

Determination of Uranium Concentrations in Brick Samples from Misan Province-Iraq using Kinetic Phosphorescence Analyzer (KPA)

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Abstract: Bricks as building materials contain radionuclides. Primordial radionuclides in building materials are one of the sources of the radiation hazards in dwellings made of these materials. It is therefore important to determine the uranium levels in bricks for public health and radiation protection. The natural uranium concentrations of brick samples which collected from 18 brick factories at 7 various areas of Misan province were measured using Kinetic Phosphorescence Analyzer (KPA-11) and an annual effective dose of the brick samples were calculated. The results showed that, the natural uranium concentrations ranged from the 0.692-46.481 mg kg⁻¹ with a mean value of 9.403 mg kg⁻¹ and the annual effective dose ranged from the 0.0000052-0.00162 mSv y⁻¹ with a mean value of 0.000391 mSv y⁻¹. The obtained results showed that all brick samples contained uranium concentrations with various rates and the annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 mSv y⁻¹. That is part of the total average annual doses of radiation from all natural sources of radiation, 2.4 mSv y⁻¹.

Key words: Uranium, brick samples, KPA, annual effective dose, Misan province, dissolution

INTRODUCTION

An established fact, all construction materials contain trace amount of natural radioactivity. This activity is a major source of external and internal radiation exposure for the occupants of a dwelling. The most commonly radionuclides in the construction material are U-238, Th-232, their decay products and K-40. Amongst the decay series product, radon is an important source of natural radiation. It found in the environment due to the radioactive decay of uranium. It is estimated that 50-55% of the average annual dose from the natural background radiation is contributed by Rn-222 alone (UNSCEAR, 1982). All building materials such as concrete, brick, sand, aggregate, marble, granite, limestone, gypsum, etc., contain mainly natural radionuclide including U-238, Th-232, their decay products and K-40 (Ravisankar *et al.*, 2011; 2012). Bricks are used as one of the main building materials, therefore, the disclosure of the basic radiological parameters and radioactive contents in bricks and others construction materials is important. This allows us to calculate the exposure of the population of the radiation from natural source (Badhan *et al.*, 2009). The major aims of this study are to estimate the concentrations and annual effective dose of uranium in brick samples collected from various locations in Misan province, Iraq and to provide data which can serve as a basis for the determination of changes in the near future.

MATERIALS AND METHODS

Experimental method

The study area: The area covered by this study (Misan province) is located in the South of Iraq, (Fig. 1). It lies between latitudes of 31°30.0'N-33°0'.00'N and between longitudes of 46°30'.0'E to 47°30'.0'E. It covers an area of approximately 16072 km². This area is very important, since, it contains many brick factories. The residents of Misan province depend on bricks as a basic building material. The disclosure of the radioactive contents within bricks is important, since, it allows us to calculate the exposure of the population of the radiation from natural source. The above causes make it necessary to measure the natural radioactivity in building materials which will serve as reference data for future studies.

Sample collection: In the present research, 18 brick samples were collected from various areas of Misan province-Iraq, (Almizbania Factory) (S₁), (Almaymuna Factories) (S₂) and (S₃) (Almajar Factories) (S₄) and (S₅), (Altubar Factories) (S₆) (S₇) (S₈) (S₉), (Kasiba Factories) (S₁₀) (S₁₁) (Altayib Factories) (S₁₂) (S₁₃) (S₁₄) and (Albatira Factories) (S₁₅) (S₁₆) (S₁₇) (S₁₈). Each one of collected samples was given a unique code and denoted with its GPS coordinates which was detected by a handheld GPS device as shown in (Fig. 2). The samples were dried for about 2 h by an oven with temperature of 80°C, then crushed to pass through 75 µm mesh sieve to

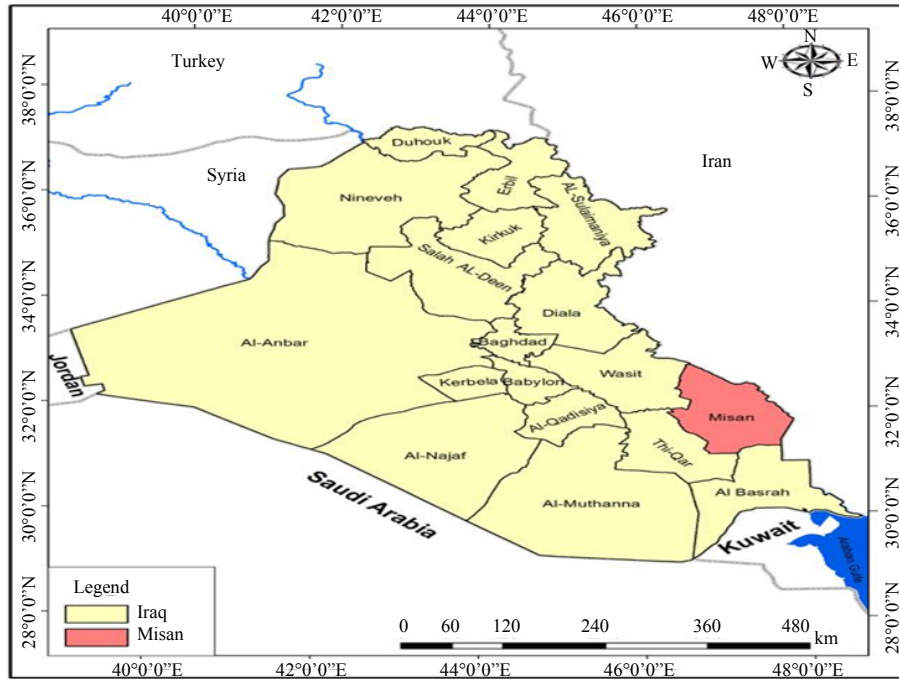


Fig. 1: Map of the administrative divisions of Iraq showing the studied area (Misan province)

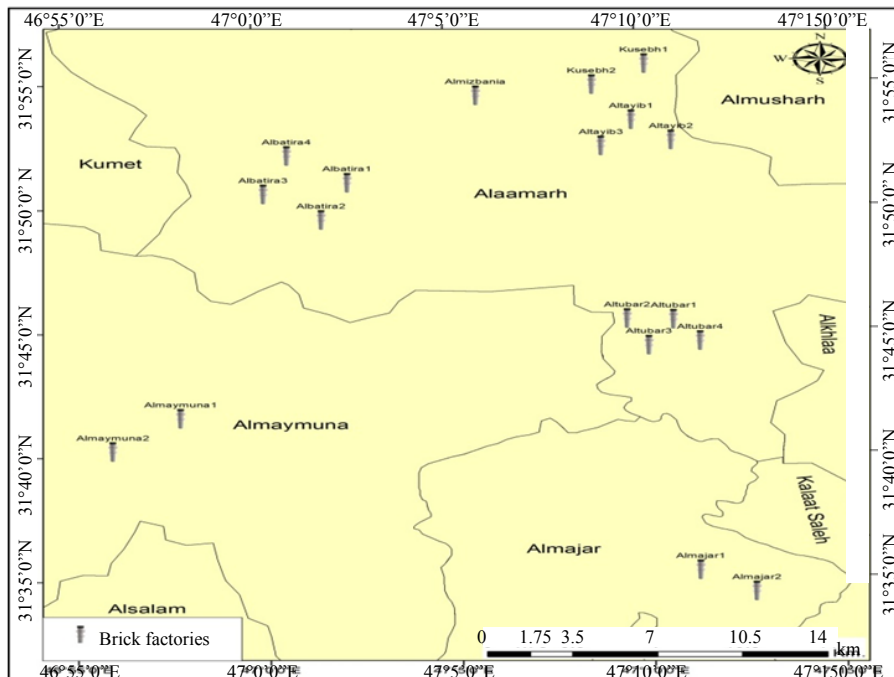


Fig. 2: Map of the administrative divisions of the study area (Misan province) showing locations of brick factories

make sure they have homogenous. These crushed brick samples were sealed in plastic containers and left until analysis (Begy *et al.*, 2013). These samples were prepared for the analysis of uranium concentrations using a Kinetic

Phosphorescence Analyzer (KPA) method (Anonymous, 2006). In the laboratory, an 0.2 gm of the sample is placed in a glass vial, then a 10 mL of HNO₃ having concentration of 8 m is added along with 0.5 mL of

Table 1: The main characteristics of the KPA-11 (Anonymous, 2006)

Parameters	Laser KPA
Selectivity	Tunable for U, Eu, Sm. Isotopes analysis
Laser source	Nitrogen laser with 337 nm wavelength
Pulse duration	3 nsec
Repetition rate	20 pulses sec ⁻¹
Emission wavelength	515 nm for U
Pulse power	120 J
Buffer	Uraplex
Sample volume	1 mL
MDL	0.01 ng·mL ⁻¹ (0.13 mBq·L ⁻¹)
Precision (RSD)	(1-3%) at U>0.01 ng·mL ⁻¹ (7-10%) at U<0.01 ng·mL ⁻¹
Analysis range	0.01 µg·L ⁻¹ -5 mg·L ⁻¹
Data processing	Computer software

H₂O₂ having concentration of 30%. Afterwards, the sample is placed and stirred on a hotplate until it reveals the salt colors that range from yellow to white. Then, the sample is heated to sub-boiling temperature for +2 h, starting with hotplate set at 200. If sample evaporates to ~2-3 mL, 8 M HNO₃ have to be added in order to return to a volume of ~10 mL. After +2 h, 8M HNO₃ have to be added again in order to return to a volume of ~10 mL. After that allow the sample to evaporate to ~2-3 mL and left to cool. Transfer the sample into 50 mL centrifuge tube rinsing vial with 2% HNO₃. Add DI water and 2% HNO₃ in order to return to a volume of 50 mL with a concentration of (0.3-1 m) HNO₃. If sample does not settle, then centrifuge or filter the sample liquid. Then, the solution is analyzed by using the KPA-11 with appropriate further dilution (ASTM., 1992).

Analysis using KPA technique: The Kinetic Phosphorescence Analyzer (KPA-11) at Radiation Protection Center (RPC), Ministry of Environment-Baghdad was used for measuring uranium concentrations in the brick samples. KPA-11 provides a rapid, precise and accurate determination of uranium concentration in aqueous solutions. It is a sensitive and selective analytical technique with low detection limits (is about 0.01 ng·mL⁻¹) (Decambox *et al.*, 1991; Shafik *et al.*, 2014a, b). The main characteristics of the KPA are showed in Table 1 (Anonymous, 2006).

Uranium standard solutions for KPA-11: Uranium standard solutions were prepared using Uranium Octoxide (U₃O₈). Firstly, a stock standard solution of 1000 mg·L⁻¹ (1000 ppm) was prepared by dissolving 117.9 mg of U₃O₈ in 100 mL of 0.82 M nitric acid (HNO₃) in volumetric flask. To construct the calibration curve for kinetic phosphorescence, analysis series of calibration standard were prepared to cover a wide range of uranium concentration which is expected in brick samples. Uranium concentrations in the series of standards were 0.5, 1, 2, 3, 4, 5, 7, 8 and 10 µg·L⁻¹. This set of standards was used to construct the calibration curve. In addition, background measurements as in calibration were

performed using nine calibration standard solutions for each analytical range, ranging in concentration from the detection limit up to 10 µg·L⁻¹. A blank sample of 0.82 m HNO₃ was used to determine the background and reagent uranium concentration. The blanks phosphorescence intensity was subtracted from all KPA measurements (Shafik *et al.*, 2014a, b).

RESULTS AND DISCUSSION

The result of uranium concentrations, activities of uranium-238, uranium-235 and annual effective dose on the public for brick samples of Misan province are shown in Table 2. Uranium Concentration (UC) in the sample was calculated using the (Eq. 1) (Anonymous, 2006):

$$\ln U * t = \ln U * 0 - (K_p - K_q) t \quad (1)$$

Where:

- U_i : Population of excited uranium ions at time i = t or 0
- k_p : Rate constant for phosphorescent decay
- k_q : Rate constant for all other relaxation processes

The KPA-11 is controlled by KPA Win Software operating under Windows for automatic calculation of uranium concentration (Anonymous, 2006).

The uranium concentrations in brick samples ranged from 0.692 mg kg⁻¹ (S₆) (Al-Tubar factory) to 46.481 mg kg⁻¹ (S₁₂) (Al-Tayib factory) with a mean value of 9.403 mg kg⁻¹.

By measuring the uranium concentrations, the activities of uranium-238, uranium-235 can be calculated for these isotopes by knowing the specific activity and the mass fraction of them (Table 3) (Majali, 2005) by using the (Eq. 2) (Majali, 2005):

$$A \left(\frac{\text{Bq}}{\text{kg}} \right) = UC \left(\frac{\text{mg}}{\text{kg}} \right) \times \text{I.A.M} (\%) \times \text{S.P.A} \left(\frac{\text{Bq}}{\text{mg}} \right) \quad (2)$$

Where:

- A : Activity
- UC : Natural Uranium Concentration
- I.A.M : The Isotopic Abundance (%) by Mass fraction
- S.P.A : The Specific Activity

The activity of uranium-238 in brick samples ranged from 8.545 Bq kg⁻¹ (S₆) (Altubar factory) to 574.025 Bq kg⁻¹ (S₁₂) (Altayib factory) with a mean value of 116.126 Bq kg⁻¹ and the activity of uranium-235 ranged from 0.398 Bq kg⁻¹ (S₆) (Altubar factory) to 26.773 Bq kg⁻¹ (S₁₂) (Altayib factory) with a mean value of 5.416 Bq kg⁻¹.

The annual effective dose H_E (mSv y⁻¹) on the public can be calculate using RESRAD program. The annual effective dose on the public in brick samples ranged from

Table 2: Results of natural uranium concentration activity of uranium-natural uranium concentration, activities of uranium -238, uranium-235 and annual

Code of brick samples	Location	Natural uranium concentration (mg kg ⁻¹)	Activity of U-238 (Bq kg ⁻¹)	Activity of U-235 (Bq kg ⁻¹)	Annual effective dose H _e (mSv y ⁻¹) Unat
S ₁	Almizbania	27.718	342.308	15.965	0.00021
S ₂	Almaymuna	4.746	58.611	2.733	0.000073
S ₃		5.104	63.032	2.939	0.000075
S ₄	Almajar	1.870	23.093	1.077	0.000042
S ₅		7.652	94.499	4.407	0.000084
S ₆		0.692	8.545	0.398	0.0000052
S ₇	Altubar	7.284	89.955	4.195	0.0000823
S ₈		4.423	54.622	2.547	0.00041
S ₉		3.885	47.978	2.237	0.00029
S ₁₀	Kasiba	1.827	22.562	1.052	0.000207
S ₁₁		5.439	67.169	3.132	0.000611
S ₁₂		46.481	574.025	26.773	0.00162
S ₁₃	Altayib	4.255	52.547	2.450	0.00063
S ₁₄		2.204	27.218	1.269	0.00034
S ₁₅		2.107	26.020	1.213	0.00031
S ₁₆	Albatira	32.144	396.968	18.514	0.00094
S ₁₇		2.668	32.948	1.536	0.00032
S ₁₈		8.758	108.158	5.044	0.00079
	Mean value	9.403	116.126	5.416	0.000391

Effective dose H_e (mSv y⁻¹) in brick samples measured within this research

Table 3: Radioactive properties of natural uranium isotopes (Majali, 2005)

Isotope	Isotope half-life (year)	Specific activity (Bq mg ⁻¹)	Isotopic abundance (%)	
			By mass	By activity
²³⁸ ₉₂ U	4.51×10 ⁹	12.44	99.2745	48.2
²³⁵ ₉₂ U	7.1×10 ⁸	80	0.7200	2.2
²³⁴ ₉₂ U	2.47×10 ⁵	230.700	0.0055	49.6

0.0000052 m Sv y⁻¹ (S₆) (Altubar factory) to 0.00162 mSv y⁻¹ (S₁₂) (Altayib factory) with a mean of 0.00039 mSv y⁻¹.

The results (Table 2) showed that for Misan province, the values of the natural uranium concentrations and activities of uranium-238, uranium-235 in all brick samples have various rates.

The annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 m Sv y⁻¹ that is part of the total average annual doses of radiation from all natural sources of radiation, 2.4 m Sv y⁻¹ (Ford, 2004).

CONCLUSION

The final results obtained indicates that the uranium concentrations in brick samples of the studied area overall have various rates and hence, the annual effective dose of gamma rays that resulted from the dissolution of uranium isotopes in brick samples were less than the total gamma rays dose which is 0.5 m Sv y⁻¹ that is part of the total average annual doses of radiation from all natural sources of radiation 2.4 m Sv y⁻¹. Therefore, it can be concluded that the brick samples of the area under the study, i.e., Misan province are radioactively safe to be used as construction materials and does not impose any serious threat to the health of locals. This study can be used as a reference for more extensive studies within the same subject in future.

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