

**SURFACE WATER POLLUTION IN WEST BLACK SEA COASTAL REAGION:
A Case Study in Zonguldak City and Its Surrounding Province (Turkey)
POLLUTION DE L'EAU DE SURFACE EN REGION CÔTIÈRE DE LA MER
NOIRE OCCIDENTALE: Une étude de cas de la ville de Zonguldak et sa province
environnante (Turquie)**

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Abstract: In this study, the hydrogeological and pollution characteristics of surface water, tap water and local spring water along 40 km coastal regions in Zonguldak city and its surrounding province having approximately 175 thousands population were examined. In addition, pollution loads of domestic and industrial wastewater and their effects to surface water quality were investigated and proposals for protection of surface water resources were put forward. For these aims, 28 sampling points were selected to sample surface water in respect to population activities in different time periods of 2006. During the sampling, temperature, conductivity and pH were measured in-situ by using portable equipments. In laboratory studies, Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , SO_4^{2-} , HCO_3^- , CO_3^{2-} , fecal and total coliform etc for drinkable water and COD (chemical oxygen demand), BOD_5 (biological oxygen demand), TSS (total suspended solids), NO_2^- , NO_3^- etc for wastewater and surface water were examined.

Key words: Surface water pollution, West Black Sea, Hydrogeology.

Résumé : Dans cette étude, les caractéristiques hydrogéologiques et de pollution de l'eau de surface, de l'eau du robinet et de l'eau de source locale le long des régions côtières sur 40 kilomètres dans la ville de Zonguldak et sa province environnante ayant approximativement 175 milliers de population ont été examinées. En outre, des charges de pollution de l'eau domestique et industrielle de gaspillage et de leurs effets sur la qualité de l'eau extérieure ont été étudiées et des propositions pour la protection des ressources d'eau de surface ont été formulées. Pour ces objectifs, 28 points de échantillonnage ont été choisis pour prélever l'eau de surface en ce qui concerne des activités de population dans des périodes différentes de l'année 2006. Pendant le prélèvement, la température, la conductivité et le pH étaient in-situ mesuré en utilisant les équipements portatifs. Dans des études de laboratoire, la coliforme etc. de Ca^{2+} , de Mg^{2+} , de Na^+ , de K^+ , de Cl^- , de SO_4^{2-} , de HCO_3^- , de CO_3^{2-} , fécale et totale pour l'eau buvable et la demande chimique d'oxygène, demande biloque d'oxygène, se montent aux solides, au nitrite, au nitrate, etc. suspendus pour l'eau usagée et l'eau de surface ont été examinées.

Mots clés : Pollution d'eau de surface, la Mer Noire Occidentale, Hydrogéologie.

INTRODUCTION

In the marine environment, human-caused disturbances take many forms. Owing to source strengths and pathways, the greatest effects tend to be in the coastal zone. Waters and sediments in such regions bear the brunt of industrial and sewage discharges and are subject to dredging and spoil dumping. Agricultural runoff may contain pesticide residues and elevated nutrients producing eutrophication in the water bodies or anoxic conditions in underlying sediments.

The Black Sea having an area of 4.2×10^5 km² and average depth of 1240 m is the world's largest land-locked sea. Approximately, 90% of its water mass is anoxic, representing the world's largest anoxic water mass (Sorokin, 1983). The Black Sea have been suffering from extensive pollution for last two decades due to unmanaged fishing, unrestricted shipping, mineral exploitation, dumping of toxic wastes, discharge of domestic wastes from coastal cities and pollutants carried by rivers (Mee, 1992).

Unregulated and unplanned freshwater withdrawal for irrigation purposes, hydro- and thermal-power generation, the use of coastal areas for several types of construction and uncontrolled solid waste deposition field, and the many untreated domestic, industrial and agricultural wastes (solids or liquids) discharged into the rivers that eventually drain into the Black Sea causing all detrimental effects on its health. The main sources of pollution in the Black Sea are the rivers which flow into the region discharging huge amount of pollution (i.e. Danube pollution load is 203 km³ yr⁻¹). In addition to these major rivers loads, especially along the Turkish and Bulgarian coast a great number of small rivers flow into the Black Sea causing substantial marine pollution (Tuncer et al., 1998). Data on both the levels of pollutants and annual loads to the Black Sea from individual sources are very scarce. Although several studies have been conducted in Turkish coasts of the Black Sea for determination of the pollutants levels and annual loads (i.e., Tuncer et al., 1998; Bakan and Büyükgüngör, 2000 and Bakan and Ariman, 2004), more studies are needed to carry out at nationwide in microscale for understanding marine pollution in details in the whole region.

In this study, surface, domestic tap water and local spring water pollution in the part of the west Black Sea region, hereafter called Zonguldak region, along 40 km coastal regions in Zonguldak city and its surrounding province having approximately 175 thousands population were examined. In addition, pollution loads of domestic and industrial wastewater and their effects to surface water quality were also investigated. Zonguldak region with its industrial (iron and steel complex, 300 MW thermal power plant and hard coal exploitation and processing), domestic (no treatment plant available) and agricultural activities is the most polluted region in the whole Black Sea region.

EXPERIMENTAL STUDIES

Study area and hydrological and geological settings

Study area, which is comprised of Kilimli and Kozlu towns, and also Zonguldak city centre, having 40 km coastline and approximately 175000 inhabitants is located in the West Black Sea Region of Turkey (Fig. 1). In the region, mean annual temperature is 13.6 °C, high and relative moisture content, annual mean precipitation amount is 1235 mm according to observations during 72 years (Özölçer and Aksoy, 2006). Most substantial surface water is Çatalağzı, Kilimli, İnağzı, Üzülmez, Kozlu, Değirmenağzı and Ilıksu streams. Their average flow rates are 3.170, 0.317, 0.100, 5.780, 0.464, 0.350

and 1.100 m³/s, respectively (Zonguldak Governorship, 2004; Tuncer et al., 1998).

Most important coal area of the Turkey is situated in Zonguldak and its province. The coal series occurred during Westfaalen (Carboniferous) are lower-bounded by Visean aged karstic limestones and upper bounded by Aptian-Barremian aged karstic limestones.

Sampling

In this study, there were 28 sampling points of which 7 of them was spring and tap water used for drinking purposes, 14 of them were sampled from streams discharged into the Black Sea. The rest of the samples (7) were taken from wastewater discharge points directly flow into the sea or to the streams (Fig. 1). During the study, the sampling was carried out three different time (February, May, July) of 2006 representing wet and drought seasons.

Laboratory studies

During the sampling, Conductivity (EC), pH and Temperature were measured in-situ by using portable equipments. In the laboratory, major ions and cations (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , SO_4^{2-} , HCO_3^- , CO_3^{2-} , etc.), and characteristics of wastewater (COD, BOD₅, TSS, NO_2^- , NO_3^- , total and fecal coliform etc.) were conducted according to methods described in APHA (1981).

RESULTS AND DISCUSSION

Hydrogeochemistry

In the region, all of the surface water is generated from karst aquifers containing mostly Ca^{2+} and HCO_3^- ions. Mg^{2+} and SO_4^{2-} ions were also found in deep dolomitic karst aquifer (Visean aged).

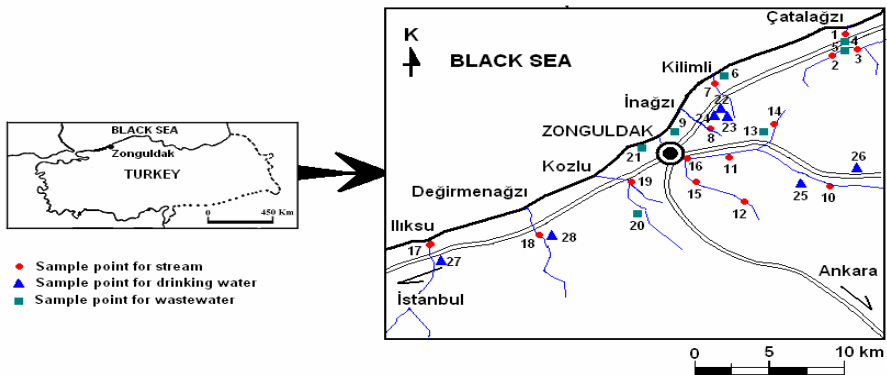
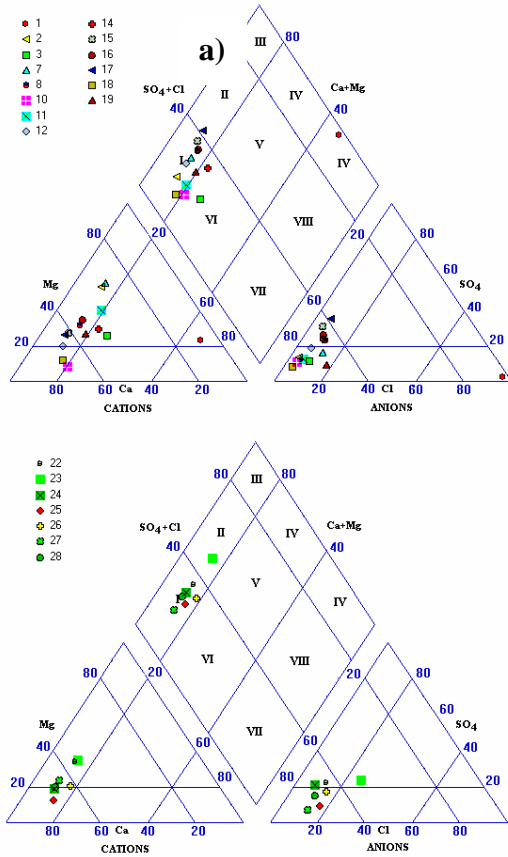


Fig. 1. Location map of study area and sample points



b)

Fig. 2. Piper diagrams for all streams (a) and drinking water samples (b).

Shoeller and piper diagrams presented for all samples were given in Fig. 2 and Fig. 3. As seen in these figures; Although, surface water contains mostly Ca^{2+} and HCO_3^- ions, samples taken from Çatalağzı and Kilimli regions (sampling point numbered 2, 3, 7, 8, and 22) contain relatively more Mg^{2+} cation than others. This phenomenon expressed that these waters are generated from old geological series of which Visian aged lime stones. Water sampled from Üzülmez, Kozlu and Değirmenağzı regions (Aptian and Barremian aged karst aquifers) have had less Mg^{2+} cations then waters sampled from Çatalağzı and Kilimli region.

As seen in the Fig. 2, sample points 1 and 23 have high Cl^- concentration levels. In reality, Cl^- content for samples taken from this region is expected in low level. The reason of this is sea water used as cooling water in the power plant and discharged into the stream just before the sampling point 1. For sampling point 23 (spring water used for drinking purpose), high Cl^- concentration may be attributed to wastewater contamination from residential houses.

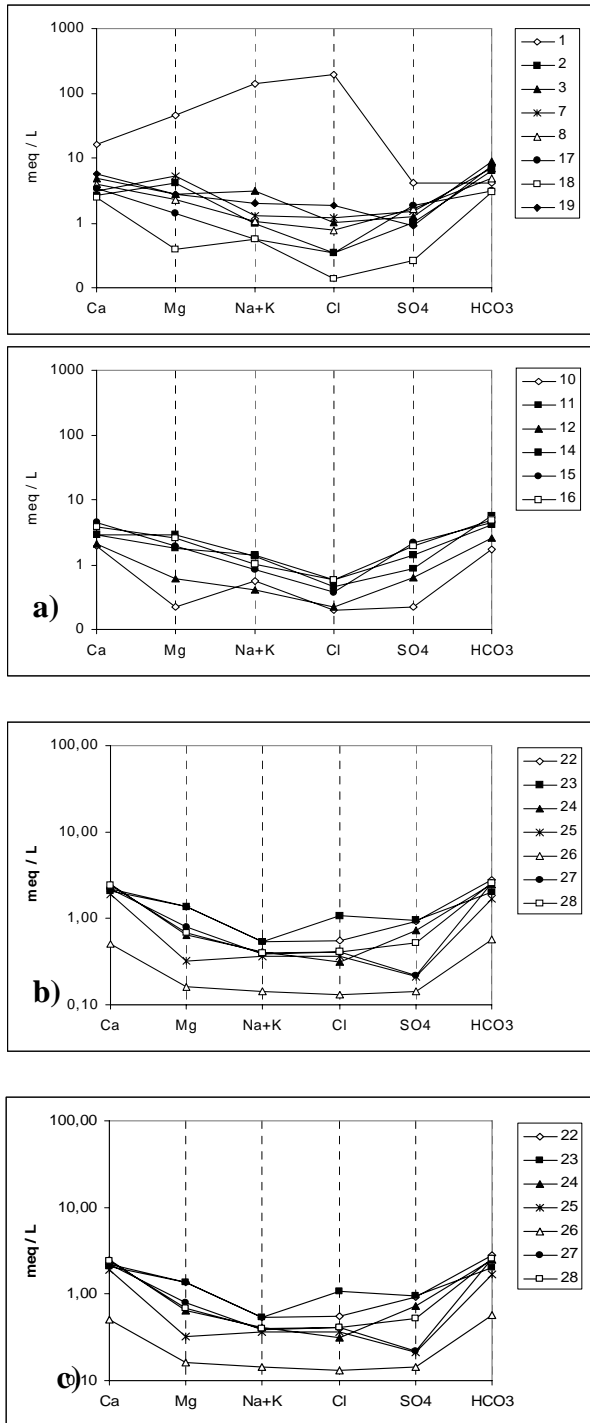


Fig. 3. Schoeller diagrams for all streams [(a) and (b)] and drinking water samples (c).

Wastewater discharges in the study area

In Zonguldak region, most discharge of the wastewater occurred from residential areas. Some of those wastewater are collected and discharged into sea or streams by a main sewage collection system. Industrial wastewaters are consists of a hard coal processing water, a power plant wastewater, a slaughter house wastewater and a small scale organized industrial region. The general characteristics of wastewaters from power plant (sample points 4 and 5) and hard coal mines (sample point 13) are given in Table 1. As seen in the Table 1, COD and TSS were found very low for sample point 13. In sampling point 4 (power plant cooling water), EC and water temperatures values were determined very high in respect to other sampling points causing thermal and salinity pollution in Çatalağzı stream. In sampling point 5 (power plant wastewater), wastewater can be characterized as light type of domestic wastewater.

Domestic wastewaters of Zonguldak region were given in Fig. 4 showing COD, BOD₅, COD/BOD₅ ratio and TSS parametres. COD and COD/BOD₅ ratio of city center wastewater are higher then Kilimli and Kozlu towns (sampling points 6 and 20). In summer term, pollution loads of wastewater are decreased due to increase in water usage. In July, for sampling point 6 (wastewaters of Kilimli town), COD level were found in very high level. This is attrubuted to slaugther house wastewater was discharge during the sampling.

Table 1. General characteristics of wastewaters discharged from Çatalağzı Thermal Power Plant and Zonguldak coal mines.

Location	No	T(°C)			pH			EC (µS/cm)			TSS (mg/L)			COD (mg/L)			NO ₃ (mg/L)			NO ₂ (mg/L)		
		Feb.	May	July	Feb.	May	July	Feb.	May	July	Feb.	May	July	Feb.	May	July	Feb.	May	July	Feb.	May	July
Çatalağzı	1	16,1	31	35,3	8,26	7,84	8,17	16750	25400	25000	102	76	192	ND	ND	ND	33,3	40,9	34,8	0,019	0,011	0,013
Çatalağzı	2	9,0	21	22,2	8,12	8,40	7,91	347	941	949	111	277	600	ND	815	665	39,4	27,3	43,9	0,035	0,004	0,108
Çatalağzı	3	9,8	21,5	22,7	8,32	8,41	8,16	988	1026	984	226	313	605	262	839	678	31,8	16,7	37,8	0,034	0,018	0,074
Kilimli	7	12,0	21	22,8	8,05	8,14	7,85	914	880	848	513	2282	296	392	814	669	7,6	48,4	30,3	0,083	0,018	0,016
İnağzı	8	10,3	22	21,7	7,78	7,74	7,65	545	656	786	27	180	110	ND	51	109	4,6	7,6	12,1	0,050	0,003	0,009
Üzülmez	10	8,3	20,2	***	8,16	7,70	***	212	377	***	6	1	***	ND	ND	***	6,1	7,6	***	0,005	0,006	***
Üzülmez	11	10,3	27,6	28,9	8,41	8,36	8,35	392	876	754	26	94	20	34	64	86	ND	16,7	10,6	0,027	0,004	0,230
Üzülmez	12	8,4	21,4	22	8,46	8,20	7,92	142	394	418	26	2	4	ND	ND	16	6,1	3,1	***	0,008	0,017	0,016
Üzülmez	14	11,3	22,7	23,8	8,08	7,96	7,73	478	584	597	52	3	ND	12	45	70	ND	6,1	12,1	0,036	0,006	0,284
Üzülmez	15	10,8	22,4	24,5	8,80	7,95	7,91	383	851	833	39	***	ND	44	38	44	ND	28,8	***	0,027	0,008	0,081
Üzülmez	16	10,4	19,4	25,1	8,48	8,01	7,83	426	756	815	42	85	80	ND	68	135	3,1	72,6	15,2	0,045	0,036	0,005
İlksu	17	9,2	21,8	25,8	8,20	8,05	8,37	284	598	662	8	1	ND	35	44	60	6,1	9,1	3,1	0,008	0,005	0,002
Değirmenağzı	18	9,2	14,6	16,2	8,19	8,03	7,95	220	346	345	15	16	ND	25	27	31	10,6	13,6	3,1	0,006	0,004	0,004
Kozlu	19	11,5	23,3	26,6	8,27	8,06	8,05	530	935	1339	45	86	29	109	62	60	24,2	4,6	9,1	0,031	0,008	0,142

*** Could not determined

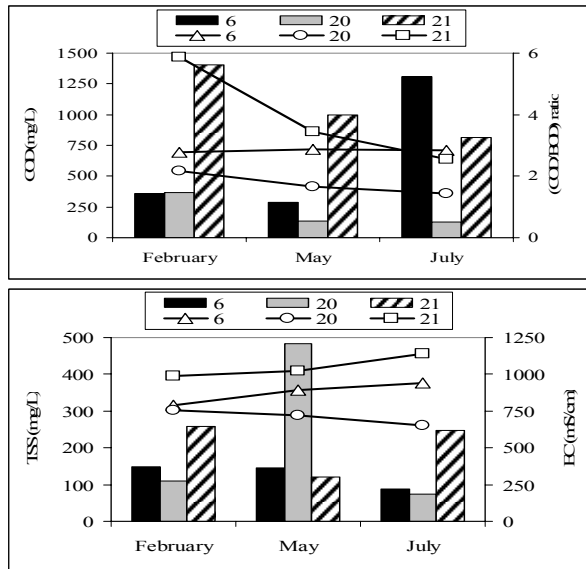


Fig. 4. Wastewater characteristics of domestic discharges in Zonguldak

COD/BOD₅ ratio is a representing parameter for biological treatability of a wastewater (Moraes and Bertazzoli, 2005). As shown in Fig. 4, for Kilimli (sample point 6) and Zonguldak city centre (sample point 21), COD/BOD₅ ratio is varied between 2 and 6. These results show that wastewater samples 6 and 21 can not be treated efficiently by using conventional biological treatment methods. Advance biological treatment methods and/or chemical treatment methods should be required to treat this kind of wastewaters (Fan et al., 2005).

Pollution loads discharged into the Black Sea

Pollution loads from domestic and industrial wastewater were estimated using COD and TSS values. Averages measured COD and TSS values were determined as 1075 and 210 for Zonguldak city centre and 435 and 175 mg/L for other town centres, respectively. For per calculating wastewater amount, 165 L/day per person value were assumed (Karagözoğlu et al., 2003; Türkman, 1986 and Orhon et al., 1997). In the calculations, total population of the Zonguldak region was used as 175000 (105000 for Zonguldak city centre and 75000 for the other towns). Hence, the result for COD and TSS load were given in Table 3.

Table 2. Pollution loads into the Black Sea calculated from experimental results for Zonguldak region.

Parameters	Zonguldak city centre	Other towns	Total
COD (ton/year)	6798	1834	8632
SS (ton/year)	1328	738	2066

Surface water pollution in the region

In the Zonguldak region, there are 7 important surface water streams. Generally,

domestic and industrial wastewaters have been discharged into the streams. In this section, the effect of these discharge on to the water quality of the streams were investigated and results presented in Table 3.

Çatalağzı Stream

Çatalağzı stream consist of two differet streams coming from Muslu and Çatalağzı town centre. Both towns discharge their wastewater into these streams. Therefore, for sampling point 2 and 3, COD (between 260-840 mg/L), TSS (between 110-600 mg/L), NO_2^- (between 0,040-0,108 mg/L) and NO_3^- (between 16,7-43,9 mg/L) values were determined very high (Table 3). Increase in draught caused decrease in water flow rate of Çatalağzı stream resulting high pollution level. Power plant cooling water (collected from the sea) causes tempreture increase in the stream up to 30°C (measured in sampling point 1) and its salinity level reaches marine water level ($\text{EC} > 16750 \mu\text{S/cm}$).

Kilimli and İnağzı Streams

Kilimli stream (sampling point 7) is the discharge media receives wastewater from coal enreachment plant, shipping construction yard, wastewater of the residential houses in Kilimli town. The other half of this wastewater discharges in to the sea via main sewage collector. As seen in Table 3, sampling point 7 has high level of COD and TSS indicating higher COD and TSS concentration than sampling point 6 taken from Kilimli sewage system (Table 1 and Fig. 4). İnağzı stream (sampling point 8), in summer season drought may happen, is coming off small fountain. Along the stream, there are about 100 shanty houses, and discharge their wastewaters into the stream. Therefore, during summer season, stream water mostly consists of wastewater from these shanty houses. As seen from Table 3, during the summer season COD value is higher then winter season indicating most pollution in the streams causes from shanty houses ($\text{COD} > 100 \text{mg l}^{-1}$).

Üzülmez Stream

Since there is no any sewage collections system and treatment plants, Üzülmez stream is the discharge media receives wastewater from residential houses, different industrial sectirs, mining sectors and commercial sectors in Zonguldak city centre. In this area, six sampling were carried out numbered as sampling points 10, 11, 12, 14, 15 and 16. Sample 13 was evaluated as mining wastewater in section 2 (wastewater discharge). In sampling point 10, during summer term due to drughtness sampling could not be performed. At this sampling point, water quality was determined very high because of no settlement around region.

First wastewater discharge into the Üzülmez stream was released from sampling point of 13 (coal mine wastewater) and 14 (residential houses wastewater). As seen from Table 3, at these sampling points COD values varies between 12-70 mg/L, TSS between 1-52 mg/L, NO_2^- between 0.06-0.284 mg/L and NO_3^- between 0-121 mg/L.

At sampling point 16, last sample point of Üzülmez stream before mixing with the sea, stream water quality was highly low. Because all kind of wastewater were mixed just before reaching this point, COD value reached up to 135 mglt, TSS 80 mg/lt and NO_3^- 72.6 mg/lt at the summer season.

Kozlu Stream

Since there is no any sewage collection system, most of the residential wastewater was discharged from elevated point in to the Kozlu stream. The main sewage collector (sample point 20) is also discharged in to the stream. Characteristics of sample point 20 were given before in Table 1.

Sample point 19 was the only point representing characteristics of the stream water just before mixing with the sea. During winter season, the water quality of the stream was worse than summer season. For example COD value was about 60 mg/L at summer season, but it was about 100 mg/L at winter season. This was because wastewater discharge into the stream rose due to high water usage containing low pollution at winter season.

Değirmenağzı and Ilıksu Streams

Due to no contamination from any source around the stream, değirmenağzı stream is a sort of surface water resource supplying drinking water to Kozlu town and some part of Zonguldak city. Its flow rate and water quality does not change with seasonal variation as shown in the Table 3 (sampling point 18). Its COD values varied between 25-91 mg/L, TSS 1-16 mg/L and NO_3^- 3.1-10.6 mg/L.

Ilıksu stream was examined as sampling point 17. Around this stream, at the weekends people come to the place for recreational purposes. Therefore, especially summer season waste discharge in to the stream rose. As seen in the Table 3, COD level was varied between 35-60 mg/L in indicating pollution level increases during the summer season.

Drinking water pollution in the region

In the region, water used for drinking purposes has been supplied from two different resources. First of the water resource is called Kozlu dam, was constructed on the Ulutan stream (a branch of Kozlu stream). Reservoir has approximately 19 km³ volume of water and supplies the drinkable water to the Zonguldak Metropolitan Area. Second water resource for the region is a small weir which supplies the drinking water to the Kozlu region and to same part of the Zonguldak city. In addition to these, there is a lot of small scale springs collected in small concrete tank resources used for drinking purposes without any pretreatment. In these small concrete tanks, water disinfection and routine clarification were not applied to observe water quality for public health.

In this study, representing Kozlu dam water, sampling point 24, was examined. For small scale spring water, sampling point 22 and 23 were examined. Sampling points 25 and 26, located near the Ulutan stream, and sampling points 27 and 28 water samples, located near the Ilıksu and Değirmenağzı stream, were other small scale spring water examined. Results of the microbiological tests of these drinking water samples were given in Fig. 5 showing total coliform found at sampling points 22, 23 and 25 and total and fecal coliform found at sampling points 22 and 23. This clearly showed that there was a contamination in a place between tank and offspring from sewage. If we ask where to pollution take place in the pipeline we may evaluate variation of ammonia, nitrite and nitrate concentration of the water sampled. As known, if there is just concentration from sewage, nitrogen content of water is form of ammonia. But by time ammonia is reduced initially into nitrite then finally into the nitrate. According to this process, if water contains high level of nitrite and ammonia, it indicates that contamination has just happened. But, if water includes nitrate, it shows that contamination was started in long time ago.

Table 3. Water characteristics of the stream samples for different seasons.

Location	No	T(°C)			pH			EC (µS/cm)			TSS (mg/L)			COD (mg/L)			NO ₃ (mg/L)			NO ₂ (mg/L)		
		Feb.	Ma	July	Feb.	May	July	Feb.	Ma	July	Feb.	Ma	July	Feb.	Ma	July	Feb.	Ma	July	Feb.	May	July
Çatalağzı	1	16,1	31	35,3	8,26	7,84	8,17	6750	2540	2500	102	76	192	ND	ND	ND	33,3	40,9	34,8	0,019	0,011	0,013
Çatalağzı	2	9,0	21	22,2	8,12	8,40	7,91	347	941	949	111	277	600	ND	815	665	39,4	27,3	43,9	0,035	0,004	0,108
Çatalağzı	3	9,8	21,5	22,7	8,32	8,41	8,16	988	1026	984	226	313	605	262	839	678	31,8	16,7	37,8	0,034	0,018	0,074
Kilimli	7	12,0	21	22,8	8,05	8,14	7,85	914	880	848	513	2282	296	392	814	669	7,6	48,4	30,3	0,083	0,018	0,016
İnağzı	8	10,3	22	21,7	7,78	7,74	7,65	545	656	786	27	180	110	ND	51	109	4,6	7,6	12,1	0,050	0,003	0,009
Üzülmez	10	8,3	20,2	***	8,16	7,70	***	212	377	***	6	1	***	ND	ND	***	6,1	7,6	***	0,005	0,006	***
Üzülmez	11	10,3	27,6	28,9	8,41	8,36	8,35	392	876	754	26	94	20	34	64	86	ND	16,7	10,6	0,027	0,004	0,230
Üzülmez	12	8,4	21,4	22	8,46	8,20	7,92	142	394	418	26	2	4	ND	ND	16	6,1	3,1	***	0,008	0,017	0,016
Üzülmez	14	11,3	22,7	23,8	8,08	7,96	7,73	478	584	597	52	3	ND	12	45	70	ND	6,1	12,1	0,036	0,006	0,284
Üzülmez	15	10,8	22,4	24,5	8,80	7,95	7,91	383	851	833	39	***	ND	44	38	44	ND	28,8	***	0,027	0,008	0,081
Üzülmez	16	10,4	19,4	25,1	8,48	8,01	7,83	426	756	815	42	85	80	ND	68	135	3,1	72,6	15,2	0,045	0,036	0,005
Ilksu	17	9,2	21,8	25,8	8,20	8,05	8,37	284	598	662	8	1	ND	35	44	60	6,1	9,1	3,1	0,008	0,005	0,002
Değirmenağzı	18	9,2	14,6	16,2	8,19	8,03	7,95	220	346	345	15	16	ND	25	27	31	10,6	13,6	3,1	0,006	0,004	0,004
Kozlu	19	11,5	23,3	26,6	8,27	8,06	8,05	530	935	1339	45	86	29	109	62	60	24,2	4,6	9,1	0,031	0,008	0,142

*** ; Dry stream or couldn't there is no analyzed ; ND ; Under or upper the dedection limits

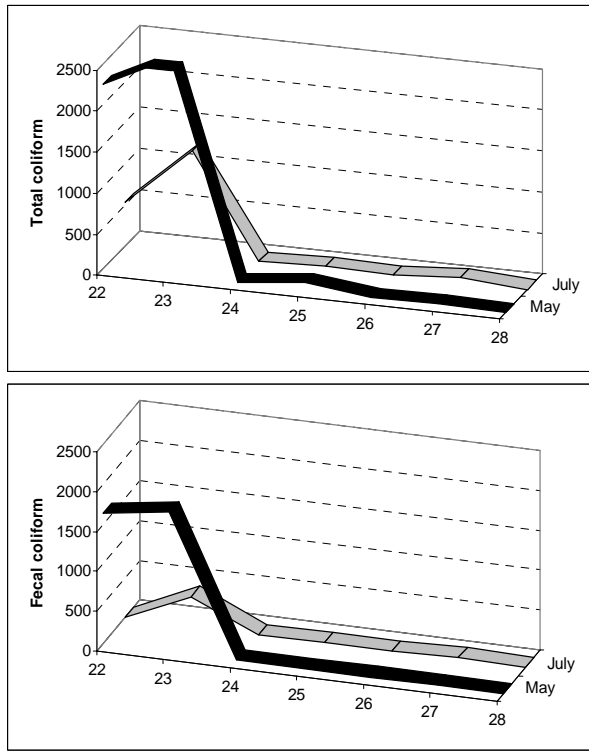


Fig. 5. Total (a) and fecal (b) coliform numbers for drinking water samples

In Fig. 6, for all drinking water, nitrite and nitrate concentration were presented. As seen in the Fig. 6, at sampling point 23, contamination was probably near the offspring place. But, at sampling point 22, contamination was probably near the tank. For sampling points 24, 27 and 28, although there was no bacteriological contamination, there were high levels of nitrate and nitrite concentration from agricultural activities and/or natural organic matter decay. Similar case may be conducted for sampling points 25 and 26.

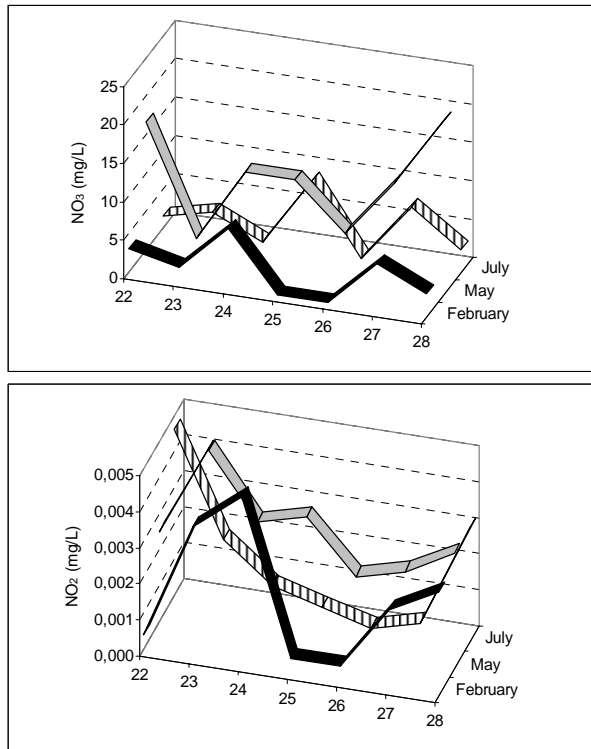


Fig. 6. Nitrate (a) and Nitrite (b) concentrations for drinking water samples.

CONCLUSION

This work is the first study to evaluate water pollution in the Zonguldak region. Following conclusions can be drawn from this study:

- In the region, surface and drinking water are rich in $\text{Ca}^{2+}/\text{Mg}^{2+}$ and $\text{HCO}_3^-/\text{SO}_4^{2-}$ due to water generated from karst aquifers.
- All streams in the region were used as a discharge medium for domestic and industrial wastewater. For this reason, large amount of organic matters and nutrients (nitrogen and phosphore) were discharged into the Black Sea. Pollution loads were evaluated as 8632 kgCOD/year and 2066 kgTSS/year.
- All of the drinking water supplied from natural way (without any pretreatment) was contaminated by means of sewage leakage and agricultural activities. This problem is the results of unplanned development of the region. Especially, shanty house around in the region (comprises 80 % of the residential house) causes unmanageable infrastructure. In recently, local authorities of the region tried to build main sewage system together with wastewater treatment plant in order to overcome these problems.

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