Turk J Engin Environ Sci
24 (2000) , 229 – 236.

Uranium, Radium and Field Measurements in the Water of Gediz River

M. BAKAÇ
Dokuz Eylül University, Faculty of Education,
Department of Physics, Buca, İzmir-TURKEY
M. N. KUMRU
Ege University, Institute of Nuclear Sciences, Bornova, İzmir-TURKEY

Received 17.03.1999

Abstract

Gediz River drains a large area of the central Aegean region of western Turkey. The geological structure contains mostly metamorphic and volcanic rocks of the Palaeozoic, Mesozoic and Neogene ages. Water samples were collected from the river at 1-km intervals. The temperature, pH, Eh, specific conductivity, total alkalinity of the water, and beta and gamma activity in the vicinity of river were measured. The concentrations of radium and uranium in the water of Gediz River were determined by the collector-chamber and fluorimetric methods, respectively. The radium and uranium concentrations ranged from 0.021 to 1.041 Bq/l and from 0.4 to 10.4 ppb, respectively. Furthermore the temperature, pH, Eh, conductance, alkalinity of the water and gamma activity also varied between 10 and 30°C, 5.55 and 8.28,-108 and 36 mV, 8 and 5940 μS/cm, 94 and 750 mg/l and 56 and 176 cps, respectively. The reason for different radium values is probably the different geological structures in these three zones. The aim of this study was to determine the level of natural radioactivity in the Gediz Basin along the river, and whether this radiation causes any danger to health.

Key Words: Gediz River, geological structure, natural radioactivity, Turkey.

Gediz Nehri Sularında Uranyum, Radyum ve Saha Ölçümleri

Özet

Batı Türkiyedeki Gediz nehri merkez Ege Bölgesindeki geniş bir alanı drenaj ederek akmaktadır. Bölgenin jeolojik yapısı genellikle Paleozoik, Mezozoik ve Neojen çağlı metamorfik ve volkanik kayaçlardan oluşmaktadır. Su örnekleri nehirden 1-km aralıklarla toplandı. Sularda sıcaklık, pH, Eh, öz iletkenlik, toplam alkalinite ve nehir kenarında beta ve gama aktiviteleri ölçüldü. Suların içeriğindeki radyum ve uranyum konsantrasyonları sırasıyla kollektör odası ve fluorometrik yöntemler ile belirlendi. Radyum ve uranyum konsantrasyonları sırasıyla 0.021 1.041 Bq/l, ve 0.4 - 10.4 ppb aralığında değişim gösterdi. Bundan başka sularda sıcaklık, pH, Eh, iletkenlik, alkalinite ve gama aktivitesi sırasıyla 10-30°C, 5.55-8.28, -108 - +36 mV, 8-5940 μS/cm, 94-750 mg/l ve 56-176 cps aralığında değişim gösterdi. Radyumdaki farklı değerlerin sebebinin bu üç bölge arasındaki jeolojik yapıdan kaynaklandığı sanılmaktadır. Bu çalışmanın amacı, nehir boyunca Gediz havzasındaki doğal radyoaktivite seviyesini belirlemek ve bu radyoaktivitenin sağlık açısından herhangi bir tehlike oluşturup oluşturmadığını tayin etmektir.

Anahtar Sözcükler: Gediz Nehri, jeolojik yapı, doğal radyoaktivite, Türkiye.

Introduction

^{226}Ra is often used as a standard for the evaluation of contamination by transuranic elements. The ^{226}Ra in the environment is widely distributed, being present in various concentrations in waters, soils, sediments and rocks. The quantities of natural radionuclides in water depend upon the quality of the parent rock. The radioactivity of natural radionuclides in granite is somewhat higher than in other rocks.

Uranium and radium belong to the group of primordial radionuclides, as they have always been present in the earth. The radionuclides ^{238}U, ^{235}U and ^{232}Th, which decay through three distinct series of radionuclides, are of great importance in the nuclear fuel cycle. Not only are they present in the human body and foodstuffs but some gaseous radionuclides may also be inhaled. ^{226}Ra (half-life 1600 years), ^{224}Ra and ^{228}Ra (half lives 3.6 days and 5.8 years, respectively, and both usually mixed with uranium ore) are of radiological importance because radium behaves chemically like calcium, being deposited on bone surfaces and areas of mineral metabolism (Evans, 1974).

It is known that interactions between rivers and seawaters, which take place in estuaries, are very complex. Thus, the trace element inputs from rivers to the sea can be seriously affected by processes which take place across the gradient of salinity produced in the mixing area of the estuary (Martinez-Aguirre and Garcia-León, 1993).

The determination of uranium concentrations in geological samples is very important in the exploration of the natural resources of this element. Of these geological samples, natural waters have special importance as indicators of uranium anomaly, not only at the surface, but also at deeper levels because its anomaly in the soil and the rocks should show up as an increase in the uranium concentration of groundwater (Bolivar et al., 1979).

Brown et al. (1983) found uranium concentrations of 0.5-1.0 ppb in the waters of South Greenland. In his survey of radioactivity in Büyük Menderes River, Turkey, Kumru (1995) detected uranium and radium concentrations of 0.24-17.65 ppb and 0.016-0.751 Bq/l respectively. In another study, it was found that the mean ^{226}Ra concentration in the Tejo and Douro Rivers respectively was 0.0162 ± 0.0067 Bq/l and 0.0198 ± 0.0087 Bq/l (Carreiro and Sequeira, 1988). A source of ^{226}Ra was discovered by Cassels (1990) during a study of radium in the natural water supply in Kakadu National Park. Bolivar et al. (1983) conducted a hydrogeochemical and stream-sediment reconnaissance in the Montrose quadrangle (Colorado) and determined that the uranium concentration in waters there varied within the range 0.02-856 ppb.

In this paper, some physical and chemical parameters of the water of Gediz River were measured and measurements of uranium, radium and gamma activity of the area for the natural radioactivity were determined. Our aim was to determine the level of the natural radioactivity in Gediz Basin along the river, and determine whether this radiation causes any danger to health.

1. Materials and Methods

1.1. Physiography and geology of study area

Gediz River is one of the main rivers (~401 km long) of the Aegean area in Turkey. Fig.1 shows a geological map of the study area. The river flows into the Aegean Sea near the Menemen Plain and is the major river which drains the watershed. It originates in the vicinity of Gediz near Kütahya and flows through the mountains of Murat, Simav, Umurbaba, Nif and Bozdağ and the surrounding area. The river flows between 38±04'-39±13' North and 27°48'-28°04' East. The bedrock structure of the river is composed mainly of metamorphic and volcanic rocks of the Palaeozoic, Mesozoic and Neogene ages. Uranium deposits in the Köprübaşı area of western Turkey occur in fluvial sedimentary rocks, which are underlaid by high-grade metamorphic rocks of the Menderes Massif (Yılmaz, 1981). On geological maps of Turkey, many granite masses are identified in the region between Buldan, Eşme, Demirci, Köprübaşı, Gördes and Alaşehir.

1.2. Sampling and field measurements

With a nominal sampling density of one sample per km, a total of 321 water samples were collected and a set of field measurements made along the course of the river. The sampling of the river waters and measurements in Gediz Basin were carried out between June 20 and August 20, 1996. All the sites were reached by boat or on foot.

In general, the field measurements were taken using small, lightweight, battery-operated, portable field instruments. Total gamma measurements (cps and μR/h) were made using a single-channel gamma-ray analyser (Scientrex BGS-4) at the points where the samples were collected. A standard pH and Eh meter (Hanna HI 8314) was used. Conductivity meters, similar in size and weight to the pH meters and measuring up to 199.9 μS/cm, were readily calibrated with a standardized KCl solution (Hanna HI 8633). The water temperatures were measured with precalibrated thermometers and recorded to the nearest degree Celsius.

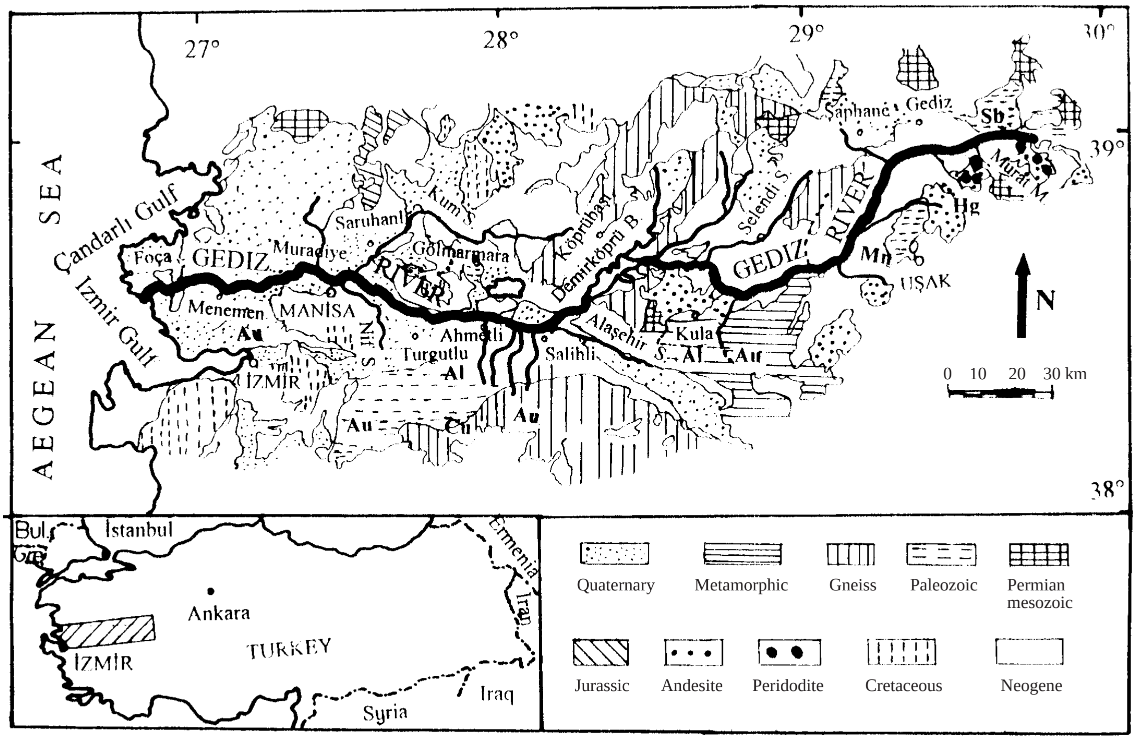

Figure 1. Geochemical map of study area

Field observations were recorded at the sampling points of the river water on cards suitable for use as a keypunch source document. The sample number, location and predominant rock type were noted. The width, depth, flow rate and turbulence of the water, as well as water and sediment colour, precipitates, sediment composition, water temperature, leaching, elevation, dominant vegetation, pollution source, plant type and the time and date of collection were duly recorded. Stream-water samples were collected from the water current flowing away from the bank. Some water samples contained abundant suspended material. This suspended material required filtration because it was thought to contain additional radium that was not in solution.

Water samples were collected in one-litre polyethylene bottles. The volumes for the determination of trace and heavy metals, radium and uranium were 500, 100 and 50 ml, respectively. The sample volume for the measurement of pH, Eh and conductivity was 100 ml. For total alkalinity, a 50-ml aliquot of the sample was used. It was expressed in parts per million CaCO3. After measuring the original pH, sulphuric acid was titrated into the sample at the last step. Giritlioğlu (1975) gives the description of the methods. The pH, Eh and specific conductance measurements were carried out using unfiltrated water samples and alkalinity measurements were conducted on filtrated water samples in the field laboratory. All the water samples were filtered through a 0.45 μm membrane and acidified to pH $\leq$ 1.0 with nitric acid to avoid the growth of microorganisms and to minimise water-wall interactions.

In order to determine the level of radium in the water, the collector-chamber method was used. In the laboratory, 100 ml was taken from each of the water samples. All the samples were transferred to radon bubblers, purged and sealed, flooded with pressurised aged air for about 20-30 minutes, and stored for about seven days or longer. The radium content was calculated from the radon that was generated during the storage time. Alpha disintegrations were then directly measured by an alpha scintillation counter system (Eberline model SAC-4). The counting - time was 20 minutes for each sample. The collector chamber was calibrated with radium standards obtained from NIST. The accuracy of the estimation for radium was 10%.

Uranium in the river water was determined using the laser-excited fluorescence technique (Scintrex UA-3) at the Institute of Nuclear Sciences. A uranium analysis was carried out on 33 river-water samples collected at 10-km intervals from the 321 sampling points along the course of the river. Using standard procedures, the 25-ml water sample was vigorously shaken and 0.20-ml aliquots of water were transferred to platinum dishes in duplicate. The determination of uranium was based on the fluorescence of a uranyl complex formed by the addition of the reagent Fluran (Scintrex) to the sample during analysis. Fluran, which has several functions, is a buffered inorganic complexing reagent that converts the various uranyl species in the water sample into a single form. Thus, uranyl fluorescence is enhanced by a factor of about 80. Its other function is masking quenchers such as iron and manganese. In the absence of the proprietary reagent, a dilute solution of sodium hexametaphosphate was substituted in order to establish optimum conditions for uranium fluorescence (Harms et al., 1982). The detection limit was better than 0.05 ppb.

2. Results and Discussion

Gediz River has a flowing rate and density which change depending on the geomorphology of the area between its spring and the point at which it joins the sea. Taking these into consideration, the river was divided into the three main parts in our study: the lower, the middle and the upper Gediz. Summary statistics for the temperature, conductivity, pH, Eh, alkalinity, gamma radiation, radiometric uranium, radium and uranium concentrations of the river water samples are shown in Tables 1 and 2.

Table 1. The statistical results of the field measurements of Gediz River

Statistics	Field measurements	Field results (total) (n=321)	Field results from different sectors		
			Lower Gediz (n=60)	Middle Gediz (n=110)	Upper Gediz (n=151)
Maximum	Gamma activity (cps)	176	176	116	130
Minimum		56	65	68	56
Mean		97	113	90	95
Std. Dev.		18	18	10	18
Mode		93	110	93	102
Median		93	110	93	102
Maximum	Gamma activity (μR/h)	16	16	13	14
Minimum		6	7	8	6
Mean		11	12	10	10
Std. Dev.		2	2	1	2
Mode		10	11	10	11
Median		10	12	10	11
Maximum	Uranium (beta) (cps)	3	3	3	3
Minimum		0	0	0	0
Mean		1	1	1	0
Std. Dev.		1	1	1	1
Mode		1	1	1	0
Median		1	1	1	0
Maximum	Uranium (beta) (μR/h)	14	14	13	12
Minimum		0	5	0	3
Mean		7	8	7	7
Std. Dev.		2	2	2	2
Mode		6	6	7	8
Median		7	7	7	7

Table 2. The statistical results of the water samples from Gediz River

Statistics	Field measurements	Field results	Field results from different sectors		
		(total) (n=321)	Lower Gediz (n=60)	Middle Gediz (n=110)	Upper Gediz (n=151)
Maximum	Temperature (°C)	30	30	24	25
Minimum		10	20	16	10
Mean		20	26	20	17
Std. Dev.		4	3	2	4
Mode		21	30	21	20
Median		20	26	20	18
Maximum	Specific conductance (μS/cm)	5940	5940	2882	5460
Minimum		8	210	8	32
Mean		1219	3026	490	1031
Std. Dev.		1403	1109	550	1328
Mode		50	2990	10	58
Median		532	3070	304	256
Maximum	pH	8.28	7.48	6.52	8.28
Minimum		5.55	5.87	5.79	5.55
Mean		6.41	6.71	6.29	6.38
Std. Dev.		0.53	0.51	0.13	6.67
Mode		6.30	6.09	6.25	6.30
Median		6.30	6.81	6.31	6.27
Maximum	Eh (mV)	36	28	16	36
Minimum		-108	-54	-21	-108
Mean		-8	-14	-8	-5
Std. Dev.		28	24	8	37
Mode		-13	-24	-13	-2
Median		-5	-16	-9	0
Maximum	Total alkalinity (mg/1)	750	328	318	750
Minimum		94	142	94	268
Mean		323	200	221	445
Std. Dev.		144	42	42	116
Mode		232	172	232	384
Median		270	185	232	392
Maximum	Ra-226 (Bq/l)	1.041	0.765	0.999	1.041
Minimum		0.21	0.021	0.021	0.021
Mean		0.287	0.235	0.313	0.289
Std. Dev.		0.165	0.144	0.189	0.149
Mode		0.191	0.085	0.191	0.340
Median		0.276	0.202	0.276	0.297
Maximum	U (ppb) (n=33)	10.4	6.4	10.4	9.8
Minimum		0.4	0.4	2.6	3.2
Mean		4.5	2.8	3.9	5.7
Std. Dev.		2.3	2.0	2.2	1.8
Mode		6.4	-	3.4	6.4
Median		3.6	2.5	3.2	5.4

The water temperatures were generally stable throughout the course of the river with the exception of those downriver. Water samples had low temperatures of 12°C at the headwaters of the river. Conductivity provides a direct indication of water salinity. There was a sharp increase in specific conductance in the first 35 samples at the estuary. In summer, the confluence of the river and the sea is closed so that the river water can be used for irrigation; however, the seawater mixes with the river water because there is no difference in the water levels of this area and the sea. As shown in Table 2, there were small changes in pH, Eh and alkalinity measurements throughout the river. Generally, water with high uranium and radium concentrations also exhibited higher conductivity and pH. In the case of uranium it is believed that the slightly higher pH, conductivity, and lower alkalinity could account for the somewhat greater difference in content than that observed for the other elements.

The gamma radiation and radiometric uranium measurements in the vicinity of the river varied noticeably. All the measurements were plotted graphically. The gamma activity (cps), radium (Bq/l) and uranium (ppb) concentrations are given in Figs. 2, 3, and 4, respectively.

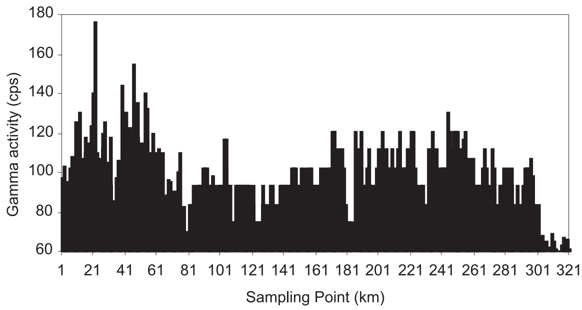

Figure 2. The gamma values of Gediz Basin

The radium concentration was found to correlate well with the local background gamma count rate. The radium concentration was found to vary between 0.021 and 1.041 Bq/l. In drinking water, the maximum allowed value for radium concentration is 0.185 Bq/l (USEPA, 1976). When this limit value is taken into consideration, sharp increases are observed at some points, as shown in Fig.3. This element contains a tremendous amount of economical as well as geological information. For example, the middle and upper Gediz ranges are very noticeable. The geochemical structure at Demirköprü Dam, Murat Mountain and in the surrounding are a may have an effect on the origin of the concentration of this element. This is because, there are uranium beds in the vicinity of Demirköprü Dam, to the north (Gördes, Demirci) and south (Salihli, Kula) and in the province of Uşak (Birsen, 1985). The 291^{st} sampling point and its surroundings (upper Gediz) boundaries. The reason are within Uşak's provincial for the anomaly in the radium concentration anomaly at this point may be the uranium (U_3O_8) found in Fakılı Village (Uşak). Furthermore, (U_3O_8) and thorium beds have also been found in Gediz (Kütahya) at the foot of Murat Mountain and in Eskişehir (to the northeast of Murat Mountain), respectively (M.R.E.I. Authors, 1980). The uranium-bearing conglomerate consists mainly of poorlysorted unconsolidated conglomerate with minor coarse-to-fine-grain sandstone, and siltstone, mudstone and very thin horizons of oxidized carbonaceous plant remains. Due to the stream and the underground waters found at Murat Mountain, the higher radium values were measured in water upriver in these areas due to wear caused by friction.

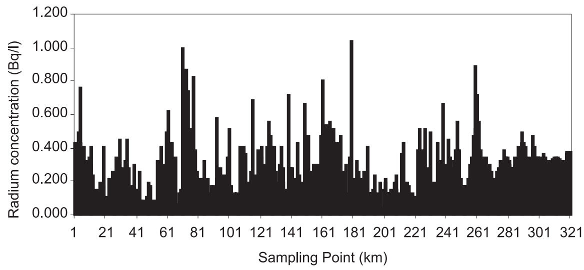

Figure 3. The radium values of the water samples

The concentration of uranium in the natural waters varied in the range 0.1-10 ppb. As confirmed by past experience, a concentration of uranium greater than 10 ppb definitely requires follow-up (King et al., 1976). The uranium concentration in the river water in the present study varied between 0.4 ppb and 10.4 ppb. Like radium, the level of uranium was also found to be higher where the gamma radiation and radium levels were high. The diversion of water for irrigation during the summer diminishes the water flow in the river basin, which also supplies water to Demirköprü Dam.

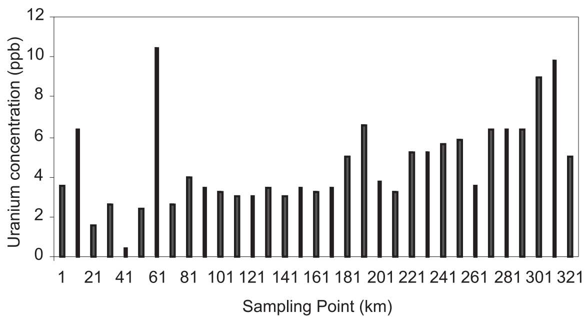

Figure 4. The uranium values of the water samples

Detection of uranium in the water implies the presence of nearby granite-type rocks. The middle gediz and neighbouring uranium deposits are located in Köprübaşı and its surroundings. The metamorphic rocks consist of banded gneiss and biotite gneiss with pegmatite and quartz veins. Following the deposition of the Palaeozoic units of the cover series, it is probable that the Menderes Massif was affected by the late phases of the Variscan Orogenesis and was metamorphosed a second time. However, the conditions of this metamorphism are as yet unknown. The Early-to-Middle Triassic leucocratic granites, about 240-230 Ma in age, postdate the Variscan event (Dora et al., 1995).

Finally, it can be concluded that Gediz River and the surrounding area consist of a number of radioactive fields which are not conducive to human health according to the level of uranium and field measurements. The reason for the increase in radium, especially the fields in Köprübaşı, is associated with the known uranium deposits in the Gediz River basin (M.R.E.I. Authors, 1980). However, in a study carried out in this zone, it was seen that this radiation does not have any significant effects on the people there part (Kılınç, 1995).

For example, Martinez-Aguirre and Garcia-Leon (1993) carried out a radioactivity survey in order to determine natural radioactivity in Guadalquivir River in southern Spain. They collected water samples from the river for the measurement of uranium and radium concentrations, which were found to be higher in the estuarine part of the river. However, compared to the levels reported in the present study, those noted in Guadalquivir River were low. In another study carried out in the area around a nuclear plant (Valencia, Spain), the uranium concentration was studied in water samples from Jucar River, with the help of low-level α-spectrometry. It was found that the pH, temperature, conductivity and uranium values varied from 6.97 to 7.87, from 6 to 8.5°C, from 671 to 20,000 μS/cm and from 0.00705 to 0.0296 Bq/l, respectively (Rodrigez-Alvarez and Sanchez, 1995). In their study, the uranium concentration was throught to vary with the salinity and pH.

References

Birsen, N., "Dünya uranyum kaynakları, yeterliliği ve Türkiyenin uranyum potansiyeli", Publications of the Turkish Atomic Energy Foundation, Ankara, 1985.

Bolivar, S.L., Dwight, E.H., "Uranium hydrogeochemical and stream sediment reconnaissance of the Craig NTMS Quadrangle, Colorado, including concentrations of forty-three additional elements", Los Alamos Scin. Lab., New Mexico, 1979.

Bolivar, S.L., Campbell, K. and Wecksung, G.W., "R-mode factor analysis applied to uranium exploration in the Montrose Quadrangle, Colorado", J. Geochem. Explor., 19, 723-743, 1983.

Brown, A., Steenfelt, A. and Kunzzenorf, H., "Uranium districts defined by reconnaissance geochemistry in South Greenland", J. Geochem. Explor., 19, 127-145, 1983.

Carreiro, M.C.V. and Sequeira, M.M.A., "^{226}Ac and ^{228}Ac in a fresh-water ecosystem", Radiat. Protect. Dosim. 24 (1/4), 133-137, 1988.

Cassels, B.M., "Radium-226 in natural water supplies in Kakadu National Park", Radiat. Protect. in Australia, 8 (2), 31-35, 1990.

Dora, O.Ö., Candan, O., Dürr, St. and Oberhänsli, R., "New evidence on the geotectonic evolution of the Menderes Massif", International Earth Sciences Colloquium on the Aegean Region, İzmir/Güllük-Turkey, 53-69, 1995.

Evans, R.D., "Radium in man", Health Phys., 27, 497-510, 1974.

Giritlioğlu, T., "İçme suyu kimyasal analiz metotları", İller Bankası, Ankara, 1975.

Harms, T.F., Ward, F.N. and Erdman, J.A., "Laser fluorometric analysis of plants for uranium exploration", J. Geochem. Explor., 15, 617-623, 1981.

Kılınç, R., "Köprübaşı ilçesi uranyum sahasından alınan toprak, bitki ve su örneklerindeki radyoaktivite miktarları ve çevresel önemi", I. Gediz Havzası Erozyon ve Çevre Sempozyumu, Salihli, Türkiye, 182-190, 1995.

King, J., Tauchid, M., Frey, D., Basset, M., Çetintürk, İ., Aydanoz, F., Keçeli, B., "Exploration for uranium in Southwestern Anotolia, Exploration for Uranium Ore Deposits", I.A.E.A. Publications, 1976.

Kumru, M.N., "Distribution of radionuclides in sediments and soils along the Büyük Menderes River", Proceedings of the Pacistan Academy of Sciences, 32 (1/4), 51-56, 1995.

Martinez-Aguirre, A. and Garcia-León, M., "Natural radioactivity in the Guadalquivir River at the South of Spain", J. Radioanal.and Nucl. Chem., 178 (2), 337-350, 1993.

Mineral Resource and Exploration Institute Authors, "Türkiye maden envanteri", M.T.A. Press, No: 179, Ankara, 1980.

Rodriguez-Alvarez, M.J. and Sanchez, F., "Behavior of uranium along Jucar River (Eastern Spain): Determination of ^{234}U / ^{238}U and ^{235}U / ^{238}U ratios", J. Radioanal. Nucl. Chem. 190 (1): 113-120, 1995.

USEPA, 1976, "Determination of radium removal efficiencies in water treatment process", Technical Note, ORP/TAD-76-5 Illinois EPA Springfield.

Yılmaz, H., "Genesis of uranium deposits in Neogene sedimentary rocks overlying the Menderes metamorphic massif, Turkey", Chem. Geol., 31, 185-210, 1981.