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in situ Determination of Radioisotopes in Certain Spring Waters and Spring Mud in Erzurum, Turkey

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In this study, the qualitative determination of radioisotopes of spring waters in Ilica, Köprüköy and Hasankale and spring mud of Köprüköy was made using a portable counting system coupled to NaI(Tl) detector. It was determined that an isotope is present when identified by the measuring system in all replicate measurements. In order to confirm the existence of the less identified radioisotope, a second high probable transition energy of the radioisotope was determined from the isotopes table and this transition energy was searched for in the measured spectra. The radioisotopes in three spring waters and a spring mud were determined using newly described analysis procedures. It is concluded that these waters and mud have the same aquifer.

Keywords: Radioisotope, Spring water, Spring mud, Qualitative analysis, NaI(Tl) detector.

INTRODUCTION

Turkey is rich in spring, thermal and mineral waters. Recently, usage of thermal and spring waters for health and relaxation has become increasingly popular in a certain population groups, especially among the elderly. It is well known that some underground waters contain an elevated level of dissolved natural radionucleide compared to tap water [1-3].

Although several countries have made significant surveys to assess the radiation that citizens received from water consumption people who use the springs, thermal and natural mineral waters attract a radiological risk from these sources worldwide [4-10]. The presence of radioisotopes and their concentrations in these waters varies over a very wide range [9] and is influenced by the physical (*e.g.* temperature), chemical (*e.g.* dissolved ions, pH) and geological properties of the aquifer; and depending upon the radionucleide content of the aquifer bedrock and soil, human activities may also influence radioactivity levels [10].

Because of specific health hazards, it is important to control human consumption of these radioactive waters. The investigation of the radionucleide content of these waters aids in understanding the geological and hydro-geological characteristics of the environment [11-14], especially as it can also act as a natural tracer [15-19]. Despite numerous studies on groundwater radioactivity that have been carried out in the literature, no report on a systematic and detailed work of these waters in Turkey. For these reasons, the aim of the present work is the *in situ* qualitative determination of the radioisotopes in Hasankale, Köprüköy and Ilica springs and in Köprüköy thermal mud near Erzurum.

EXPERIMENTAL

The measurements were carried out using a portable counting system (PCS) including a Scintillation detector, which is a hermetically sealed assembly that includes a high resolution NaI(Tl) crystal, a photomultiplier tube, an internal magnetic/light shield, an aluminium housing and a 14-pin connector coupled to a Canberra EasySpec Hand-Held NaI MCA. The detector specifications are outlined in Table-1. The PCS was calibrated using a ^{137}Cs test source with 185 kBq activity and checked with a ^{60}Co radioactive source at the laboratory. Since the measurement areas were hot and humid, the same procedures were repeated to test whether the system was correctly running in the thermal springs and mud and it was found that the system was running correctly in thermal spring conditions. Since the NaI(Tl) detectors are effected by temperature, the calibration was redone before each data acquisition. During the measurements the detector contacted water and mud. If the temperature exceeded 50 °C the measurement was stopped, since the operating temperature range of the detector is –10 to +50 °C.

To obtain good counting statistics, seven different measurements were carried out in springs.

TABLE-1
PCS SPECIFICATIONS

Data acquisition		
Channels	:	1024
Maximum Counts/Channel	:	2×10^6
Acquisition Mode	:	PHA add
Preamplifier		
Rise time	:	< 100 ns
Fall time	:	100 µs
Output noise	:	< 300 µV rms
Analog to digital converter		
Conversion rate	:	Wilkinson type with 66 MHz clock
Conversion time	:	18 µs maximum
Integral non-linearity	:	> 0.3 %
Differential non-linearity	:	± 1 channel
Lower level discriminator	:	45 keV
Upper level discriminator	:	2.1 MeV
Shaping amplifier		
Active circuit	:	4-pole Gaussian pulse-shapping filter
Shapping time	:	0.5 µs
Detector performance		
Spectrum shift	:	< 1 % of the 662 keV peak position from ^{137}Cs source for a change of counting rate from 0 to 30 kcps in the spectrum
Nuclide identification energy tolerance window	:	4 %
FWHM	:	7.5 %
Minimum dose rate	:	10 µR/h
Maximum dose rate	:	10 mR/h

The radioisotopes found in different spring waters and Köprüköy spring mud are listed in Tables 2 and 3. These tables show the radioisotopes determined in these measurements, their most intense γ-ray energies E, N_0/N_T ratio-N_0 is the number of determination at N_T measurements, their second intense γ-ray energies E_{sec}, which were taken from Lederer *et al*. [20] and existence of E_{sec} at the spectra.

N_0/N_T have been most important parameters to determine whether there is presence of radioisotopes in spring waters, because when a radioisotope is defined at all measurements it can definitely decide the presence of that radioisotope in spring water. Regarding $N_0/N_T \leq 4/7$, we have to consider another parameter E_{sec}. A peak with E_{sec} energy on spectra confirms that radioisotopes are existent in spring waters. Otherwise, we have to conclude that the radioisotopes are not existent in spring water or mud.

Some radioisotopes with no γ-ray peak with E_{sec} energy, were marked with (—); the peaks with almost identical energies also were marked with one of the symbols; √, # and * in these Tables. Since the γ-ray energies of radioisotopes are same or very close to each other it is difficult to determine which peak belong to which radioisotopes. To confirm the existence of these radioisotopes we used the following method: first, we determined whether or not there were most intense γ-ray energies (E_{sec}) of these radioisotopes taken from the "Table of Isotopes" [20]. The peak with E_{sec} energy for a given radioisotopes in the spectra were also investigated. If E_{sec} energy was observed at all spectra but for the other radioisotopes it could not be found in all spectra, it was concluded that the radioisotopes were also present, although the others were not.

RESULTS AND DISCUSSION

Certain radioisotopes were determined by using PCS in three spring waters and spring mud in Erzurum region of Turkey. These radioisotopes are presented in Tables 2 and 3. The radioisotopes from the energies of their most intense and second intense γ-rays were also determined.

The determined radioisotopes using analysis procedures are presented in Table-4. It is clearly shown that all radio-

TABLE-2
RADIOISOTOPES IN ILICA AND HASANKALE SPRING WATER

Ilica spring water						Hasankale spring sater					
Radioisotope	E (keV)		N_o/N_T	E_{sec} (keV)*	Existency of E_{sec}	Radioisotope	E (keV)		N_o/N_T	E_{sec} (keV)*	Existency of E_{sec}
^{40}K	1660.810		7/7			^{124}I	602.708	√	7/7	722.1298	Exist
^{65}Ni	1481.840		7/7			^{124}Sb	602.708	√	7/7	2260.890	Not exist
^{124}I	602.708	√	7/7	722.1298	Exist	^{133}Xe	80.997		7/7	356.280	Exist
^{124}Sb	602.708	√	7/7	2260.890	Not exist	^{134}Cs	604.697	√	7/7	563.1667	Not exist
^{133}Xe	80.997		7/7	356.280	Exist	^{214}Bi	609.312	√	7/7	1228.1120	Exist
^{134}Cs	604.697	√	7/7	563.1667	Not exist	^{93}Sr	590.280	*	7/7	–	–
^{232}Th	59.000		7/7	–	–	^{40}K	1660.810		6/7		
^{241}Am	59.540		7/7	98.944	Not exist	^{65}Ni	1481.840		6/7		
^{214}Bi	609.312		6/7	1228.1120	Exist	^{106}Ru	621.840	*	6/7	–	–
^{106}Ru	621.840		6/7	–	–	^{65}Zn	1115.520	#	6/7	–	–
^{214}Pb	351.921	#	5/7	534.481	Exist	^{138}Cs	1435.860		6/7	2630.1010	Not exist
^{211}Bi	351.100	#	5/7	–	–	^{46}Sc	1120.510	#	6/7	889	Exist
^{138}Cs	1435.860		5/7	2630.1010	Not exist	^{232}Th	59.000		5/7	–	–
^{133}Ba	356.005	*	5/7	302.382	Not exist	^{241}Am	59.540		4/7	98.944	Not exist
^{131}I	364.480	*	5/7	364.637	Exist	^{109}Cd	83.032		4/7		
^{93}Sr	590.280		5/7	–	–	^{152}Eu	1407.954		4/7		
^{46}Sc	1120.510		4/7			^{59}Fe	1099.220		4/7		
^{59}Fe	1099.220		4/7			^{152}Eu	1407.954		4/7		
^{152}Eu	1407.954		4/7								

*Taken from Lederer *et al.* [20].

TABLE-3
RADIOISOTOPES IN KÖPRÜKÖY SPRING WATER AND KÖPRÜKÖY SPRING MUD

Köprüköy spring water						Köprüköy spring mud					
Radioisotope	E (keV)		N_o/N_T	E_{sec} (keV)*	Existency of E_{sec}	Radioisotope	E (keV)		N_o/N_T	E_{sec} (keV)*	Existency of E_{sec}
^{232}Th	59.000		5/5			^{232}Th	59.000		5/5		
^{42}K	1524.655		5/5			^{42}K	1524.655		5/5		
^{134}Cs	604.697	√	5/5	563.1667	Not exist	^{214}Bi	609.312	√	4/5	1228.1120	Exist
^{214}Bi	609.312	√	4/5	1228.1120	Exist	^{106}Ru	621.840		4/5	–	–
^{124}I	602.708	√	4/5	2088.2740	Not exist	^{109}Cd	83.032		4/5		
^{106}Ru	621.840		4/5	–	–	^{88}Y	1836.010	#	3/5	–	–
^{124}Sb	602.708	√	4/5	2260.890	Not exist	^{88}Rb	1836.010	#	3/5	–	–
^{133}Xe	80.997		4/5	356.280	Exist	^{241}Am	59.540		3/5	98.944	Not exist
^{90}Mo	257.340	#	2/5			^{92}Y	934.460		2/5		
^{138}Xe	258.310	#	2/5			^{234}Pa	946.000		2/5		
^{65}Ni	1481.840		2/5			^{124}Sb	602.708	√	2/5	2260.890	Not exist
^{109}Cd	83.032		2/5			^{133}Xe	80.997		2/5	356.280	Exist
						^{134}Cs	604.697	√	2/5	563.1667	Not exist
						^{124}I	602.708	√	2/5	2088.2740	Not exist

*Taken from Lederer *et al.* [20].

TABLE-4
DETERMINED RADIOISOTOPES IN DIFFERENT SPRING WATERS AND KÖPRÜKÖY SPRING MUD

Radioisotopes	Ilica spring water	Hasankale spring water	Köprüköy spring water	Köprüköy spring mud
^{40}K	+	+		
^{42}K			+	+
^{46}Sc	+	+		
^{59}Fe	+	+		
^{65}Ni	+	+	+	
^{65}Zn				
^{88}Rb				?
^{88}Y				?
^{90}Mo			?	
^{92}Y				+
^{93}Sr	+	?		
^{106}Ru		?	+	+
^{109}Cd		+	+	+
^{124}I	+	+		
^{124}Sb	+			
^{131}I	+			
^{133}Xe	+	+	+	+
^{133}Ba				
^{134}Cs	+			
^{138}Cs	+	+		
^{138}Xe			?	
^{152}Eu	+	+		
^{211}Bi				
^{214}Bi	+	+	+	+
^{214}Pb	+			
^{232}Th	+	+	+	+
^{234}Pa				+
^{241}Am	+	+		+

isotopes determined using our analysis procedures definitely present in Ilica spring water. The ^{93}Sr and ^{106}Ru radioisotopes in Hasankale spring water, ^{90}Mo and ^{138}Xe radioisotopes in Köprüköy spring water and the ^{88}Rb and ^{88}Y radioisotopes have identical or very close energies to each other. However, it has not been determined which radioisotopes are present in spring water or mud using our analysis procedures as shown in Tables 2 and 3, no peak with E_{sec} energies were recorded at spectra for both radioisotopes in these spring waters and mud. For this reason, we have not determined which radioisotopes are available. To show this we use the symbol "?" in Table-4.

From Table-4, it is found that Ilica and Hasankale spring waters have almost identical radioisotopes contents. As a consequence, it is concluded that these spring waters have the same aquifer when their bedrock and soil are the main source of these radioisotopes. If so, we can determine that the possible presence of ^{93}Sr radioisotope is higher than the possibility of the ^{106}Ru radioisotope, because while the ^{93}Sr radioisotope is available in Ilica Spring water, ^{106}Ru is not available. Since the Köprüköy spring water and mud have identical same contents we may say that their aquifer is the same.

An interesting result is ^{241}Am, which is an isotope of the tansuranium Am element and is not a natural radioisotope. It has been recognized seven times in Ilica, four times in Hasankale, three times in Köprüköy thermal mud with the PCS. So, it is can conclude that

• Some artificial radioisotopes such as ^{241}Am can be formed deep under the surface under special pressure and thermal conditions [21].

• The presence of artificial ^{241}Am radioisotopes in three spring waters systems may have same aquifer.

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