

The Effects of Lithology on Water Pollution: Natural Radioactivity and Trace Elements in Water Resources of Eskisehir Region (Turkey)

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Abstract The high radioactivity and trace elements in drinking water are common concerns for human health. The aim of this study was to investigate the eligibility of groundwater for drinking purpose in terms of both radioactivity and trace element contents in Eskisehir Region (Turkey). The study area is located in a highly populated residential area where water supply is mostly met from groundwater. The area is about 20,000 km², where igneous, metamorphic, and sedimentary rocks are exposed. The 209 water samples collected from 84 water resources (including thermal waters) were analyzed with respect to major ions, trace elements, and radioactivity (gross alpha and gross beta) during both in wet and dry seasons. Based on the analysis results, trace elements in 49 samples of 84 water

resources were over the limits of Code TS 266 1997 (Turkish Drinking Water Standards) and WHO 1993 standards. Particularly, Fe, Mn, Al, As, Ba, Zn, Cr, Cu, and B ion concentrations exceeded the limits. The gross alpha values in 18 locations and gross beta values in three locations also exceeded the limits of aforementioned standards in terms of radioactivity (gross alpha=0.1 Bq L⁻¹; gross beta=1 Bq L⁻¹). Furthermore, water radioactivity levels were close to the allowable limits in 33 water resources. The obtained results explicitly indicate that there is a strong relationship between the higher radioactivity–trace element contents and geochemical composition of rocks, which controls the radioactivity and trace element concentrations present in the aquifer.

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1 Introduction

According to the recent scientific researches, water resources, which are essential for the sustaining of living life on the Earth, are increasingly exploited and this causes a global water shortage. Therefore, it is essential to protect water resources and keep them clean for the next generations. However, water pollution is continuously being elevated in two ways: natural and anthropogenic. Either natural or anthropogenic, pollution still affects the quality of groundwater consumed for drinking and domestic uses. This study mainly deals with natural pollution.

The most likely origin of natural pollution in groundwater is the lithological and mineralogical properties of rocks (Freeze and Cherry 1979; Şahinç 1991; Apello and Postma 1992; Appleton et al. 1996). Many scientific researches stated that concentration of trace elements and radionuclides in drinking and domestic water might exceed the allowable limits due to the lithology of aquifer units (Choubey 1997; Choubey and Ramola 1997; Nishimura 1997; Focazio et al. 1998; Singh et al. 1999; Abrahams 2002; Moussa and El Arabi 2003; Yang et al. 2003; Brofferio et al. 2004). Large amounts of radioactivity in water resources and neighboring rocks may probably be the reason of widespread and fatal cancer diseases (Rangel et al. 2001; Vesterbacka 2005). For instance, the National Research Council in USA reported that, annually, 30,000–32,000 people die from lung cancer resulting from radon inhalation in dwellings near highly natural radioactive areas as well as from the groundwater extracted from the same area (Yu and Kim 2004; Zapecza and Szabo 1988). The origin of radioactivity is generally uranium and thorium series which are found in the minerals of various rock types.

In Turkey, few studies have been realized by a number of researchers on lithology-based radioactivity. Akyil et al. (1996) have investigated the α activity of ground water in West Anatolia. The gross alpha and beta levels in different surface and tap waters of Elazığ region and in the drinking water of Emendere-Village (Sındırğı-Balıkesir) region have been analyzed by Dogru et al. (2000a, b), respectively. Örgün

et al. (2004) have examined hydrogeochemical properties and radioactivity contents of the tap waters in West Anatolia. Örgün et al. (2005) studied the hydrogeochemical properties and radioactivity contents of tap waters in the region between Sivrihisar and Beylikova towns. They have also investigated the effects of the lithological properties of Kızılcaoren ore deposits and Kaymaz-Sivrihisar plutons on the chemical and radioactive contents of waters. Örgün et al. (2007) have examined the natural and anthropogenic radionuclides in rocks and beach sands at Ezine region (Çanakkale), Western Anatolia, Turkey. Simsek (2008) assessed the natural radioactivity levels in aquifers bearing uranium ore in Koprubasi-Manisa, Turkey. The trace elements and gross α - β values of drinking water resources nearby Kütahya-Tuncbilek power plant have been examined by Özürk and Yilmaz (2000).

As can be followed from the literature study given above, it can be concluded that few studies have been directed to examine lithological-based radioactivity in Turkey. The main goals of the present study were to examine the natural contamination of groundwater with respect to natural radioactivity and trace elements of the host and wall rocks in the aquifers of Eskisehir region (Fig. 1), and to understand the impacts of contamination on the environment. In order to achieve this goal, major ions, trace elements, gross alpha and gross beta analyses were performed on 209 water samples (including thermal water samples) collected from 84 different locations during wet and dry seasons in the study area. The obtained results showed that the 49 of 74 fresh water resources were over the allowable limits for drinking purpose with respect to trace elements (Fe, Al, Ba, As, B, Mn, Zn, and Cr). According to TS 266 (1997), the gross α - β values were over allowable limits in three water resources while gross alpha values exceeded the eligible limit in 15 water resources. Furthermore, gross alpha contents in 20 water resources and gross beta contents of 12 water sources were close to the allowable limits. As expected, the trace elements (B, Fe, As, Mn, Al, Ba, and Zn) were higher in the thermal waters (sample nos. 7, 23, 30, 56, 57, 58, 59, 60, 61, and 76). However, gross α - β values of thermal waters were within the allowable limits according to the Turkish Natural and Mineralized Water Regulation. In four water resources, nitrate content was found to be above 50 ppm (the limit value for adults according to TS 266 2005 and WHO 1993),

and in 36 water sources nitrate was above 10 ppm (limit value for infants according to EPA 2003).

1.1 Geological Setting

The study area attracted many investigators due to its complex tectonic structure, different lithological units, and various mineralogical and geochemical features (Kulaksız 1981; Ercan 1986; Okay and Tüysüz 1999; Delaloye and Bingöl 2000; Arda 1976; Özgenç 1993; Öztürk et al. 1995; Gültekin et al. 1999; Gültekin and Örgün 2000; Gültekin et al. 2003). In the present study, geological–lithological–mineralogical information were mainly based on observations provided by Gözler et al. (1996) and Orgun et al. (2005).

The study area comprises igneous, metamorphic, and sedimentary rocks (Fig. 1) and is located in the collision zone that occurred following the close-up of

the northern part of the Tethys (Gözler et al. 1996). It contains metamorphosed ophiolitic mélange slabs resulting from its abovementioned tectonic position and prevailed deformations. The Sivrihisar, Eskişehir, İnönü, and Mihalıccık metamorphics and the Karkin formations are characterized by Lower Triassic crust material containing slabs of ophiolitic suites and mélanges. These units are overlain by Jurassic–Cretaceous sediments. Upper Cretaceous units are cut by the Topkaya, Karakaya, Y. Karacaoren granodiorites, and the Kaymaz granite. These units are overlain discordantly by Paleocene and Eocene while Miocene and Pliocene overlies all these units as the young sediments.

In the study area, igneous rocks are characterized by granite, subalkaline monzodiorite, gabbro, ophiolitic suite (radiolarite, chert, diabase), and peridotite. Volcanic rocks are characterized by rhyodacite, dacite,

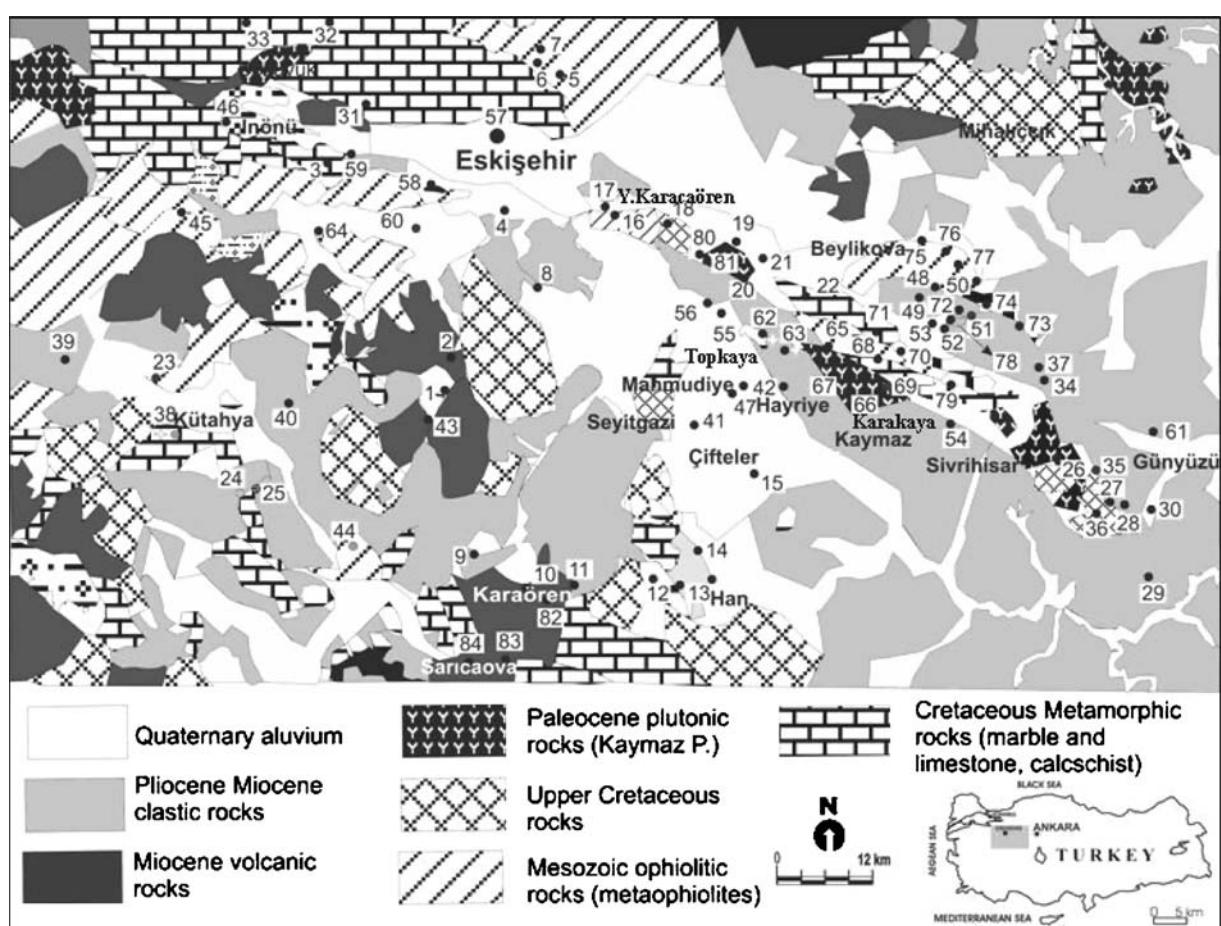


Fig. 1 The geological and sampling location map of the study area (modified from MTA 2002; Gözler et al. 1996; Kulaksız 1981)

rhyolitic tuff, and dacite–andesite; metamorphic rocks by marble, schist, talcschist, and sedimentary rocks by limestone, dolomitic limestone, recrystallized limestone, conglomerate, clayey limestone, marl, and gypsum. Early and Late Miocene volcanic rocks are observed especially around Phrygian Valley, Yazılıkaya, Han, Kirka, and Karaören at the southern part of the study area while Triassic aged metamorphics prevail as the basement rocks at the northern part. The eastern part comprises Jurassic–Cretaceous igneous rocks, Paleogene and Neogene volcano-sedimentary units together with the Kaymaz granite and Sivrihisar monzodiorite. The Sivrihisar and Kaymaz plutons have been developed as a result of the compressional tectonics in Upper Cretaceous–Eocene period. The Kaymaz granite, granite intrusions in ophiolites, ore-bearing hydrothermal alterations, and pegmatites are best observed near Karakaya village. The Kaymaz granite, which has a high silica content, is composed of quartz, orthoclase, oligoclase, biotite, hornblende, zircon, apatite, tourmaline, sphene, pyrite, and magnetite. The Sivrihisar monzodiorite contains the same minerals as Kaymaz granite, except that it has relatively low silica content. In both plutons, radioelements ^{238}U , ^{232}Th , and ^{40}K are common. Activity concentrations of ^{238}U , ^{232}Th , and ^{40}K in both plutons were found to be quite higher than the normal limits suggested by UNSCEAR (1993) (Orgun et al. 2005). Uranium is relatively more enriched in Kaymaz granite than that of Sivrihisar monzodiorite. Malachite, azurite, bornite, and pyrhotite minerals were determined along the dyke of the Kaymaz granite where it is in contact with the ophiolitic suites (Gozler et al. 1996). There are metaophiolitic complexes rich in tremolite–actinolite amphiboles, albite, quartz, chlorite, epidote, sphene, and prehnite minerals at the northern part of Kaymaz town (Erkoyun et al. 2006). Phonolite, basalt, tuff, and agglomerate which resulted from young volcanism (Miocene–Pliocene) can be observed at the southern part of Kaymaz. Complex ore deposits (fluorite, apatite, rare earth elements—REE) are formed as a result of hydrothermal activity along the E–W-trending fault zone where alkaline volcanic rocks are exposed. The phonolite found in the area mainly contains SiO_2 and Al_2O_3 , leucite, nepheline, amphibole, and apatite. It causes the existence of REE depending on carbonatite dykes (Gozler et al. 1996). Tuffs contain thorium, barite, Mn, and REE.

2 Materials and Methods

In June and December of 2005–2006, a total of 209 water samples were collected from springs, wells, and fountains from 84 different water resources, each representing wet and dry spells. Water samples were filtered by 0.45 μm filter paper and acidified to $\text{pH} \leq 2$ by adding 0.5 N HNO_3 and sent to a certified isotope and water chemistry laboratory of the Technical Research and Quality Control Department of State Hydraulic Works of Turkey (DSİ-TAKK). Major ions and trace elements were analyzed by ion chromatography and by KP-M device using the EPA 200-8 method, respectively. The gross alpha and gross beta analyses were performed by Low Level Alpha/Beta Counting System. The analytical errors in the gross alpha analyses (excluding Rn progenies) were $\pm \text{min } 0.002 \text{ Bq L}^{-1}$ and $\pm \text{max } 0.056 \text{ Bq L}^{-1}$. The errors in gross beta analyses were $\text{min } \pm 0.02 \text{ Bq L}^{-1}$ and $\text{max } \pm 0.15 \text{ Bq L}^{-1}$. The error limits increased for the higher gross alpha and gross beta values.

To examine the water–rock interaction, host rock samples were collected from different lithologic units and their petrographical properties were analyzed in the soil analysis laboratory of DSİ-TAKK. The in situ measurements of EC ($\mu\text{mho cm}^{-1}$), pH, and Eh (mV) were performed by the portable instrument YSI-556 Multi-parameters.

3 Results and Discussion

The analysis results of water samples with respect to major ions, trace elements, and gross α –gross β values are given in Table 1. These results have been evaluated in terms of lithology-dependent water quality and the eligibility for drinking water, considering the Turkish Drinking Water Standards (TDWS Code TS 266) (Table 2). Petrographical analyses of the rocks were performed only for those having relatively high radioactive levels (shown in Table 2 in bold sampling numbers).

The detailed evaluation of water chemistry analyses results are presented in the following:

3.1 Major Ions

In most of the water samples, calcium, magnesium, and bicarbonate were dominant ions. Thus, this water

type is representative for the lithology of tuff, granite, mélange, and marble, which are cropped out in the area. As to the eligibility for drinking purpose, nitrate values were over the permissible limit of 50 mg L^{-1} in the four water locations (sample nos. 2, 20, 66, and 67). Since these sampling locations are close to the settlement areas, these waters are contaminated by anthropogenic reasons either from sewage or from agricultural activities. Nitrate values in 36 locations were over the permissible limit (10 mg L^{-1}) for infants (EPA 2003). The consumption of water having high NO_3^- may cause methemoglobinemia (blue baby syndrome) and may link with stomach cancer (Abrahams 2002).

3.2 Trace Elements

The columnar diagrams (Fig. 2) show the appropriateness to the TDWS Code TS 266 in terms of trace element contents of water samples for wet and dry spells (Table 1). Figure 2 indicates that trace elements in water samples in dry spell are higher than those of the samples in wet spell because of dilution factor by precipitation. The areal scattering of trace elements were drawn by means of Surfer™ and shown in Fig. 3. The locations, in which trace elements contents were high (except for locations of 42 and 47) can be attributable to mineralogical composition of the aquifer rocks. Maximum concentrations of trace elements were observed in granitic rocks (sample 64), U–Th hydrothermal alteration zones (samples 48, 49, 50, 51, and 75), tuff (samples 1, 10, 11, 82, 83, and 84), limestone (samples 24, 25, and 44), ophiolitic suite (samples 19, 44, 52, 53, 64, 72, 77, and 78), and boron-containing formations (sample 9). Waters from the Kizilcaoren ore complex have higher fluorite, barite, and REE (samples 48, 49, 50, 51, and 75). The high Mn and Fe contents were determined in phonolitic and pegmatitic rocks where Cr–Fe ore deposits are cropped out (Erkoyun et al. 2006). High trace elements in water sample nos. 66 and 67 were collected around the Kaymaz subalkaline granite containing high Si, Al, magnetite, pyrite, chalcopyrite, F, B, Pb, and Zn. The Kaymaz granite was formed under compressional tectonic regime that occurred later than Upper Cretaceous and earlier than Eocene (Orgun et al. 2005; Erkoyun et al. 2006).

Cu is the highest amongst the other trace elements in water sample nos. 66 and 67 due to exposure of Cu ores (malachite, azurite, and bornite) (Gozler et al.

1996) along the contacts where ophiolitic suites were cut by granite veins in which barite, apatite, zircon, and magnetite are exposed. Furthermore, Cu also enriched in the ryolitic tuffs (samples 82, 83, and 84). The Cr-rich peridotites and an abandoned chromite mine area are found in the catchment area (sample no. 5). The waters from ophiolite were enriched in Mn, Fe, Al, and Cr depending on the lithology of radiolarite, chert, gabbro, diabase, and chromite. Th, U, fluoride, Mn, and REE were abundant in waters circulating within the rhyolitic tuffs (samples 2, 8, 11, 12, 13, 14, 82, 83, and 84).

Lower Miocene aged volcano-sedimentary units contain fluorite, barite, Mn, and REE besides alkaline volcanic products such as silicified tuff, volcanic breccias, and agglomerate. High trace element concentrations in sample nos. 42 and 47, which are the locations close to each other, indicate that these waters most probably come from the same aquifer. The water sample of 47 was taken from a borehole drilled in an abandoned waste disposal area having excessive amounts of Fe and Mn. Fe and Mn can easily be leached in groundwaters due to anoxic environment. Under reducing conditions, oxidation of pyrites might cause the increase in SO_4^{2-} content in nos. 42 and 47. Moreover, humic and pulvic acids, which may be found in such an environment, can play an important role in dissolution of trace elements (Apello and Postma 1992; Hem 1975; Vanloon and Duffy 2000). If humic substance exists in a metal ion complex, this complex can readily be dissolved in oxidative conditions. Accordingly, silicates can be dissolved in acidic water as it happened for water sample no. 84. Apatite (Gozler et al. 1996), existing in granitic rocks, is a source of higher fluorite and arsenic contents in water. Arsenic concentrations of water samples nos. 42 and 47 were determined in higher concentrations than the acceptable limit. Arsenic may increase in water by microbially reducing Fe-oxyhydroxide in aquatic sediments and adsorbed arsenic can be released to groundwater due to oxidative environment. Thus, the reducing conditions in an aquifer facilitate the mobilization of As^{3+} into groundwater. In this case, increasing of arsenic can be observed equivalently with increasing of Fe^{2+} (Hem 1975; Apello and Postma 1992). Accordingly, arsenic in sample nos. 42 and 47 is thought to be released into groundwater by this way. Higher As in drinking water may cause serious illness problems (Abrahams 2002).

54	2005 DP	8	1.05	58.1	5.6	7.14	18.8	184	0.03	0.00	0.00	0.00	0	0.01	0.05		
	2005 WP	11	0.78	66.9	6.5	5.36	13.5	199	0.01	0.00	0.03	0.00	0.02	0.01	0.05		
	2006 DP	8	1.41	56.9	6.6	7.40	16.3	188	0.00	0.00	0.00	0.00	0.00	0.01	0.15		
	2006 WP	6.6	0.98	58.3	5.6	7.50	17.5	171	0.01	0.01	0.39	0.00	0.9	0.29	0.58		
55	2005 DP	146	6.53	202	49.5	53.57	21.1	1197	0.01	0.01	0.39	0.10	0.00	1	0.39	0.11	
	2005 WP	161	7.04	228	58.4	48.21	19	1186	0.01	0.02	0.18	0.10	0.00	0	0.11	0.51	
	2006 DP	158	7.65	232	61.2	58.50	24.2	1220	0.01	0.02	0.18	0.12	0.00	0	0.12	0.87	
56	2005 DP	36	6.89	111	78.1	21.43	29.8	803	0.09	0.00	0.01	0.27	0.09	0.2	0.28	0.02	
	2005 WP	39	7.25	122	91	21.3	25.7	656	0.00	0.04	0.01	0.01	0	0.3	0.06	0.31	
	2006 DP	35	7.59	106	74.6	23.2	24.5	776	0.00	0.00	0	0.1	0.05	0.05	0.14		
57	2005 DP	15	1.17	45.5	2.0	5.714	12.5	272	0.00	0.03	0.00	0.00	0	1.8			
	2005 WP	124	6.83	54.2	27.6	17.1	10.0	596	0.00	0.01	0.24	0.10	0	1.7	0.2	0.42	
	2006 DP	126	6.25	58.1	30.3	15.4	24.0	596	0.00	0.03	0.00	0	1.6	0.10	0.4		
	2006 WP	127	6.78	60.5	32.2	19	33	570	0.00	0.03	0.00	0	1.6	0.09	0.31		
	2005 WP	118	5.99	53	26.2	18.1	28.0	500	0.00	0.00	0	0.2	0.08	0.06	0.13		
59	2005 DP	22	2.38	62.7	31.1	2.86	29.8	373	0.00	0.00	0.06	0.02	0	0.2	0.12	0.02	
	2005 WP	24	2.35	66.1	34.0	3.93	28.5	354	0.00	0.00	0.06	0.02	0	0.2	0.12	0.1	
	2006 DP	24	2.47	69.5	35.3	6.25	36.8	363	0.00	0.01	0.01	0	0.2	0.06	0.29		
	2006 WP	23	2.94	62.1	31	3.57	7.15	328	0.00	0.03	0.00	0	0.5	0.03	0.18		
60	2005 DP	39	2.84	90.5	18.4	32.1	16.5	391	0.00	0.00	0.03	0.02	0	0.4	0.07	0.13	
	2005 WP	42	2.35	90	18.8	31.1	15.0	383	0.00	0.00	0.03	0.02	0	0.4	0.07	0.13	
	2006 DP	41	3.13	94.7	20.4	41.9	21	384	0.00	0.00	0.01	0	0.4	0.10	0.27		
	2006 WP	41	3.18	87.2	17.5	36.4	17.0	337	0.00	0.00	0.01	0	0.4	0.06	0.02		
61	2005 DP	9.3	1.23	50.1	18.5	2.86	4.31	269	0.00	0.00	0.02	0	0.1	0.03	0.05	0.14	
	2005 WP	9.2	1.17	54.7	20.0	3.21	5.0	255	0.00	0.00	0.02	0	0	0.03	0.16	0.25	
61	2006 DP	9.6	1.14	55	21.3	5.25	25.7	257	0.01	0.01	0.01	0	0.1	0.03	0.15		
	2006 WP	7.8	0.97	48.7	18.1	3.43	4.97	233	0.01	0.01	0	0.49	0.00	0.1	0.14	0.2	
62	2005 DP	7	418	2.1	0	75.2	5.2	390	0.48	0.23	18.2	0	0.00	0.03	0	0.02	
	2005 WP	7	415	3.9	0	75.4	5.2	355	0.48	0.24	10.6	0	0.00	0.32	0.00	0.2	
64	2005 DP	6	1,953	116	0	238	50.4	58.9	7.21	1172	0.0	0	0.05	0.03	0.01	0.23	
	2006 DP	8	590	163	3.9	86.2	25.2	709	6.25	363	16.3	0.00	0.02	0.53	0.02	0.1	
	2006 WP	8	596	4.6	4.4	30.8	0.4	12.8	9.5	386	0	0.01	0.06	0.21	0.02	0.09	
65	2005 DP	8	373	2.1	0	64.5	7.04	2.48	1.92	220	17.1	0.15	0.12	0	0.22	0.00	
	2005 WP	8	680	20.0	7.04	93.2	18.2	32.3	48.6	244.0	79.6	0.01	0.16	0	0.71	0.13	0.21
66	2006 DP	7	578	148	14	115	3.3	31.6	32.7	238	77.8	0.00	0.01	0.00	2.03	0.04	0.16
	2006 WP	8	661	14	4.84	93.6	0.1	34.1	34.4	305	0.01	0.02	0.01	2.3	0.01	0.15	
67	2005 DP	7	385	3	0	62.7	8.99	3.19	0.48	226	10.1	0	0	0.48	0.04	0.38	0.11
	2006 DP	7	251	153	5.1	0	40.3	0.9	2.13	33.2	67.1	0.00	0.01	0.00	0.7	0.03	0.05
67	2006 WP	7	242	5.3	4.65	32.6	0.03	8.2	22.1	106	0.08	0	0.00	0.26	0.00	0	0.28
68	2005 DP	7	386	3	0	64.5	7.8	3.0	220	12.7	0.01	0.04	0.02	0.00	0.65	0.08	0.27
	2006 WP	8	378	2.2	4.1	65.2	0.04	7.80	8.66	229	0.04	0.01	0	0.00	0.44	0.00	0
69	2005 DP	7	408	5.1	0	53.3	16.2	3.19	12	206	12.5	0	0.01	0	0.46	0.18	0.27
	2005 WP	8	357	0.9	3.91	62.7	6.1	2.84	6.73	204	8.46	0.01	0.02	0	0.45	0.05	0.23
74	2005 DP	7	391	100	14.1	3.91	0.9	0	158	100	14.1	3.90	0.9	0.00	0.74	0.08	0.20
	2006 WP	8	378	2.2	4.1	65.2	0.04	7.80	8.66	229	0.04	0.01	0	0.00	0.44	0.00	0
72	2005 DP	8	408	5.1	0	53.3	16.2	3.19	12	206	12.5	0	0.01	0	0.46	0.18	0.27
	2005 WP	8	357	0.9	3.91	62.7	6.1	2.84	6.73	204	8.46	0.01	0.02	0	0.45	0.05	0.23
74	2005 DP	7	391	100	14.1	3.91	0.9	0	158	100	14.1	3.90	0.9	0.00	0.74	0.08	0.20

Table 1 (continued)

S. no	Period	pH	EC	Eh	Major ions						Trace elements										Radioactivity				
					Na mg L ⁻¹	K mg L ⁻¹	Ca mg L ⁻¹	Mg mg L ⁻¹	Cl mg L ⁻¹	SO ₄ mg L ⁻¹	HCO ₃ mg L ⁻¹	NO ₃ mg L ⁻¹	b mg L ⁻¹	Zn mg L ⁻¹	Cr mg L ⁻¹	Mn mg L ⁻¹	Fe mg L ⁻¹	Cu mg L ⁻¹	Al mg L ⁻¹	As mg L ⁻¹	B mg L ⁻¹	Si mg L ⁻¹	Ba mg L ⁻¹	G. alpha Bq L ⁻¹	G. beta Bq L ⁻¹
75	2005 DP	8	458	22	0	38.7	24.3	5.67	17.8	234	1.51	0.02	0.29	0	0.01	0.31	0.43	0.33	0	0	16	1.47	0.11	0.33	
	2006 DP	8	461	119	23	0	54.1	14.7	4.61	18.8	256	1.86	0.00	0.03	0.03	0.32	0.81	0.03	0.00	0	0	11	1.02	0.06	0.24
	2006 WP	9	441	27	4.56	32.2	0.2	21.0	22.0	187	0.02	0.02	0.00	0.01	0.37	0.01	0.32	0	0.1	0.1	14	1.47	0.10	0.26	
76	2005 DP	7	1,113	38	3.91	94.8	77.7	14.9	41.8	706	5.49	0.01	0.25	0	0.00	0.63	0.4	0.16	0.9	0.9	24	0.6	0.04	0.45	
77	2006 DP	8	431	0.9	3.91	79.2	6.9	2.48	7.69	265	31	0.15	0.08	0	0.00	0.93	0.1	0.34	0.00	0	0	4.5	0.2	0.01	0.1
79	2005 DP	8	608	13	3.91	46.9	45	6.73	23.1	338	36	0.01	0.28	0	0	0.34	0.45	0.11	0.00	0	0	21	0.51	0.03	0.15
	2005 DP	7	588	5.1	3.91	96.6	9.7	10.6	1.92	305.0	9.35	0.19	0.06	0	0.00	0.9	0.06	0.25	0.00	0	5.5	0.22	0.03	0.23	
80	2005 DP	8	417	39	0	49.1	12.4	7.75	50.5	229	43.5	0.00	0.01	0	0.00	0.63	0.05	0.28	0	0	11	0.33	0.01	0.13	
81	2005 DP	8	517	56	0	60.9	9.7	25.9	65.4	212	1.28	0.15	0.15	0	0.01	1.67	0.11	1.55	0	0.1	13	0.29	0.03	0.14	
82	2005 DP	7	272	13	10.9	39.7	4.1	5.67	20.7	150	7.97	0.1	0.32	0	0	0.51	0.75	0.18	0	0.0	0	31	0.17	0.33	0.44
	2006 DP	7	274	57.9	8	0	43.1	0.6	5.32	2.89	145	6.33	0.00	0.01	0.01	0.00	0.69	0.01	0.00	0	0	36	0.01	0.01	0.38
	2006 WP	8	274	7.9	4.89	33.0	0.1	11.4	7.27	157	0.1	0.01	0	0	0.2	0.04	0.06	0	0	0	32	0.17	0.35	0.32	
83	2005 DP	7	145	3	3.13	23.6	0	2.13	10.1	70.8	0	0.5	0	0	0.41	1.03	0.95	0	0	0	257	0.16	0.02	0.17	
84	2005 DP	6	84	8	0	10	0.6	1.77	13.9	34.2	1.82	0	0.24	0	0	0.37	0.39	0.72	0	0	0	169	0.18	0.04	0.21
	2006 DP	6	92	169	3.9	0	9.02	5.8	1.77	26.9	29.9	0.00	0.01	0.01	0.00	0.21	0.04	0.04	0.00	0.00	0	18	0.01	0.06	0.17
	2006 WP	7	82	5.7	4.48	6.2	0.02	8.9	14.2	27.5	0	0.03	0	0.00	0.45	0.03	0.71	0.00	0.1	0.1	19	0.18	0.05	0.23	
Maximum Contamination Levels (MCLs) of nitrate and trace elements recommended by TS-266 1997 and 2005					50	10	5	0.05	0.05	0.2	3	0.2	0.01	1	0.3	0.1	1.0								

WP wet period, DP dry period

Aluminum (Al), which are found in most of the samples, mainly come from acidic igneous rocks enriched in alumino-silicates namely granite, tuff, and rhyolitic tuff outcropping in the study area. Soil acidification may cause increasing solubility and leaching of Al. Medical scientists have revealed that the higher Al content in drinking water has been conducted with the development of Alzheimer's disease (Abrahams 2002). High barium (Ba) was observed in water samples (nos. 11, 48, 49, 50, 66, 67, 75, 80, and 81) interacted with granite, rhyolitic tuff, ophiolitic suite (especially in zeolite mineral), and marble rocks.

The Kizilcaoren village is the most significant fluorite ore deposit in Turkey which formed by hydrothermal alteration. Fluoride concentrations in groundwater vary from 4.6 to 9.2 mg L⁻¹ and lead to endemic fluorosis near the Kizilcaoren village, and fluorine (F) level (Fidancı et al. 1994) in plant samples was reported up to 658.9 mg L⁻¹. On the other hand, the maximum permissible level of F is 1.5 mg L⁻¹ for drinking water, which is recommended by the TS 266 (1997) and WHO (1993). Fluorine concentrations in plants picked from Kizilcaoren village show a relationship with that of soil, and serious fluorosis symptoms have been seen in sheep living around the Kizilcaoren (Fidancı et al. 1998; Uslu 1982; Kirikoglu 1988).

3.3 Radioactivity

Gross alpha and gross beta activities were measured to determine natural radioactivity levels of water (Table 1). As it is well known, the gross alpha activity is presented by ²³⁸U series and gross beta activity is presented by ²³²Th series and ⁴⁰K (Abdel Hady et al. 1994; Saied et al. 1994; Abdel-Monem et al. 1996; El-Shershaby 2002). Bedrock, soil, and water are in interaction with each other. As a result of this interaction, some radionuclides can penetrate into groundwater from bedrock and soil during their percolation from surface into aquifer. Gross alpha and gross beta in water are mostly controlled by the lithology of granitic and volcanic rocks in the study area. Accordingly, high gross α and gross β values in the water samples of the present study mostly depend on the host and wall rock composition consisting of granite, tuff, rhyolitic tuff, pegmatite, phonolite, limestone, and marble. Thus, waters coming from granitic and volcanic rocks have been naturally

Table 2 Chemical analyses of water samples and mineralogical properties of some selected high-radioactive rocks

	Sample no.	Sample name	Rocks	Minerals	MCL	RCL
Springs	1	Kalabak	R. tuff		Al	
	4	Kalabak bottled water				Al, alpha
	15	Sakarbaşı	Conglomerate	Fe		As, Zn, alpha
	19	Aktepe	O. mélange	Fe, alpha		Al, Ba, Cr, beta
	21	Kosmat	Marble		Excellent	
	23	Yoncalı thermal	Grovak	Fe, As		
	25	Porsuk Çiftliği	Limestone	Fe		Al, As, Ba, Cr
	26	Dinek	Granite			Fe, alpha
	27	Atlas	Marble			Fe, alpha
	34	Nasrettin Hoca	D. limestone		Excellent	
	35	Gecek	Granite			Fe
	37	Babadat	Marble	Ba, Fe		Al, Cu, Zn
	38	Sofu	Marble		Excellent	
	39	Aliköy	Limestone	Fe		Al, As, Ba, Cr
	45	Muratdere	Marble		Excellent	
	46	Karasu	Marble		Excellent	
	52	Karkin Ali	O. mélange	Al, Ba, Fe		
	56	Uyuzhamamı thermal	O. mélange	Fe		As, Zn, Mn, Al
	58	Kızılınlı thermal	Silicified cong.	Fe		As, B, Mn, Al
	59	İnönü thermal	D. limestone			As
	60	Eskar thermal	Tuff			As, Al
	61	H. karahisar thermal	Conglomerate			
	62	Balcıkhisar	Marble		Excellent	
	64	Kümbet	O. mélange	Fe, Cr		As, alpha
	67	Karakaya	A. granite	Al, Ba, Fe, alpha		As, Cr, Pb, beta
	69	İlipinar Okçu	Schist+marble	Ba, Fe		Al, Fe
	70	Okçu Soğukpinar	Schist+marble		Al, Ba, Fe, Cu, Zn	
	71	Okçu Sakarya	D. calcschist	D+Qtz+Ms	Al, Fe	Ba, Cr, Cu, Zn, Pb, alpha
	73	Dümrek spring	Marble		Al, Ba	Fe
	74	Zeyköy spring	Marble (serisit, klorit)		Fe	Al, Ba
	76	Yalınlı Dogrul	O. mélange		Fe, Ba, As	Zn, Cu, B, Al
	77	Dumluca	O. mélange		Al, Fe	Ba
	79	Sarıkaynak–Akpinar	Marble		Al, Fe	Ba, Pb
	82	Karaören–Kirka Hatap	R. tuff		Fe, alpha	Al, As, Ba, Cr, Cu, Zn, Pb, beta
	84	Sarıcaova–Çitbaşı	Silicified R. tuff	Qtz+Sa+Pl+Bt	Al, Fe	Ba, Cr, Cu, Zn, alpha
Fountains and reservoirs	3	İnönü	D. limestone			Excellent
	5	Bozdag (Kulahmet)	O. mélange	Qtz+Px+Hbr+Chr		Cr
	6	Yayla	O. mélange			
	11	Yazılıkaya	R. tuff	Qtz+Sa+Pl+Bt	Al, As, Ba, Fe, alpha, beta	Cu, Zn
	12	Aglarca	Calcschist–marble		Al, As, Ba, Fe	Zn
	13	Han	R. tuff	Qtz+Pl+Bt	Fe	Al, Ba, Cu, Zn
	16	Kulahmetli	Peridotite			Excellent
	17-A	Yörük Karacaören	Granite		Al, Ba, Fe	Cr, alpha
	18	Gökceoğlu	Schist			Excellent
	20	Uyuz Hamam	Granite	Fe, alpha		Ba, Cr, beta
	22	İkipinar	Calcschist–marble	Alpha		Fe
	24	Gelinkayası	Limestone	As, alpha		Fe
	31	Zemzemiyə	Marble		Excellent	

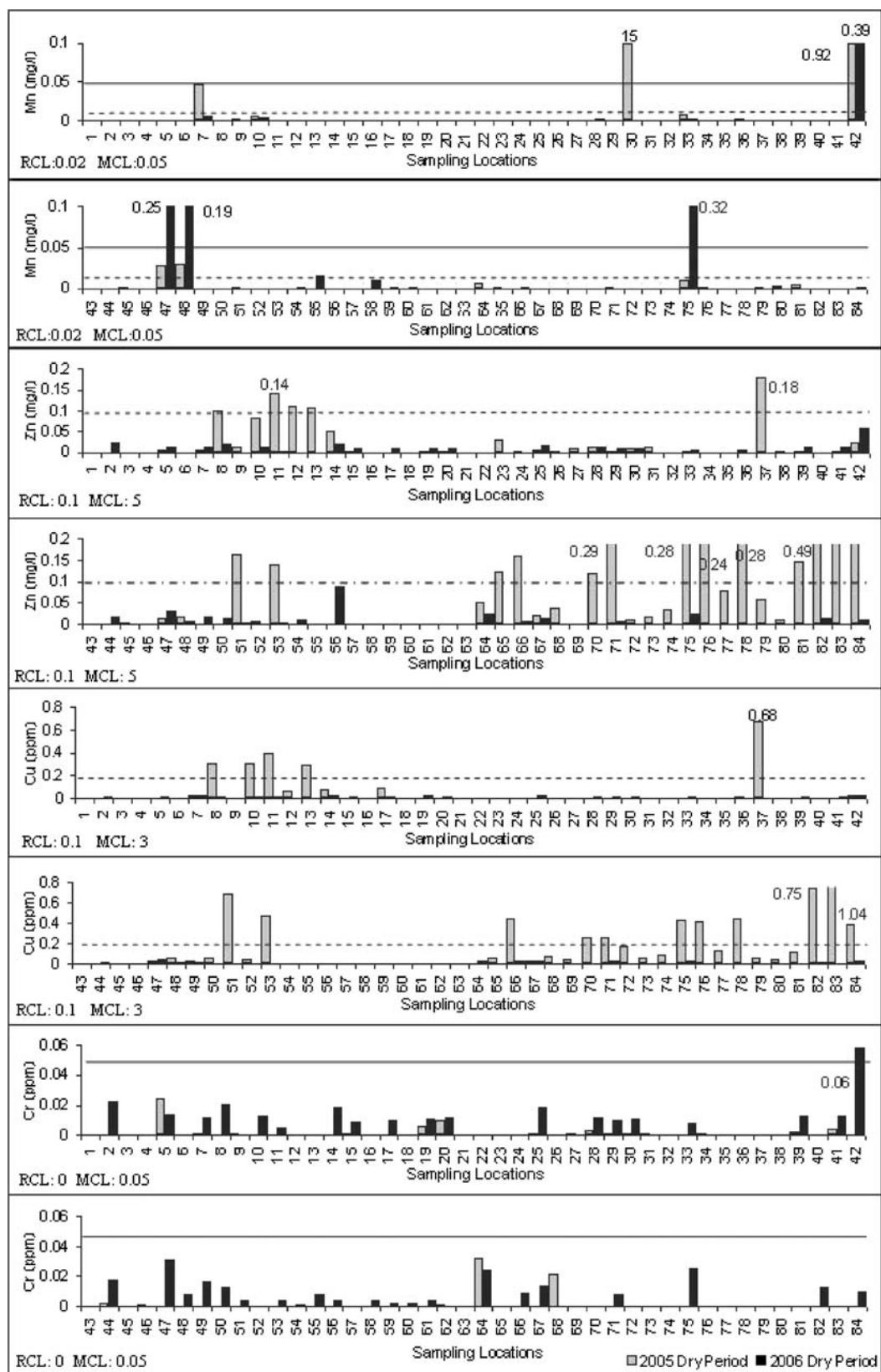
Table 2 (continued)

	Sample no.	Sample name	Rocks	Minerals	MCL	RCL	Depth (m)
Wells	32	Sögüt	Marble		Fe, alpha		
	33	Küre Asmalı	Calcschist–marble		Fe	Ba, Cr	
	40	Perli Killik	Limestone			Fe, alpha	
	43	Kütahya–Güllüdere	R. tuff		Mn	As, Fe, alpha	
	49	Yayla	U+Th ore deposit	Qtz+Cal+Pl+Fl+Brk	Ba, Fe, alpha	Cr, beta	
	50	Adalet	U+Th ore deposit		Ba, Fe, alpha	Al, Cr, beta	
	51	Karapınar	U+Th ore deposit		Al, Ba, Fe, alpha	Cu, Zn	
	53	Kayacık	O. mélange		Ba, Fe	Cu, Zn, alpha	
	68	Kutahya Okçu Köyü	Schist+marble		Al	Ba, Fe, Cr	
	72	Karkin Yayla	O. mélange			Ba, Fe, Cu	
	75	Kızılıcaören Dam	Schist+C. schist+U		Al, Ba, Fe, alpha	As, Cr, Cu, Zn, Pb, beta	
	83	Kırka-Zöhre Uslu	R. tuff		Al	Fe, Cu, Zn	
	2	Y. Kalabak	R. tuff			Alpha, As, Ba, Fe, Cr, beta	85
	7	S. İlica thermal	O. mélange		B, Fe, Mn, As		100
	8	Derbent	R. tuff	Pl+Bt	Mn	As, Fe, Cr, Cu, Zn, alpha	100
	9	Kırka	Tuff		B	As, Ba, Fe	187
	10	Karaören	R. tuff	Qtz+Pl+Bt	Alpha, beta, Fe	Ba, Cr, Cu	170
	14	Han	R. tuff	Qtz+Pl+Bt	Al, Ba, Fe, alpha	Cr	100
	28	Kayakent	Marble		Fe	Al, Cr, alpha	93
	29	Yeni Çıktı no. 56969	Marble		Fe	Cr, alpha	170
	30	Gümüşkonak thermal	Gypsum–marl	Gp+Qtz+Chl+Cal	Mn, Fe, Al	Cr, As	215
	36	Kuzören	Marble			Excellent	100
	41	Hayriye	Clay limestone+tuff		Fe	Ba, Cr, alpha	95
	42	TJK	Marble		As, B, Fe, Cr, Mn	Al, Ba, Cu, beta, alpha	63
	44	Ahiler no. 57177	Of. (granit+aplit)		Ba, Fe	As, Cr, Cu, alpha	83
	47	Mahmudiye	Marble		As, B, Fe, Mn, alpha	Cr, beta	144
	48	Kızılıcaören	U+Th ore deposit		Al, Ba, Fe, Mn, alpha, beta	Cr	300
	54	Sivrihisar no. 49863	Conglomerate			Excellent	175
	55	Uyuzhamam	Granite		Alpha	Beta, Al, B, Fe	40
	57	Eskişehir thermal	Limestone		B	Al, As	110
	63	Balcıkhısar	Calcschist–marble			Excellent	118
	65	Kaymaz	Marble		Al	Ba, Fe, Zn, Pb	51
	66	Karakaya	A. granite	Fl+Qtz+AM	Ba, Fe, alpha	As, Cr, Cu, Zn, beta	178
	78	Karkin	O. mélange		Ba	Al, Fe, Cu, Zn	60
	80	Alpu no. 57887	Marble		Al, Fe, Ba	As	232
	81	Kelkaya no. 57888	Marble		Al, Fe, Ba	As, Zn, Pb	191

R rhyolithic, O ophiolite, D dolomite, C calc, Qtz quartz, Ms muscovite, Fl feldspar, Sa sanidine, Bt biotite, Pl plagioclase, Gp gypsum, Chl chlorite, Cal calcite, Px pyroxene, Hbr hornblende, Chr chromite, Brk brookite, AM accessory minerals (zircon, apatite, sphene, tourmaline, pyrite), MCL maximum contamination level, RCL recommended contamination level

contaminated in terms of radioactivity and trace elements due to the dissolving of the minerals such as quartz, feldspar, and mica. These rocks may also contain fluorine, boron, and some trace (Pb, Zn, Cu,

Fig. 2 Columnar diagrams of trace elements of water samples for the 2006 dry spell (MCL maximum contamination level, RCL recommended contamination level). (2005 Dry Spell; 2006 Dry Spell; MCL RCL, values exceeding the scale have been marked on the top of the bars)



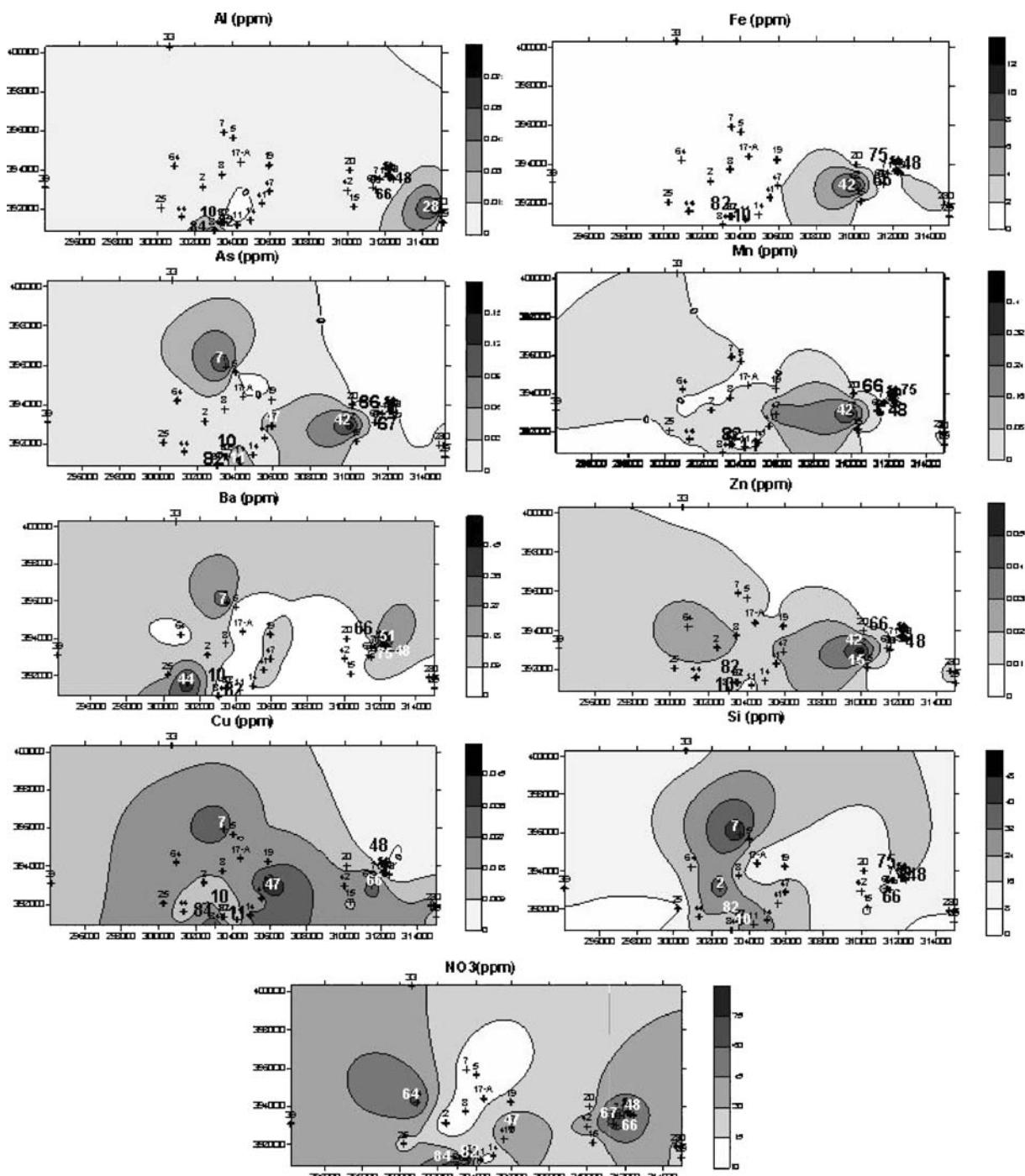


Fig. 3 The areal distribution of trace elements (for 2006 dry spell)

Hg, etc.) and radioactive elements (U, Th, Rn, Ra) having over the allowable limits for drinking water (Banks et al. 1995a,b; Apambire et al. 1997; Banks et al. 1998; Örgün et al. 2004). However, the lower

gross alpha and gross beta values have also been seen in marble and limestone besides in dolomitic limestone, ultramafic (peridotite), and ophiolitic melange due to their lithology (Gascoyne 1992). Fluorine-

contained waters, from the vicinity of Kizilcaoren village, have higher uranium content. Accordingly, the waters with high F^- in Kizilcaoren ore complex may increase the solubility of U. On the other hand, the solubility of U increases with carbonate content if Eh (oxidation-reduction potential) is high enough (Gascoyne 1992). The water samples having high Fe content show a high linear relationship with gross α values (Fig. 4) but the linearity was low in the relationship with gross β values. The strong relationship between gross alpha–beta and Eh values can only be seen if the radioactivity levels of the water samples are high (Fig. 5). Waters circulating in dolomitic limestones (samples 34, 45, and 46) have lower radioactivity because U is depleted in the rock during dolomitization process (Gascoyne 1992). On the other hand, no correlation was observed among gross alpha and beta values with SO_4 , Cl, Ca, and HCO_3 contents.

Figure 6 shows the percentage distribution of gross alpha and gross beta values in the 209 water samples. It can be seen that 17% and 3% of samples exceeded the permissible limits for gross α (0.1 Bq L^{-1}) and for gross β (1 Bq L^{-1}), respectively. The radioactivity levels of waters were examined by TS 266 (1997)

(Turkish Drinking Water Standards) and WHO (1993) standards with respect to the suitability for drinking purpose for the wet and dry periods (Fig. 7). This figure reveals that maximum permissible levels are exceeded in 18 water resources for gross alpha and in three water resources for gross beta. The high gross beta values in samples 11, 48, 49, 50, 66, 67, 75, and 82 are consistent with their high potassium contents which may come from feldspar minerals (orthoclase, microcline, and sanidin). Generally, U-Th content increases with silica content and are found in the accessory minerals of granitic rocks (Orgun et al. 2005). Accordingly, in this study, high levels of gross alpha and beta values were found in samples 56, 66, 67, 82, and 84.

Gross α and gross β values of dry spells were higher than those of wet spells possibly because of the two reasons: (1) dilution effect in the wet spell, (2) undercontrolling of reducing conditions in the dry spell. The spatial distributions of gross alpha and beta values show that (Fig. 8) the highest gross α and gross β values accumulated mainly in the three districts. These districts are located nearby the residential areas, namely: (1) Kırka, Karaören, Yazılıkaya, Han (nos. 10, 11, 14, 64, and 82, 84—

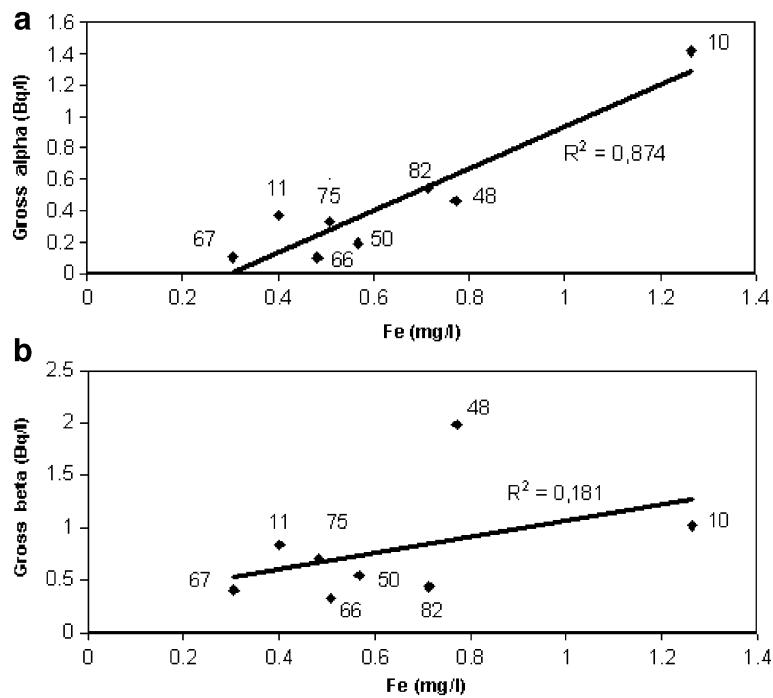


Fig. 4 Gross alpha (a) and gross beta (b) vs. Fe content (for the 2005 dry spell)

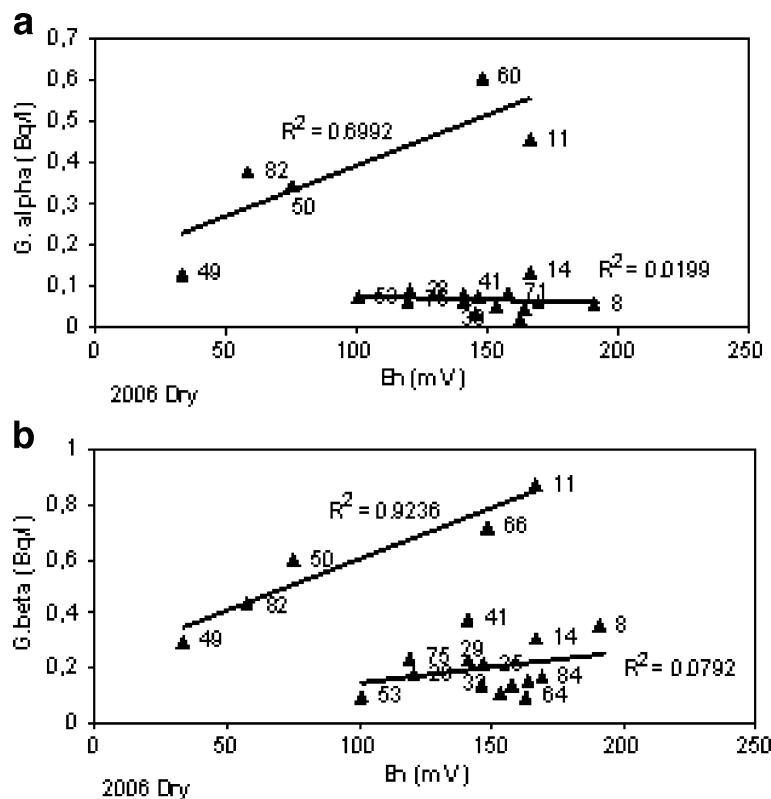


Fig. 5 Gross alpha (a) and gross beta (b) vs. Eh (for the 2006 dry spell)

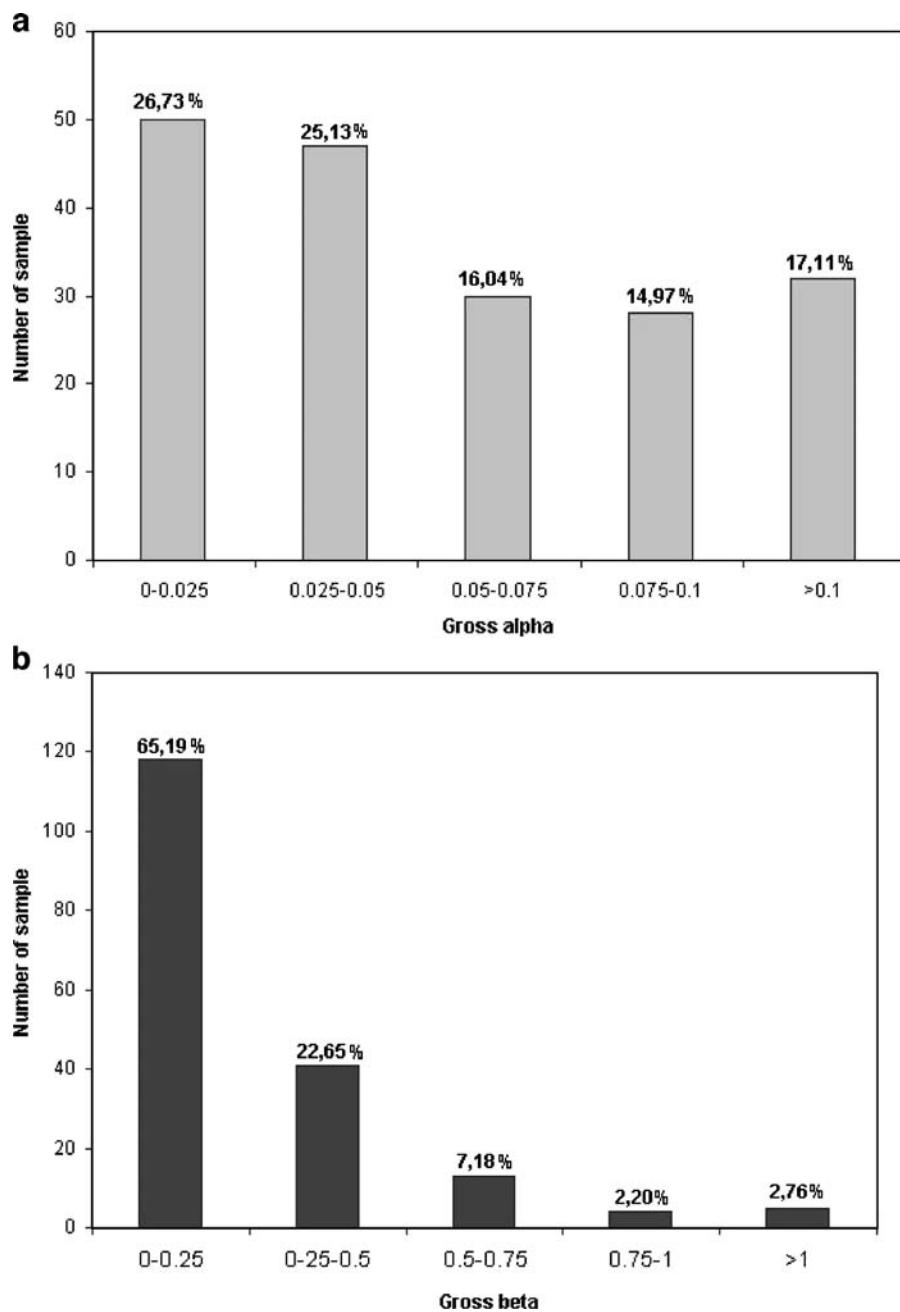
water samples from volcanic rocks); (2) Kaymaz Pluton, Karakaya, and Kizilcaoren (nos. 48, 49, 50, 51, 66, 67, and 75—water samples from granite, phonolite, and pegmatite rocks); (3) Hayriye and Mahmudiye (nos. 47 and 42—water samples from an abandoned waste disposal area). Table 3 shows the problematic water districts in terms of trace elements and radioactivity. It must be pointed out that waters belonging to these districts can be used for drinking purposes only if each progeny of U and Th series is found suitable for human health.

4 Conclusions

The aim of this study was to investigate the natural pollution of groundwater, utilized for drinking and domestic purposes, originated from lithologic properties of rocks (i.e., minerals) and to recognize its environmental effects around Eskisehir region. This research revealed that the trace element concentrations (Al, Fe, Mn, Cr, Ba, Zn, Pb, Cu, B) in 49

of 74 fresh water resources are above the limits of Turkish Drinking Water Standards (Code TS 266). The relatively high concentrations of trace elements in the water samples mostly originated from dissolution of minerals, which are contained in the lithology of aquifer or the surrounding rocks. On the other hand, samples nos. 42 and 47 taken from the boreholes have higher trace element contents because these wells were drilled in an abandoned waste disposal area having reductive conditions that may cause the increase in the solubility of trace elements in aquatic environment. Granite, rhyolitic tuff, tuff, phonolite, pegmatite, and ophiolitic suite are the main natural (lithologic) sources causing the increase of trace elements in water due to water–rock interaction. Other lithologic sources for the natural contamination of groundwater can be considered as the ore deposits produced by hydrothermal alteration around the Kaymaz plutonic rocks and ore complex found nearby the Kizilcaoren village which is characterized by thorium, uranium, barite, fluorite, and REE. Furthermore, nitrate value

Fig. 6 The percent distribution of gross alpha and gross beta



was found above the limit (50 mg L^{-1}) in four locations (sample nos. 2, 20, 66, and 67) and above the limit for infants (10 mg L^{-1} ; EPA 2003) in 36 locations. High nitrate values in those samples did not originate from lithological reason but anthropogenic and agricultural activities. The hazardous effects of the high F content in water derived from Kizilcaoren ore complex to environment (plant,

water, soil, etc.) were reported by the previous study (Fidancı et al. 1996).

Both gross alpha and gross beta values in three water resources were found over the limits as only gross alpha values were relatively high in 15 water resources. Thus, a total of 18 water resources were evaluated as over the standard limits for drinking water in terms of radioactivity. Moreover, the radio-

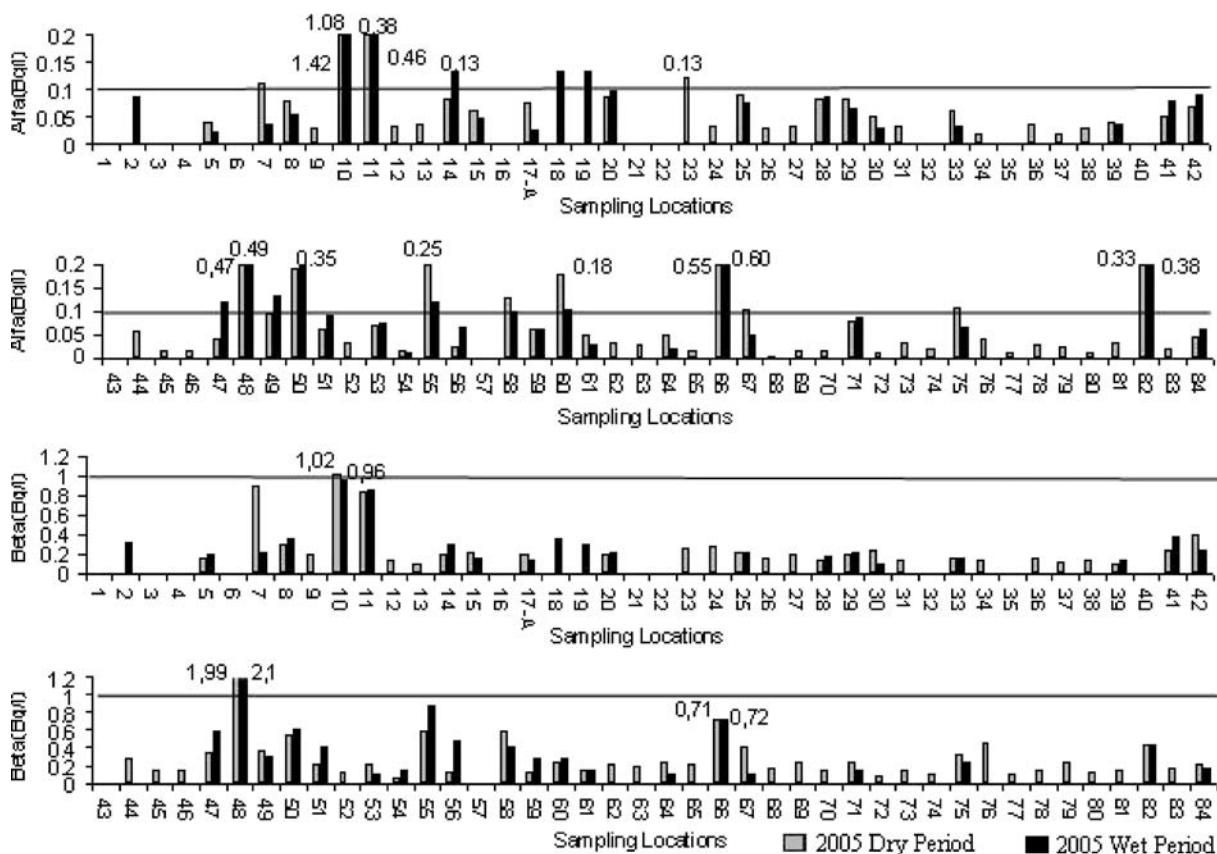


Fig. 7 Columnar diagrams of gross alpha and gross beta for the 2006 dry spell. (2005 Dry Period; 2006 Dry Period; MCL RCL, values exceeding the scale have been marked on the top of the bars)

activity values were found close to the limits in 20 locations. It is obvious that the higher gross alpha and gross beta values are sourced from the aquifer lithology. Most probably, radioactivity increased in these water resources due to the long residence time of groundwater in aquifers consisting of granite, rhyolitic tuff, pegmatite, phonolite, and marble. Water localities having highest gross alpha and gross beta values can be counted as Kırka–Karaoren–Yazılıkaya region where rhyolitic tuff is cropping out (samples 10 and 11), Kızılcaoren ore complex, and thorium–uranium mines (samples 48, 49, 50, 51, and 75). Furthermore, radioactive enrichment was also determined in the well waters from an abandoned waste disposal area (no. 42 and no. 47) due to the reductive conditions.

In conclusion, the waters containing high radioactivity and trace elements can be classified under three main districts depending on their lithology: (1)

water samples from volcanic rocks such as tuff, rhyolitic tuff, andesite, dacite (Kırka, Karaören, Yazılıkaya, Han); (2) water samples from granite, phonolite, pegmatite rocks (Kaymaz Platon, Karakaya, and Kızılcaoren); (3) water samples from the wells drilled in an abandoned waste disposal area for providing domestic water to the residential areas (Hayriye and Mahmudiye). These three water districts can be utilized for drinking purposes either by dilution with water having low content of radioactivity or by purification of trace elements. It is clear that the waters in these three districts require further analysis to determine each progeny of U and Th series in order to make an environmental risk assessment (Vesterbacka 2005).

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Fig. 8 The areal distribution of gross alpha and gross beta for the 2006 dry spell

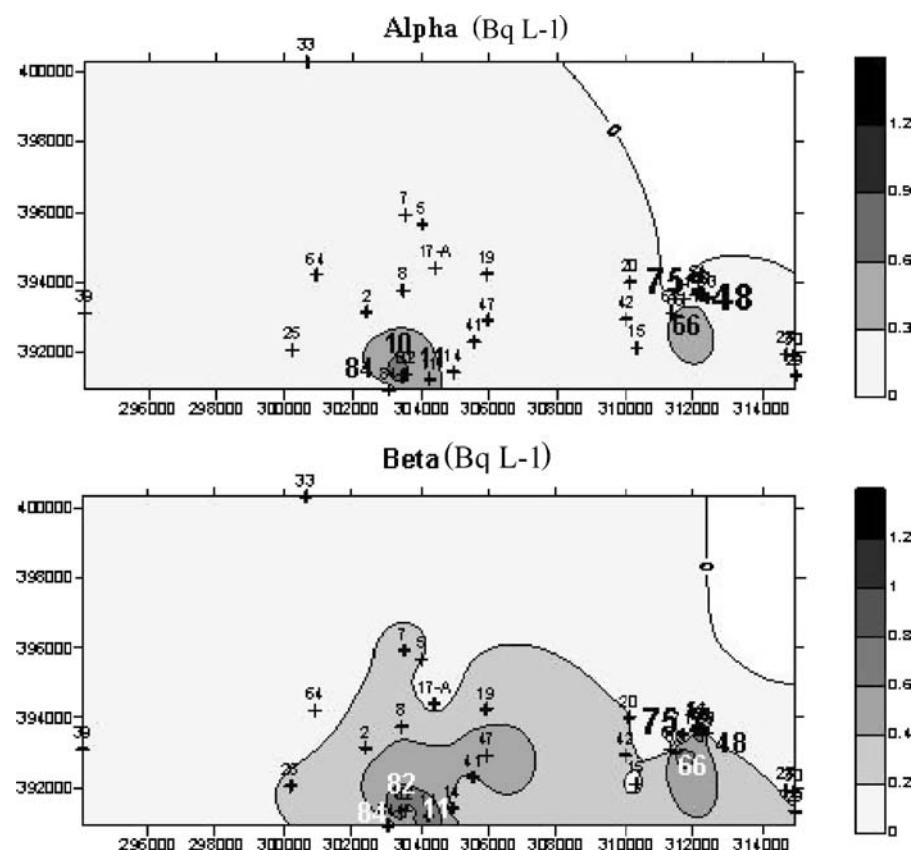


Table 3 Problematic water districts in terms of trace elements and radioactivity

District no.	Sample no.	General lithology	Type	Well depth (m)	Trace element content exceeding the limits (mg L^{-1})	Max. alpha (Bq L^{-1})	Max. beta (Bq L^{-1})	Any filtration before using	Currently used as drinking water
1	10	R. tuff	Well	170	Fe	1.42	1.02	No	Yes
	11	R. tuff	Fountain	—	Al, As, Ba, Fe	0.46	1.76	No	Yes
	14	R. tuff	Well	100	Fe, Ba, Cr	0.13	0.31	No	Yes
	64	O. mélange	Spring	—	Fe, Cr	0.07	0.23	No	Yes
	82	R. tuff	Spring	—	Fe	0.38	0.44	No	Yes
	84	R. tuff	Spring	—	Al, Fe	0.06	0.23	No	Yes
2	48	U+Th+OD	Well	300	Al, Ba, Fe, Mn	0.52	2.10	No	No
	49	U+Th+OD	Fountain	—	Ba, Fe	0.16	0.36	No	Yes
	50	U+Th+OD	Fountain	—	Ba, Fe	0.35	0.60	No	Yes
	51	U+Th+OD	Fountain	—	Al, Ba, Fe	0.09	0.41	No	Yes
	66	A. granite	Well	178	Ba, Fe	0.60	0.72	No	Yes
	67	A. granite	Spring	—	Al, Ba, Fe	0.10	0.41	No	Yes
	75	Schist+U	Fountain	—	Al, Ba, Fe	0.42	0.33	No	Yes
3	42	Marble	Well	63	As, B, Fe, Cr, Mn	0.09	0.51	No	Yes
	47	Marble	Well	144	As	0.12	0.66	No	Yes

U uranium, Th thorium, OD ore deposit

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