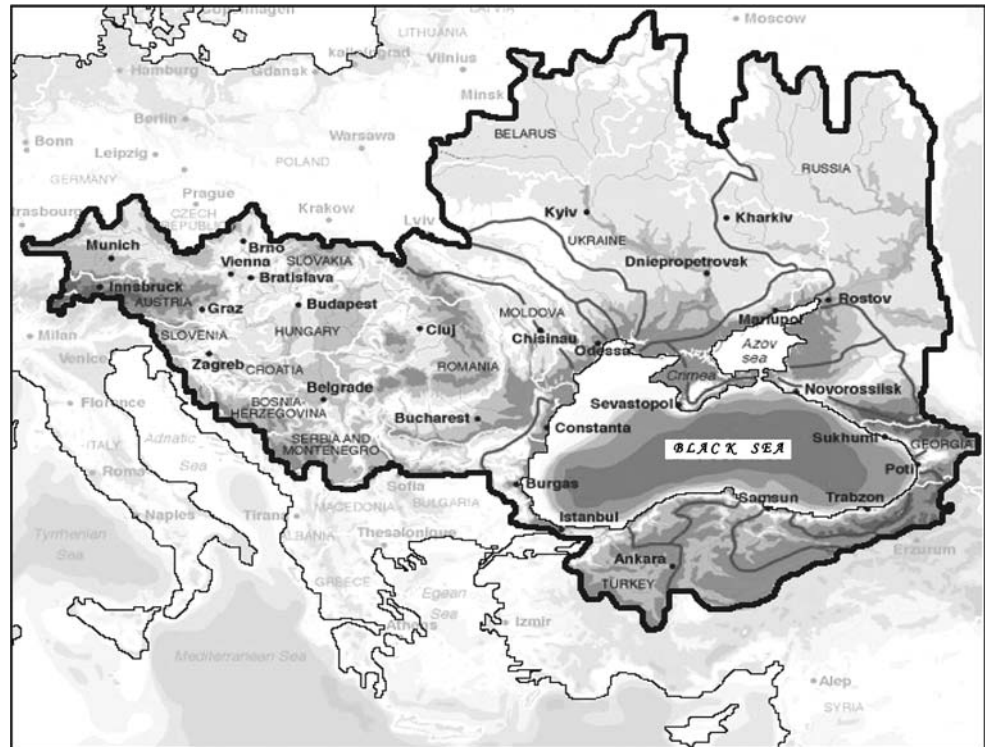
The Black Sea is a large semi-enclosed European sea (4.2×10^5 km²). It is deep (maximum depth 2,212 m) and is virtually isolated from other seas (Readman et al. 2002). The Black Sea is surrounded by six countries located in Europe and Asia: Bulgaria, Georgia, Romania, Russia, Turkey and Ukraine (Fig. 1). In fact, the Black Sea is influenced by 17 countries, 13 capital cities and some 160 million people. The distinguishing feature of the Black Sea is the shallow (150–200 m deep), oxic and biologically active top layer. The remaining 90% of its 5.37 km³ volume is anoxic because of the lack of water exchange (Tuncer et al. 1998; Readman et al. 2002). Thus, it contains the world’s largest anoxic water mass.

The main sources of pollution in the Black Sea are the rivers which flow into the region. The major rivers flowing into the Black Sea and their discharges are Danube (203 km³ year⁻¹), Dniiper (54 km³ year⁻¹), Dniesta (9.3 km³ year⁻¹), Don (28 km³ year⁻¹) and Kuban (13 km³ year⁻¹). In addition to these, a large number of smaller rivers along the Turkish and Bulgarian coasts contribute another 28 km³ year⁻¹ to the water budget of the sea (Bakan and Büyükgüngör 2000).

The Black Sea has suffered from extensive pollution over the last few years due to unmanaged fishing, unrestricted shipping, mineral exploitation, dumping of toxic wastes, discharge of domestic wastes from coastal cities and pollutants carried by rivers (Tuncer et al. 1998). Chemical and microbiological pollution, synthetic organic contaminants, heavy metals, radionuclides and oil pollution have caused the environment of the Black Sea to deteriorate dramatically in terms of biodiversity, habitats, fisheries resources, aesthetic and

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Fig. 1 The Black Sea Basin

recreational value and water quality (Bakan and Büyükgüngör 2000). Natural weathering and human activities are both sources of metals, which are transported to the marine coastal environment through rivers and the atmosphere (Angelidis and Aloupi 2000). Anthropogenic activity has introduced substantial amounts of trace metals (Ag, Co, Cr, Cu, Mn, Ni, Pb and Zn) into the oceanic environment (Matthai et al. 2002). Heavy metal contamination of aquatic ecosystems has been recognized as a serious pollution problem (Küçüksezgin et al. 2002). Pollution of coastal marine environments with trace metals is a major concern, since it may lead to deterioration of natural habitats by depleting ecologically sensitive species or eliminating or tainting commercial species (Akhter and Al-Jowder 1997). Heavy metals are generally brought to the sea by river input and/or industrial discharge. Their concentrations are generally directly correlated to the organic matter content, so that problems are exacerbated in coastal sediments where heavy metals are known to accumulate especially near population centres, river outflows and industrial outfalls (Danovaro 2003).

Materials and methods

Characteristics of study areas

The Black Sea coast of Turkey is 1,695 km long, extending from Bulgarian border in the west to the

Georgian border in the east. The area of the region is about 141,000 km² or about 18% of the total surface area of Turkey (Bakan and Büyükgüngör 2000). Some cities and towns use sewerage systems, but small towns use septic tanks. Present sewerage systems are mainly old, but new systems are being constructed.

In the Black Sea Region of Turkey, nut facilities, tea-plant factories, variable food manufactures, fishing facilities are the main industry types. On the other hand, coal mining, fertilizer factories and copper facilities are rare but important industries (Final Project Report 1997). Details on industries and their type of wastes are shown in Table 1 (Bakan and Büyükgüngör 2000).

Kızılırmak and Yesılırmak, the two most important rivers of Black Sea Region, and a lot of big and little industries (food, cement, fertilizer, pesticide, resin, plastic, textile, cigarette manufacturing) exist in the Middle Black Sea Region of Turkey. Most of these factories have no treatment plant and they have potential to create local pollution problem (Altas et al. 2001). The two important iron and steel factories of Turkey exist in the western part of the Black Sea region. On the other hand, the eastern part of the Black Sea Region has no important industrial factories, but only hazelnut facilities, floor manufacturing and fish-oil factories. Besides small industrial activities, pulp and paper factory present in this region is one of the important industries.

Table 1 The major Turkish industries and their type of waste in the Black Sea Region (Bakan and Büyüküngör 2000)

Type of industry	Probable pollutants and characteristics of effluents	Location
Food manufacturing (slaughtering, dairy products, canning of fruits/vegetables/fish, grain mill and bakery products, sugar factories, etc.)	BOD, COD, suspended, chemical and organic materials, odor, specific pollutants from each type of manufacturing such as sugar and slaughtering	Giresun, Ordu, Samsun, Sinop, Sakarya, Trabzon, Zonguldak
Manufacture of paper and paper (pulp and paper)	pH change, high amount of suspended, colloidal and dissolved materials, cellulose	Giresun, Zonguldak
Manufacture of non-metallic mineral products (mainly cement factories)	Heated cooling water, suspended solids, inorganic salts	Ordu, Samsun, Trabzon
Manufacture of wood and cork products	Organics from staining and sealing wood products	Ordu, Sakarya
Non-ferrous metal basic industries	Acid, metals, toxic, low volume, mainly mineral matter	Samsun, Trabzon
Manufacture of industrial chemicals (fertilizers and pesticides, resins and plastics)	Acids, mineral elements, suspended solids, caustic, phenols, formaldehyde	Samsun, Sakarya
Manufacture of textile	Highly alkaline, colored, high BOD, temperature and suspended solids	Samsun, Zonguldak
Beverage industries (soft drinks)	BOD, suspended and precipitable solid materials, fat and oil	Giresun, Ordu, Trabzon
Tea plant factories	Wastes of tea leaves	Rize, Artvin, Trabzon, Giresun
Cigarettes	Wastes from tobacco and its treatment	Samsun, Sinop, Trabzon
Coal mining	SiO ₂ , CaCO ₃ , Al ₂ O ₃ , Co, Cd, Li in coal ash	Zonguldak
Hazelnut	Suspended material	Trabzon, Ordu, Giresun

Sampling

Marine samples were taken from surface at certain stations including both 500 and 5,555 m (3 mile) distance from edge at same direction along the Sinop, Samsun and Ordu cities located in the Middle Black Sea region of Turkey during May 2000–October 2001 as seasonal in order to determine the impact of marine activities.

Samples were taken in 5 weeks of the year April–May 2000 and analyzed in order to designate the heavy metal pollution of certain rivers, streams, harbor and shores in Samsun’s boundary. In addition, 32 routine monitoring sampling stations—21 of them were rivers and streams, 5 were industrial discharge channels, 6 were sewage outlets—were selected at the Black Sea coast for the same purpose (Table 2). Selected rivers and streams, industrial and domestic discharge channel were sampled as seasonal during 1996. Different numbers of sample were collected from every river and stream depending on the width of them. The composite samples were then transferred to storage bottles each containing preservation reagent for a specific parameter. Water discharged by each river and stream was also measured during sampling. Samples belonging to industrial outlets and domestic sewerages were collected from the nearest part to the discharge point.

Analysis

Heavy metals—Cd²⁺, Pb²⁺, Zn²⁺, Ni²⁺, Cu²⁺, Co²⁺ and Cr⁶⁺—were determined by flame atomic absorption spectrometry (UNICAM 929), and the values of detection limits of that instrument for heavy metals mentioned above are 2, 10, 2, 10, 2, 30 and 20 µg L⁻¹, respectively. Measurements were made using the standard methods given by APHA, AWWA, WEF (1995).

Results and discussion

Investigation of marine pollution arising from heavy metals in the Middle Black Sea region of Turkey during May 2000–October 2001 as seasonal

This investigation was carried out with the intent to determine the contribution of land-based and marine activities to the Black Sea, bringing up its present state. Sampling points, date and determined heavy metal concentration of stations were given in Table 3. According to Marine General Quality Criteria given in Turkish Environmental Regulation (Table 4), it was seen that Cd²⁺ and Cu²⁺ levels generally and Pb²⁺ and Zn²⁺ levels sometimes exceeded the criterion, while Ni²⁺ concentrations are at the desired levels.

Table 2 Selected sample stations and their characteristics (Final Project Report 1997)

No.	Sampling station	Origin of wastes	No.	Sampling station	Origin of wastes
1	Sakarya River	Domestic and industrial	17	Samsun sewerage system	Domestic
2	Çark Stream	Industrial	18	Mert River	Domestic and industrial
3	Melen Stream	Industrial	19	TÜGSAŞ Fertilizer Ind.	Industrial
4	Alaplı Stream	Industrial	20	KBİ Copper Ind.	Industrial
5	Erdemir South Collector	Industrial	21	Yeşilırmak River	Domestic and industrial
6	Erdemir North Collector	Industrial	22	Miliç Stream	Domestic and industrial
7	Gülüç Stream	Industrial	23	Civil Stream	Industrial
8	Çatalağzı Stream	Domestic and industrial	24	Melet River	Domestic and industrial
9	Kozlu Stream	Domestic and industrial	25	Ordu Sewerage System	Domestic
10	Zonguldak Stream	Domestic	26	Giresun Sewerage System	Domestic
11	ÇATES-B Ash Channel	Industrial	27	Aksu Stream	Domestic and industrial
12	Filyos Stream	Domestic and industrial	28	Fol Stream	Domestic and industrial
13	Bartın Stream	Domestic and industrial	29	Söğütlüdere Stream	Domestic and industrial
14	Sinop sewerage system	Domestic	30	Trabzon Sewerage System	Domestic
15	Kızılırmak River	Domestic and industrial	31	Değirmendere Stream	Domestic and industrial
16	Kürtün Stream	Domestic	32	Rize Swerage System	Domestic

Table 3 The seasonal results of heavy metal concentrations of shore and off-shore sampling points through the Sinop, Samsun and Ordu cities in the Middle Black Sea Region of Turkey during May 2000–October 2001

SAMPLING POINT		SAMPLING DATE and POLLUTANT LEVEL ($\mu\text{g M}^{2+} \text{L}^{-1}$)																								
		M A Y 2000						O C T O B E R 2000						A P R I L 2001						O C T O B E R 2001						
		Cd	Pb	Zn	Ni	Cu	Mn	Cd	Pb	Zn	Ni	Cu	Mn	Cd	Pb	Zn	Ni	Cu	Mn	Cd	Pb	Zn	Ni	Cu	Mn	
1-Cape of Sinop	A	13	-	42	-	-	152	-	-	-	-	200	160	34	30	-	-	-	-	-	×	×	×	×	×	×
	B	×	×	×	×	×	×	4	-	-	-	180	-	50	-	538	-	-	-	-	×	×	×	×	×	×
2-Kızılırmak River	A	13	-	44	-	-	127	-	-	-	-	110	-	-	25	-	12	-	-	-	-	307	-	81	23	99
	B	×	×	×	×	×	×	6	-	-	-	180	-	-	-	-	60	-	-	-	-	-	94	15	142	
3-Kürtün Stream	A	-	-	35	-	-	135	6	261	-	-	200	-	17	-	338	-	-	-	-	-	148	11	12	20	224
	B	×	×	×	×	×	×	-	250	-	-	250	140	-	130	-	-	-	-	-	-	67	-	31	5	166
4-Samsun Harbor	A	-	-	26	-	-	127	6	-	-	-	200	-	39	20	-	-	6	-	-	-	34	133	-	26	163
	B	×	×	×	×	×	×	-	-	-	-	210	-	35	-	549	133	36	18	-	-	21	56	-	163	
5-Mert River, K.B.İ*,TÜGSAŞ**	A	30	-	13	-	-	157	-	218	-	-	240	150	20	20	207	78	-	33	-	171	3	31	20	252	
	B	×	×	×	×	×	×	-	-	-	-	40	41	10	-	-	12	56	-	91	-	10	-	250		
6-Yeşilırmak River	A	18	-	-	-	-	178	8	-	-	-	30	-	-	-	24	3	113	-	-	555	-	3	244		
	B	×	×	×	×	×	×	-	-	-	-	190	60	67	10	7	-	12	54	-	262	231	-	-	197	
7-Melet River	A	25	-	11	-	-	191	-	164	-	-	310	-	-	-	-	-	26	98	×	×	×	×	×	×	
	B	×	×	×	×	×	×	-	-	-	-	140	-	26	163	-	30	-	69	×	×	×	×	×	×	

A: Shore(500 m from edge) ; B: Off-shore(5555 m - 3 mile from edge) ; *: Copper Ind.; **: Fertilizer Ind.; ×: not sampled ; -:not detected

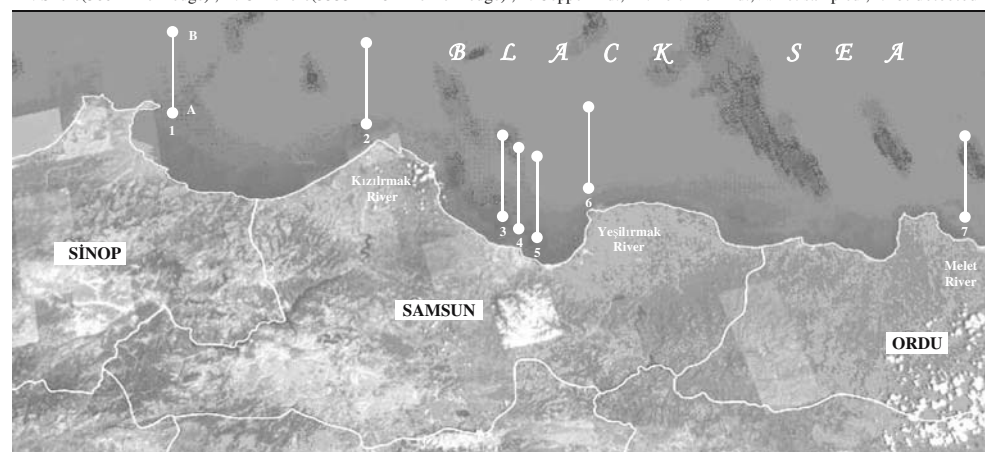


Table 4 Some part of General Quality Criteria of Marine (Turkish Environmental Regulation 1999)

Parameters (mg L ⁻¹)	Criterion
SS	30
Cu	0.01
Cd	0.01
Cr	0.1
Pb	0.1
Ni	0.1
Zn	0.1

The determination of heavy metal concentration especially at offshore sampling points means that the Black Sea is seriously exposed to pollution-originating marine activities. It is already known by world public opinion that foreign flagships remove their hazardous wastes by throwing them into the Black Sea. The results obtained in our studies show that this situation keeps going. In order to prevent the pollution, it is needed that the countries surrounding the Black Sea

fits willingly and seriously the contract which signed to each other.

Determination of weekly heavy metal concentrations at certain points in Samsun boundaries

In the land-based pollution monitoring studies, 11 sampling points comprising river, stream, shore and harbor were selected considering to be exposed the heavy metal pollution. The samples were taken between 14 April and 18 May 2000, in the 5-week period, and Cd²⁺, Pb²⁺, Zn²⁺, Cu²⁺, Ni²⁺ and Co²⁺ concentrations of these points were determined. The concentrations of heavy metals for 5 weeks and location of certain sampling points along the Samsun boundary were presented in Table 5.

Weekly examinations showed that the results were quite changeable. While water quality of a station en-

Table 5 The concentrations of heavy metals of certain sampling points along the Samsun boundary between 14 April 2000 and 18 May 2000

PARAMETER Sampling Point / Date	LEAD (µg Pb ²⁺ L ⁻¹)					CADMIUM (µg Cd ²⁺ L ⁻¹)					COPPER (µg Cu ²⁺ L ⁻¹)					ZINC (µg Zn ²⁺ L ⁻¹)					NICKEL (µg Ni ²⁺ L ⁻¹)					COBALT (µg Co ²⁺ L ⁻¹)				
	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000	14.04.2000	21.04.2000	05.05.2000	12.05.2000	18.05.2000
1.Kızılırmak River	989	-	-	-	347	13	-	15	68	13	70	-	53	-	61	-	-	2	-	-	-	-	-	-	-	-	-	-	-	-
2.Altunkum Shore	-	326	-	522	-	15	43	-	16	43	4	4	35	-	65	-	-	-	-	49	-	-	-	-	-	-	-	-	-	-
3.İncesu Stream	-	-	-	-	-	15	-	4	14	-	61	45	27	43	-	-	-	-	-	385	-	-	-	-	-	-	-	-	-	219
4.Kurupelit Stream	-	-	-	-	347	2	28	38	-	-	79	29	-	53	61	-	-	-	-	2	-	-	-	-	-	-	-	-	-	31
5.Kurupelit Shore	-	-	370	-	-	58	-	60	41	-	65	-	45	35	25	-	-	-	-	-	-	-	-	-	-	-	-	-	-	135
6.Kürtün Stream	-	-	435	-	-	-	25	-	15	9	53	33	22	27	33	-	-	-	-	-	-	-	-	-	-	341	-	-	-	-
7.Samsun Harbour	-	489	507	326	-	10	2	28	23	13	38	58	33	27	38	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-
8.Mert Stream	-	-	-	315	-	25	41	28	15	13	-	43	61	-	33	-	-	-	-	-	-	-	-	-	-	-	42	-	-	-
9.Kirazlık Stream	641	-	-	-	-	20	-	30	-	-	40	43	-	9	58	-	-	-	-	821	-	-	49	-	-	-	286	-	322	-
10.Kirazlık Shore	x	-	674	-	-	x	25	9	53	23	x	27	20	40	68	x	-	-	-	-	x	-	-	-	-	x	-	-	-	-
11.Yeşilirmak River	-	-	-	424	358	38	-	48	-	13	10	56	-	63	20	-	-	-	-	422	-	-	-	-	-	-	-	-	-	89

x : not sampled ; - : not detected

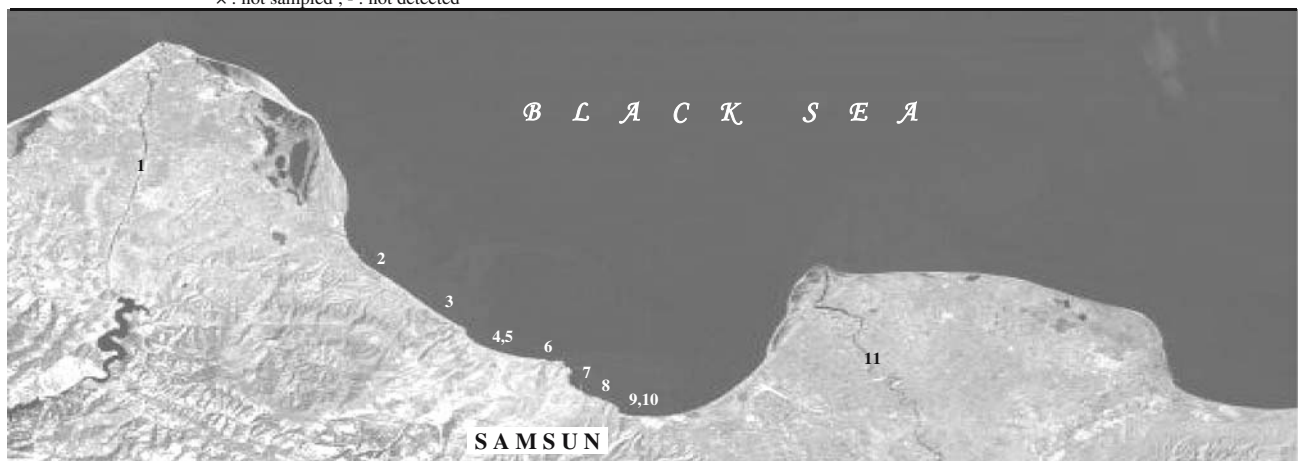


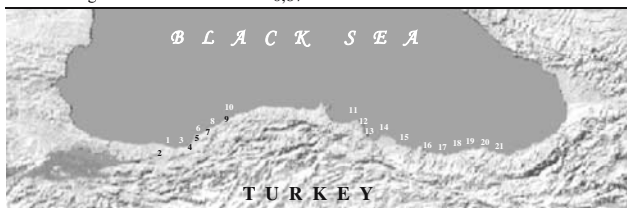
Table 6 Some part of Quality Criteria of Inland Water Sources according to the classes (Turkish Environmental Regulation 1999)

Water quality parameters	Water quality classes			
	I	II	III	IV
pH	6.5–8.5	6.5–8.5	6.0–9.0	Above 6.0–9.0
T. Phosphorus ($\mu\text{g PO}_4^{3-}\text{-P L}^{-1}$)	20	160	650	>650
BOD (mg L^{-1})	4	8	20	>20
Cadmium ($\mu\text{g Cd}^{2+}\text{ L}^{-1}$)	3	5	10	>10
Lead ($\mu\text{g Pb}^{2+}\text{ L}^{-1}$)	10	20	50	>50
Copper ($\mu\text{g Cu}^{2+}\text{ L}^{-1}$)	20	50	200	>200
T. Chromium ($\mu\text{g Cr L}^{-1}$)	20	50	200	>200
Cobalt ($\mu\text{g Co}^{2+}\text{ L}^{-1}$)	10	20	200	>200
Nickel ($\mu\text{g Ni}^{2+}\text{ L}^{-1}$)	20	50	200	>200
Zinc ($\mu\text{g Zn}^{2+}\text{ L}^{-1}$)	200	500	2,000	>2,000
Manganese ($\mu\text{g Mn}^{2+}\text{ L}^{-1}$)	100	500	3,000	>3,000

ters into Class I in a week according to the “Water Quality Criteria for the Inland Water Sources” (Turkish Environmental Regulation 1999) given in Table 6, for example in another week the station’s water quality enters into Class IV. It was found that the rivers, streams, shores and harbor which were determined pollutants belong to Class IV, Class IV, Class II–III, Class I–II, Class I–II–III and Class II–III–IV, from the point of view of Cd^{2+} , Pb^{2+} , Cu^{2+} , Zn^{2+} , Co^{2+} and Ni^{2+} , respectively.

Table 7 Annual load of heavy metals in rivers and streams along the Black Sea Coast of Turkey and sampling stations

NAME	DISCHARGE ($\text{km}^3\text{ yr}^{-1}$)	POLLUTANT (t yr^{-1})		
		T. Cr	Cd	Pb
1-Sakarya River	6,02	128,595	781,144	-
2-Çark Stream	0,31	4,086	12,867	-
3-Melen Stream	1,57	95,96	28,726	-
4-Alaplı Stream	0,27	-	11,520	13,927
5-Gülüç Stream	1,19	-	20,378	-
6-Çatalağzı Stream	0,13	2,627	3,115	-
7-Kozlu Stream	0,02	-	0,053	-
8-Zonguldak Stream	0,13	0,791	0,885	-
9-Filyos Stream	3,22	-	262,824	-
10-Bartın Stream	0,36	16,230	0,773	18,196
11-Kızılırmak River	7,39	427,101	1234,239	722,303
12-Kürtün Stream	0,16	22,588	2,084	1,239
13-Mert River	1,06	19,214	98,761	-
14-Yeşilirmak River	10,26	2549,228	1505,295	-
15-Miliç Stream	0,43	-	3,067	0,459
16-Civil Stream	0,6	3,140	0,117	-
17-Melet River	0,83	128,298	124,780	5,045
18-Aksu Stream	0,97	3,608	8,326	-
19-Fol Stream	0,20	-	-	-
20-Söğütüdere Stream	0,12	-	2,633	-
21-Değirmendere	0,87	-	-	-



The realized studies of pollution investigation along the Black Sea Coast of Turkey in 1996

Annual discharges of heavy metals in selected rivers and streams resulted from evaluation of inventories and their locations in the Black Sea Region are given in Table 7. The concentration of heavy metals in domestic discharges and their locations are illustrated in Fig. 2.

It was clearly seen that transportation of heavy metals from Yesilirmak, Kızılırmak and Sakarya rivers to the Black Sea was rather of high quantity. Although their annual discharges were low levels, Melet river and Filyos stream have more pollutant loads than streams and rivers which are of same characteristics. The heavy metal loads of Çatalağzı, Kozlu, Zonguldak, Bartın, Kürtün, Miliç, Civil, Söğütüdere streams and Aksu River are too low. Cd^{2+} , Pb^{2+} and T.Cr ions were detected at about 95, 30 and 60% of the whole of investigated streams and rivers, respectively. These heavy metal ions were not diagnosed at Fol and Değirmendere Streams.

It was found that examined rivers and streams generally belong to Class III and Class IV for Cd^{2+} , Pb^{2+} and T.Cr concentrations according to the criteria (Table 6). Consequently, it is seen that the water quality of rivers and streams so decreases at the term to be exposed to the heavy metal pollution.

Cd^{2+} , Pb^{2+} and T.Cr were found at the domestic discharge points of Ordu and Rize cities, while only one of these heavy metals were determined at all domestic discharge points except Sinop city. Detected concentrations for Cd^{2+} , Pb^{2+} and T.Cr are in allowable levels for sewerage systems according to Turkish Environmental Regulation (Table 8). It is seen that the heavy metal pollution increase gradually between 1993 and 1996, when we compare our studies with Tuncer et al. (1998) taking into consideration same parameters and sampling stations.

Fig. 2 The concentrations of heavy metals in the domestic discharge stations of Black Sea Region in Turkey during 1996

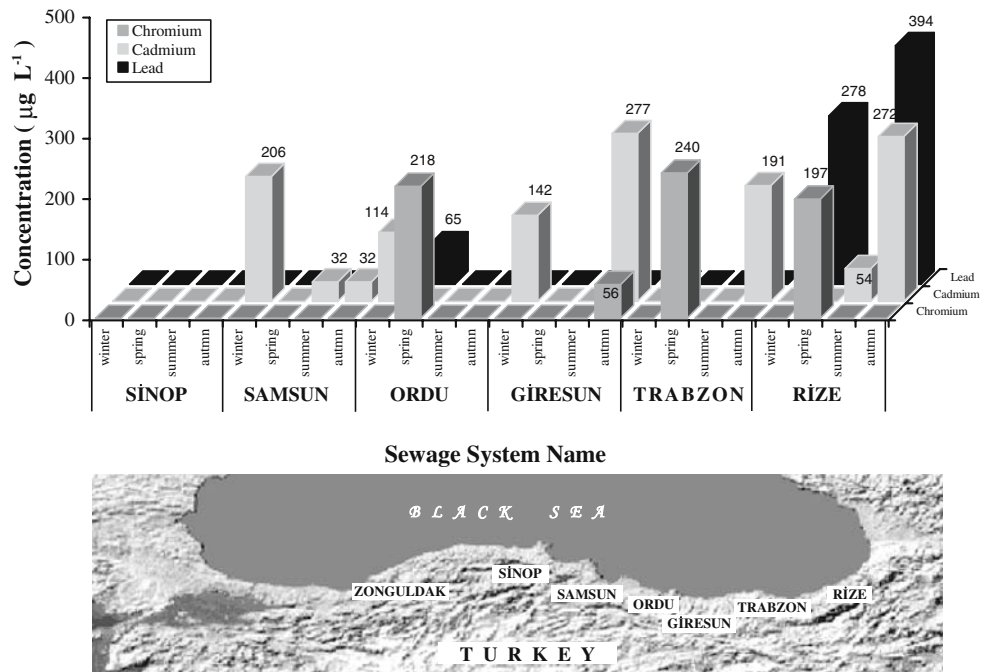


Table 8 Some part of considering standards of wastewaters for discharge to sewerage systems (Turkish Environmental Regulation 1999)

Parameter (mg L ⁻¹)	At the wastewater substructure establishments resulting whole treatment	At the wastewater substructure establishments resulting deep marine discharge
Pb	3	3
Cd	2	2
Cr	5	5
Cu	2	2
Ni	5	5
Zn	10	10

Conclusion

In three different studies realized at separate times, heavy metals were measured at approximately 60 sampling points including domestic, industrial discharges, river, stream, harbor and marine to asses the potential contribution on the local pollution problems in the Black Sea along the Turkey and its certain cities coasts located on the Middle Black Sea Region. A lot of big and little industries in the Black Sea region have potential to create local pollution problem because of no treatment plant facility. Most cities located the Black Sea Coast of Turkey suffer from local water pollution problems due to discharges of untreated domestic and industrial wastes. Observed heavy metal

concentration will decrease definite proportion, when the projects including collection and treatment of domestic and industrial wastes are completed.

Examinations as weekly and seasonal showed that the results are quite changeable. Most of the examined stations enter into Class III and Class IV according to the “Water Quality Criteria for the Inland Water Sources”. Some examined stations also enters into Class I. Consequently, we can say that the examined stations were exposed to specific contaminants at different times.

In marine pollution monitoring examination, it was found that heavy metal levels at most sampling points generally fitted the quality criteria. However, determined heavy metal concentration over the criterion, especially at some offshore stations, show that the Black Sea is polluted by marine activities.

In order to stop and prevent the pollution in the Black Sea, further dissuasive laws have to be brought into force by the countries located in the Black Sea Basin. In addition to this, continuous pollution measuring stations in the Black Sea must be established at certain points in order to observe pollution and form inventory.

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