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Distribution of Natural Radionuclides in Ama Fatma oil shale, Morocco†

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Abstract: The present work aims to analyze the distribution of radionuclides naturally occurring in the oil shale of the Ama Fatma coastal zone. For this purpose, using gamma-ray spectrometry, we analyzed the activity concentrations of uranium (U), thorium (Th) series, and potassium-40 (K-40) in oil shale samples retrieved from the study area. The concentrations of U-238 were ranged from 45 ±36 to 194±50 Bq.kg^{-1} and those of Th-232 fluctuate between 1.48±0.92 and 9.47±1.06 Bq.kg^{-1}. The measured concentrations for Ra-226, Pb-214 and K-40 oscillate between 33±5 and 138±9 Bq.kg^{-1}, 18±1 and 68±3 Bq.kg^{-1} and between 17±4 and 279±16 Bq.kg^{-1} respectively. The obtained values variation appears to be related to the deposit nature and assimilation processes of appropriate radionuclides rather than to the retrieved samples' related depth. The analysis of the ^{232}Th/^{238}U and ^{226}Ra / ^{238}U allowed an understanding of these radionuclides behaviors. The ratios ^{226}Ra / ^{238}U ranged from 0.56 to 0.74 with a mean value of 0.70 which indicates the existence of disequilibrium in the investigated oil shale samples. This disequilibrium can be attributed to significant differences in the mobility of these radionuclides.

Keywords: Natural radioactivity; Gamma spectrometry; Ama Fatma oil shale; specific activities ratios

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1. Introduction

The primary source of natural radionuclides is attributed to the three decay chains of ^{235}U, ^{238}U, and ^{232}Th, as well as ^{40}K. ^{232}Th has the longest half-life of 1.405 × 1010 years, followed by ^{238}U with a half-life of 4.47×109 years and ^{235}U with a half-life of 7.1x108 years. Natural uranium is composed of 99.27% ^{238}U, 0.72% ^{235}U, and 0.0055% ^{234}U [1]. The radionuclides activity concentrations and distribution in natural materials represent high interest as they provide useful information on environmental radioactivity [2]. The principal source of external irradiation to the human body is the gamma radiation emitted by naturally occurring radionuclides. The human external exposure to natural radiation doses is related to the geological and geographical conditions as the associated amount of gamma radiation varies from one region to another along the world [3,4]. Due to this heterogeneity, radionuclides appear in diverse levels in rocks in each region around the world [5]. Higher radiation levels are linked to igneous rocks (e.g., granite) as the content of uranium in a rock increases with silica content [6-7]. To be noted that some minerals enclose higher levels of natural radionuclides compared to others [8]. Less important radiation levels are associated with sedimentary rocks [9] with the exception of certain phosphate and shale rocks that display a relatively high content of radionuclides [10].

The examination of sedimentary rocks for uranium and thorium isotopes typically involves a somewhat laborious process of radiochemical procedures to separate the heavy

radioactive elements, followed by an analysis of the height of alpha pulses [11-15]. It would be desirable to have simpler and faster methods of analysis. One such alternative is gamma ray spectrometric analysis [16-17] which allows for the non-destructive measurement of the natural gamma ray activity of the core at energies exceeding approximately 1 MeV. In this particular aspect of the natural gamma spectrum, only three nuclides contribute: ^{40}K, ^{214}Bi, and ^{208}Tl. ^{40}K is a radionuclide that has a long lifespan, whereas ^{21}4Bi and ^{208}Tl are short-lived daughters in the decay schemes of ^{238}U and ^{232}Th, respectively.

The present study analyzes the radionuclides concentrations naturally occurring in oil shale samples retrieved from the Ama Fatma coastal zone. The study area is situated in the Tarfaya-Boujdour basin, which defines the Atlantic margin area between the towns of Tarfaya and Boujdour. This region is bounded by the Atlantic Ocean on the west and the Republic of Mauritania on the east, as depicted in Figure 1. Located approximately 80 km to the north of the city, this segment commences with a black shale layer exceeding 2 m in thickness [18-19]. The geological characteristics of the study area consist of Marl, a sedimentary rock abundant in organic matter while the mineral composition of the samples mainly comprises calcite, dolomite, and quartz [18].

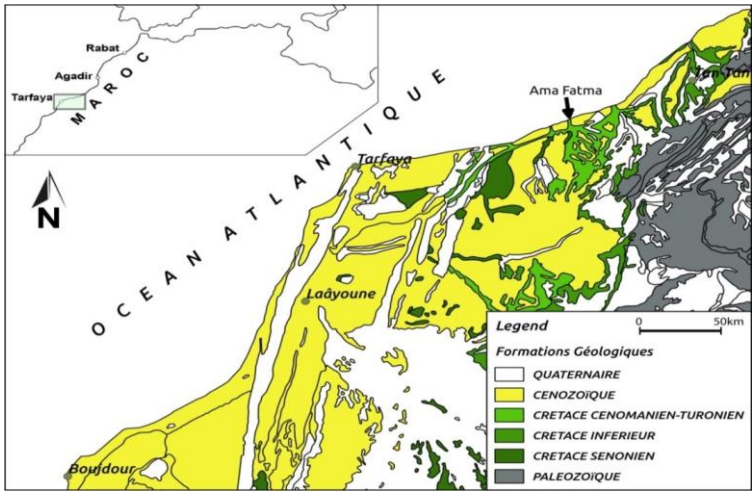

Figure 1. A sketched map of the study area showing its main geological features.

The objective of this study includes investigating the natural concentrations of radionuclide elements (^{238}U, ^{235}U, ^{232}Th, ^{226}Ra, ^{214}Pb, and ^{40}K) within Ama Fatma oil shale and assessing the disequilibrium of uranium series radionuclides by studying the ^{232}Th/^{238}U, and ^{226}Ra / ^{238}U ratios.

2. Materials and Methods

2.1. Sample collection and physical preparation

The oil shale samples under investigation were collected from the Ama Fatma region, located within the coastal basin of Tarfaya-Boujdour in Southwest Morocco (Fig. 1). The geographical coordinates of the sampling area are X= 28.202 and Y= -11.781 [19]. These samples were collected at different depths, ranging from 24.18 to 26.69 meters from the top of the Ama Fatma outcrop, as shown in Table 1. The physical parameters including total carbon, lithological parameters and mineralogical composition were analyzed and presented in [18,19] as shown in Table 1. The mineralogical composition was measured using X-ray diffractometry while the granulometry was performed using a wet Laser Diffraction equipment (Malvern Mastersizer 2000) using the Hydro 2000G dispersion Unit [18]. The total carbon was determined using Rock-Eval pyrolysis method [19].

Table 1. Chemical & mineralogical characteristics of Ama Fatma oil shale samples *[18,19].

Samples	TOC (%) (Rock Eval)*	Depth (m) (from top of outcrop)*	Mineralogical composition**							Lithology*	Age*
			Quartz	**Calcite**	**Dolomite**	**Clay**	**Halite**	**Fluorapatite**	**Pyrite**		
S_1(9)	6.19	24.18	6.8	70.2	8.6	5.5	7.3	1.6	-	Marl very rich in organic matter	Upper Cretaceous
S_1(10)	5.84	24.69	7.3	65.3	8.8	4.7	3.1	9.4	1.5		
S_1 (10-13)	5.84	24.83	17.5	55.8	14.4	7.4	4.4	0.4	0.3		
S_1 (13-14)	5.84	24.95	3.4	67.8	4 .7	3.7	5.7	13.3	1.7		
S_1(14)	5.91	25.27	6.7	66.9	4.8	3.9	2.7	5.4	9.7		
S_1(15)	-	25.8	17.2	47.9	20.6	8.1	4.1	0.4	1.6		
S_1(16)	7.64	26.69	8.4	65.1	14.8	8.3	3.3	-	-		

The obtained oil shale samples were ground to a fine powder using an electric grinder. Then homogenized samples were weighed and stored in a standard specification plastic container. These containers have been carefully sealed and stored for at least four weeks to prevent contamination of the spectrometer and the escape of ^{222}Rn and ^{220}Rn radiogenic gasses to allow radioactive secular equilibrium to be achieved in the decay chains.

2.2. Analytical methods

The natural radionuclides (^{238}U, ^{235}U, ^{232}Th, ^{226}Ra, ^{214}Pb, and ^{40}K) activity concentrations were measured by high-resolution gamma-ray spectrometry. The detector used is a low background CANBERRA high-purity coaxial germanium (50% efficiency, 1.8 keV resolution), housed in a 10-cm-thick high-purity lead shield. The gamma spectra delivered by the detector are analyzed by Genie 2000 gamma analysis software. A typical gamma-ray spectrum of an oil shale sample recorded with this system is presented in Fig. 2. The ^{234m}Pa (1001,03 keV) was used to estimate ^{238}U specific activities, while the peak area at ^{228}Ac (911,32 keV) was used to estimate ^{232}Th. For the determination of ^{235}U, ^{226}Ra, ^{214}Pb, and ^{40}K, their most intensive lines at 143,76 keV, 186,21 keV, 351,92 keV and 1460,81 keV represented their activities, respectively. It worthily noting that the spectrometry gamma analytical method requires important amount of sample compared to other alternative metods (e.g. Alpha spectrometry).

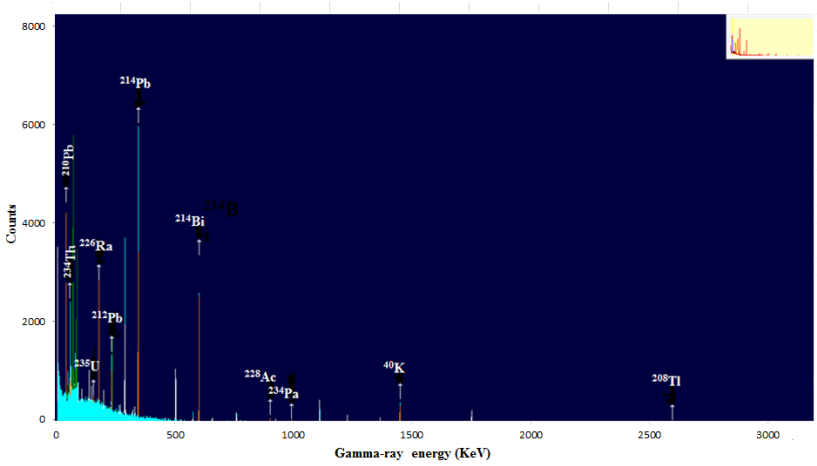

Figure 2. A typical gamma-ray spectrum of oil shale (sample S_1(9)).

3. Results and Discussions

3.1. Radionuclides Distributions

Table 2 displays the specific activities of ^{226}Ra, ^{238}U, ^{232}Th, ^{214}Pb, ^{235}U, and ^{40}K measured in oil shale samples collected at different depths from the Ama Fatma coastal zone. The activities of ^{226}Ra ^{238}U, ^{232}Th, ^{214}Pb, ^{235}U, and ^{40}K ranged between 33.4, 45.1 to 193.7 Bq.kg^{-1},

1.48 and 9.47 Bq.kg^{-1}, 17.8 and 68.2 Bq.kg^{-1}, 4.02 and 4.75 and between 16.9 and 279.2 Bq.kg^{-1} respectively. The highest values of ^{226}Ra, ^{238}U, and ^{235}U were recorded in S1(14). The S1(16) shows the highest value of ^{232}Th and ^{40}K while S1(10) indicates the highest value of ^{214}Pb. The obtained activities of ^{238}U and ^{226}Ra measured in the Ama Fatma oil shale are greater than the activities recorded in soil samples reported as not contaminated while the activities of ^{232}Th and ^{40}K are less high compared to values recorded in the abovementioned soil [20]. The high values of uranium found in the Ama Fatma oil shale samples are related to the oil shale and the sedimentary deposits abundant in organic material abilities to capture redox-sensitive elements, including Re, V, and U, under reductive conditions [10,21]. In comparison with the oil shale of Timahdit [21], the Ama Fatma oil shale shows less important activities mainly ^{238}U, ^{235}U, and ^{232}Th. It was expected that ^{232}Th, ^{238}U, and ^{40}K would demonstrate distinct environmental behavior, given their particular solubility and varying chemical reactivity [22]. The examination of the exemplary curve (Fig. 3) indicates that the specific activities of ^{226}Ra, ^{238}U, ^{232}Th, ^{214}Pb, ^{235}U, and ^{40}K in the oil shale deposits are not related to depth and the irregular recorded variation seems to be related to the lithological factors which probably controlling radionuclides distribution.

Table 2. Specific activities of ^{226}Ra, ^{238}U, ^{232}Th, ^{214}Pb, ^{235}U and ^{40}K Bq.kg^{-1} with the respective standard errors (±σ) and isotopic ratios ^{232}Th/^{238}U and ^{226}Ra / ^{238}U. .

Samples	^{226}Ra	±σ	^{238}U	±σ	^{232}Th	±σ	^{214}Pb	±σ	^{235}U	±σ	^{40}K	±σ	^{232}Th/^{238}U	^{226}Ra / ^{238}U
S_1(9)	104.0	7.7	139.9	41.7	2.79	1.15	44.8	2.3	4.02	2.90	145.8	9.0	0.020	0.74
S_1(10)	125.0	8.4	172.8	42.2	2.44	0.90	68.2	3.5	4.40	2.86	110.9	6.9	0.014	0.72
S_1(10-13)	47.7	5.4	67.1	32.6	4.79	0.69	29.8	1.6	4.27	2.78	159.4	9.2	0.071	0.71
S_1(13-14)	33.4	4.9	45.1	36.4	2.25	0.82	25.0	1.4	-	-	63.6	5.0	0.05	0.74
S_1(14)	138.3	9.2	193.7	50.3	1.48	0.92	57.7	3.0	4.75	2.84	86.4	6.7	0.008	0.71
S_1(15)	40.0	4.63	71.2	43.3	-	-	17.8	1.1	-	-	16.9	4.2	-	0.56
S_1(16)	131.0	9.5	185.7	58.5	9.47	1.06	61.5	3.2	4.13	3.83	279.2	16.2	0.05	0.70

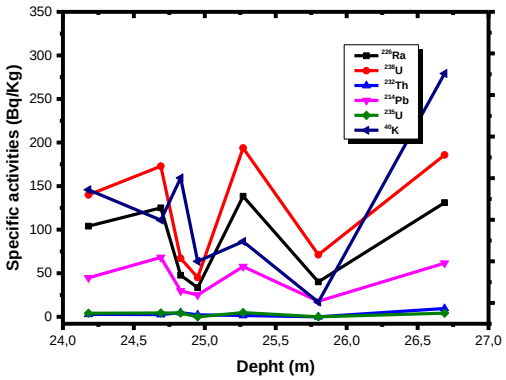

Figure 3. Variation of ^{226}Ra, ^{238}U, ^{232}Th, ^{214}Pb, ^{235}U, and ^{40}K concentrations versus depth.

3.2. Pearson correlation coefficients among radionuclides, TOC, and mineralogical composition

Pearson correlation coefficients between the measured radionuclides, mineralogical composition, and the total carbon (TOC) in the examined oil shale samples are presented in Table 3. All natural radionuclides of the 238U-series (^{226}Ra, ^{238}U, and ^{214}Pb) showed a strong positive correlation with each other, which means that the distribution mechanisms of ^{226}Ra, ^{238}U, and ^{214}Pb within the oil shale are more or less the same. On the other hand, no correlation was found with the ^{40}K, ^{232}Th, and ^{238}U series, revealing distinctively different mechanisms of control of ^{40}K, ^{238}U, and ^{232}Th concentrations in Ama Fatma oil shale. Positive correlations between the organic matter content and ^{40}K and ^{232}Th specific

Activities. Organic matter is a component of great importance because it tends to form soluble or insoluble complexes with radionuclides, which can migrate throughout the profile or be retained in the soil [23]. significative positive correlations were found between ^{232}Th and dolomite and clay, which indicate the association of these radionuclides with dolomite and clay. The ^{226}Ra, ^{238}U, and ^{235}U were found to be positively correlated with pyrite, which is in accordance with earlier results for oil shale from the Timhdit area, These correlations could be attributed to the common affinity of these elements for pyrite minerals.

Table 3. Pearson correlation coefficients between mineralogy, total organic content, and radionuclides activity concentrations in oil shale samples collected from Ama Fatma, Morocco.

	TOC	*226Ra*	*238U*	*232Th*	*214Pb*	*235U*	*40K*	*Quartz*	*calcite*	*Dolomite*	*Clay*	*Halite*	*Fluorapatite*	*Pyrite*
TOC	1,00													
226Ra	0,42	1,00												
238U	0,44	**0,99**	1,00											
232Th	**0,90**	0,15	0,18	1,00										
214Pb	0,39	**0,95**	**0,94**	0,18	1,00									
235U	-0,46	0,34	0,36	-0,53	0,30	1,00								
40K	**0,90**	0,50	0,48	**0,96**	0,54	-0,64	1,00							
Quartz	-0,03	-0,44	-0,38	0,34	-0,48	-0,15	-0,10	1,00						
calcite	0,10	0,56	0,49	-0,31	0,59	-0,05	0,34	-0,92	1,00					
Dolomite	0,58	-0,32	-0,25	**0,84**	-0,37	-0,63	0,09	0,85	-0,85	1,00				
Clay	**0,70**	-0,17	-0,11	**0,91**	-0,22	-0,65	0,41	0,75	-0,67	0,94	1,00			
Halite	-0,18	-0,42	-0,47	-0,21	-0,46	-0,71	-0,09	-0,17	0,25	-0,15	-0,12	1,00		
Fluorapatite	-0,49	0,07	0,03	-0,58	0,26	0,51	-0,22	-0,78	0,56	-0,70	-0,81	-0,09	1,00	
Pyrite	**0,99**	**0,68**	**0,70**	-0,70	0,46	**0,99**	-0,13	-0,40	0,43	-0,51	-0,51	-0,60	0,05	1,00

3.3. Study of ^{232}Th/^{238}U and ^{226}Ra / ^{238}U activity concentration ratios

The activity concentrations ratios ^{238}U/^{226}Ra and ^{226}Ra / ^{238}U were carried out to analyze their variations as shown in Table 2.

3.3.1.232. Th/ ^{238}U

The calculated ^{232}Th/^{238}U ratios ranged from 0.008 to 0.071 with a mean value of 0.03. The values of the ^{232}Th/^{238}U are very low as the oil shale samples have marine origin. The obtained low ratio values of ^{232}Th/^{238}U suggest that the studied drilling samples were deposited in a reducing environment [24].

3.3.2.226. Ra / ^{238}U

If secular equilibrium prevails in the ^{238}U chain, ^{226}Ra / ^{238}U will be approximately 1. Thus, any values of ^{226}Ra / ^{238}U other than 1 would suggest the existence of disequilibrium. The ratios ^{226}Ra / ^{238}U of Ama Fatma oil shale ranged from 0.56 to 0.74 with a mean value of 0.70. Such ratios being lower than 1 indicate the existence of disequilibrium in the oil shale samples under consideration. This disequilibrium can be attributed to significant differences in the mobility of the radionuclides present. Differential ion mobility has been widely discussed in the literature as a potential cause of such disequilibrium [25,26,27].

4. Conclusions

The study allowed analyzing the activity concentrations of the ^{238}U, ^{235}U, ^{232}Th, ^{226}Ra, ^{214}Pb, and ^{40}K in shale oil samples collected from the coastal zone of Ama Fatma. The

results show that the distribution of studied radionuclides varied independently to depth where it appears to be associated with the lithological characteristic of the zone. The analysis of the $^{232}Th/^{238}U$ and ^{226}Ra / ^{238}U allowed an understanding of these radionuclides behavior. The ratios $^{238}U/^{226}Ra$ ranged from 0.56 to 0.74 with a mean value of 0.70 which indicates the existence of disequilibrium in the investigated oil shale samples. This disequilibrium can be attributed to significant differences in the mobility of these radionuclides. This research data can be used as a valuable resource for further related studies and investigations.

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