

Efficiency Calibration Study of NaI(Tl) Detector for Radioactivity Measurements in Soils from Ain Zalah Oil Field

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Abstract: The quality of the results of gamma spectrometry measurement depends directly on the accuracy of the detection efficiency in the specific measurement conditions. In this work, The efficiency of NaI(Tl) detector with Marinelli beaker was measured in four different methods with point sources, natural radioactive samples (Thorium nitrate $\text{Th}(\text{NO}_3)_4 \cdot 4.5\text{H}_2\text{O}$, Uranyl acetate $\text{UO}_2(\text{CH}_3\text{COO})_2 \cdot 2\text{H}_2\text{O}$ and KCl powder) combined with soil and Al_2O_3 . These methods were applied to calculate the activity concentrations of ^{40}K , ^{226}Ra and ^{232}Th in soil samples from Ain Zalah oil field. The results show that the efficiency methods with soil and Al_2O_3 are favorable to those based on point sources, KCl alone. The radioactivity concentration of ^{40}K , ^{226}Ra and ^{232}Th was found to be within the recommended values.

Key words: NaI(Tl) detector • Detection efficiency • Point and natural standard sources • NORM in soil • Oil fields

INTRODUCTION

Gamma spectrometry is one of the tools commonly used for the measurement of various environmental radionuclides. Ge (Li), HPGe and NaI(Tl) detectors are commonly used to identify and measure activities of radioactive sources. NaI (Tl) detectors are known to have a comparatively poorer resolution than germanium detectors, but are preferable in some specific types of analyses (e.g. identify and measure activities of low-level radioactive sources) due to their relatively high efficiency, besides, they operate under room temperature conditions (i.e. without using of liquid nitrogen as in germanium detectors) [1].

The determination of the activity for each radionuclide requires prior knowledge of the full-energy peak efficiency at each photon energy for a given measuring geometry, which must be obtained by an efficiency calibration using known standard sources or reference mixed gamma source with multiple energy transitions, of exactly the same geometrical dimensions, density and chemical composition of the sample under study. Standard radioactive samples, if available, are costly and would need to be renewed, especially when the

radionuclides have short half-lives [2]. These reference materials should be traceable to a well-known international reference materials manufacturing organization e.g. NIST and IAEA. However, occasionally these kinds of sources might not be available for every laboratory. As a result, an alternative calibration procedure can be employed, namely relative efficiency calibration. [3]. Various experimental and calculation works have been reported for the detection efficiency work [4 - 14].

An international reference material are not available in our laboratory, so we used four experimental methods for obtaining the photo peak efficiency of NaI (Tl) detector and then calculating the radioactivity in soil samples from Ain Zalah oil field.

Experimental Details:

Efficiency Calibration: The Gamma-ray spectrometer consisted of a Scintillation detector of 3.8 cm×2.5 cm NaI(Tl) crystal connected to SPECTCH UCS-20 and computer with a program installed for data acquisition and analysis of the gamma-ray spectrum. The energy calibration of gamma-ray spectrometer was carried out using ^{22}Na , ^{60}Co and ^{137}Cs point sources. The detector and

the sources (or Marinelli beaker) were shielded by 5cm lead surrounding all sides. The absolute detection efficiency of the NaI(Tl) detector was calculated for the energies 1460.8 keV emitted from ^{40}K isotope, 352 keV, 609.3 keV and 1764.5 keV emitted from ^{214}Bi (daughter of ^{226}Ra isotope) and 2614.5 keV emitted from ^{208}Tl (daughter of ^{232}Th series) using four methods and will be discussed in the following:

Method 1- The relative efficiency calibration for NaI (Tl) detector was done in two steps in the energy range from 186.21 keV to 2447.6 keV. First, a relative efficiency curve of the detector was produced using a ^{226}Ra point source [4], with an activity of 2 μCi . The source was counted at three positions from the detector at (0.1, 5, 15) cm distance from the top surface of the detector. Most intensive gamma rays of ^{226}Ra in equilibrium with its daughters have been used. In order to achieve the relative efficiency curve, the relative intensities of the photo peaks corresponding to these gamma ray lines have been experimentally calculated (by dividing the net count of each gamma line by the net count of 609.3 keV gamma transition). The photo peak relative efficiency (R_{\square}) was obtained by dividing the calculated relative intensity of the photo peak with energy (E) on the reference relative intensity of the same photo peak, [3, 15, 16].

$$R = \frac{I_M(E)}{I_R(E)} \quad (1)$$

Where $R(E)$ is the relative efficiency in energy (E), I_M is the relative intensity measured by the detector for the photo peak with energy (E) and, I_R is the reference relative intensity of the same photo peak. The second step for relative efficiency calibration is the normalization of the average relative efficiency curve of the detector to an absolute efficiency value using the same geometry of the measured sample. This step was done using standard solutions of potassium chloride KCl containing 52.3% of natural potassium [15, 16] as a low activity standard source for efficiency calibration of the gamma ray spectrometer in measuring large volumes of low specific activity materials. In this study, the normalization was performed using a known concentration of KCl for the geometrical configuration as the real sample. Using this concentration and the corresponding counting rate, a normalizing factor for ^{40}K ($N.F(^{40}\text{K})$) can be calculated through the following equation: [3, 17]

$$N.F(^{40}\text{K}) = \frac{M * 16.238}{N_K} * T \quad (2)$$

Where, N_K the net count under the peak area at 1460.8 keV gamma line of ^{40}K in the standard solution.

T: The real counting time.

M: Is the mass of KCl ((kg).

The normalizing factor for each gamma line can be calculated as, [3, 17]:

$$N.F_{\gamma} = \frac{R_{\square}(1460.8\text{keV}) * B.R(^{40}\text{K})}{R_{\square} * B.R_{\gamma}} * N.F(^{40}\text{K}) \quad (3)$$

Where, $N.F_{\gamma}$ is the normalizing factor of gamma line corresponding to the peak energy of (γ) radionuclide.

- $R_{\square}(1460.8)$ is the relative efficiency of the gamma line ^{40}K .
- $R_{\square}(\gamma)$ is the relative efficiency of gamma line corresponding to the peak energy of (γ) radionuclide.
- $B.R(^{40}\text{K})$ is the emission probability of the gamma line corresponding to the peak energy of ^{40}K .
- $B.R(\gamma)$ is the emission probability of the gamma line corresponding to the peak energy of (γ) radionuclide.

Method 2- The experimental photo peak efficiencies of NaI (Tl) detector as a function of gamma-ray energies were calculated using ^{22}Na (6564.9 Bq), ^{137}Cs (31941.7 Bq) and ^{60}Co (15742 Bq) point sources placed on the top surface of the detector. The measurement time for each source was 1800 s. The experimental efficiency for the energies 511 keV and 1274.5 keV emitted from ^{22}Na , 661.6 keV emitted from ^{137}Cs and 1173.2 keV, 1332.5 keV and 2505.7 keV emitted from ^{60}Co were calculated using the following formula:

$$\square = \frac{N}{(A.I.T)} \quad (4)$$

Where: \square is the detector efficiency. N is the net counts under the peak of the gamma-line after subtraction of the background under the peak line, A is the radionuclide activity (in Bq), I is the photon emission probability at energy. T is the measurement time (in second).

The absolute detection efficiency of the NaI (Tl) detector at 1460.8 keV gamma line from ^{40}K was calculated by using 250g homogenized powder of standard KCl transferred into a standard Marinelli beaker [15, 16]. The measurement time was 7200 s with the same gamma-ray spectroscopy system under the same condition. The efficiency at 1460.8 keV was calculated using equation (4).

Method 3- Standard samples of ^{232}Th (3.35 Bq), ^{238}U (25.925 Bq) and ^{40}K (125.75 Bq) were prepared in our laboratory by weighing accurate amounts of Thorium

Table 1: The sample number, codes, density and the distance from the oil well head

S. No.	1	2	3	4	5	6	7	8	9	10
Oil well	Az.11a	Az.11b	Az.15	Az.17a	Az.17b	Az.18	Az.21	Az.27a	Az.27b	Az.28
Density(g/cm ³)	1.02	1.06	0.97	1.04	1.11	1.15	1.19	0.93	1	1.09
Distance from well head (m)	25	30	15	2	30	2	5	5	10	20

south. Ain Zalah is a prominent surface anticline consisting of a series of elongated ridges a low range of hills. The rock succession in the area belongs to the Euphrates and lower is characterized by natural fractures and production is dependent on fractures [22].

Sampling: Sediment samples of the TE-NORM were taken from the waste associated with Oil well drilling, production from Ain Zalah region. Table 1 shows the sample number, codes, density and the distance from the oil well head. The sample bulk contained mainly the soil disposed around oil wells originated from drilling process. The samples dried for 4 h at 110°C in an oven to a constant weight and subsequently sieved into equal particle sizes using 1mm sieve. For activity concentration measurement, 250 g of each soil sample was packed in Marinelli beaker, closed tightly, sealed and stored for 1 month to establish the secular equilibrium between the parent radionuclides and their respective daughters, especially between ²²⁶Ra and its Progenies (²²²Rn, ²¹⁴Pb and ²¹⁴Bi). The samples were analyzed non-destructively using NaI(Tl) gamma-ray spectrometry. An empty container identical to that of the sample was also counted in the same counting time under the same geometry to determine the background distribution spectrum.

Calculation of Radioactivity Concentration: According to our procedure for efficiency calibration of NaI(Tl) detector, the activity concentration for any radionuclide (i) is determined through the equation, [3, 17]:

$$C_i = \frac{1}{B.R._\gamma * \epsilon * T * W} * N \quad (5)$$

Where,

- C_i : is the activity concentration of radionuclide (i) (Bq/kg)
- $B.R._\gamma$ (or I mentioned in equation 4): the emission probability of the gamma line corresponding to the peak energy (γ) of the radionuclide (i),
- ϵ : the spectrometer's efficiency corresponding to the peak energy (γ) at the specific geometry,
- N : The net counts under the peak of the gamma-line after subtraction of the background under the peak line.

- W : The mass of the sample (kg).

Equation (5), can be written as, [3, 17]:

$$C_i = \frac{N.F._\gamma * N}{W * T} \quad (6)$$

RESULTS AND DISCUSSION

In method 1(²²⁶Ra point source + standard solution of KCl), rational model fitting efficiency curve was established between the calculated relative efficiency of each gamma line at three positions from the detector surface versus gamma energy as shown in Figure 2. The average values for the relative efficiency obtained from Figure 2 is plotted against the gamma ray and fitted using rational equations as shown in Figure 3. The normalizing factor of gamma line corresponding to the peak energy (352, 609.3, 1460.8, 1764.5 and 2614.5) keV were calculated from equation (3) and listed in Table 2.

In method 2 (²²Na, ⁶⁰Co and ¹³⁷Cs point sources + standard powder of KCl), the experimental efficiencies versus energies were fitted using the power equation as shown in Figure 4. All efficiencies from point sources were normalized to the absolute efficiency at 1460.8 keV (Marinelli beaker) [15, 16] and fitted using the power equation as shown in Figure 4.

In method 3, (standard samples from ²³²Th, ²³⁸U and ⁴⁰K+ Al₂O₃) the efficiencies versus energies were fitted using rational equations as shown in Figure 5.

In method 4, (standard samples from ²³²Th, ²³⁸U and ⁴⁰K+ sandy soil) the efficiencies versus energies were fitted using rational equations as shown in Figure 6.

The absolute efficiency of NaI(Tl) detector as a function of gamma-ray energy from the above four methods with average are shown in Figure 7. It is clear from this figure that the efficiencies obtained from methods 3 and 4 are comparable with the average value. The discrepancy is very pronounced in the low energy region, while in the region of high energies all methods are close to each other.

Table 2: The normalizing factors for different gamma lines

Energy (keV)	352	609.3	1460.8	1764.5	2614.5
Normalization factor	139.91	200.11	1492.2	1160.3	206.06

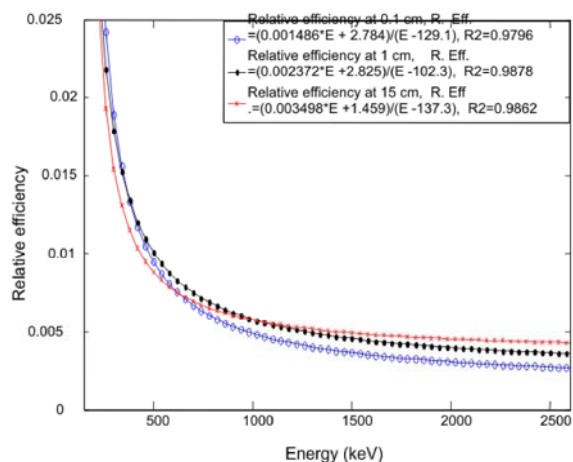


Fig. 2: The relative efficiency curves obtained from ^{226}Ra point source at three distances from the face of the detector as a function of the photon energy

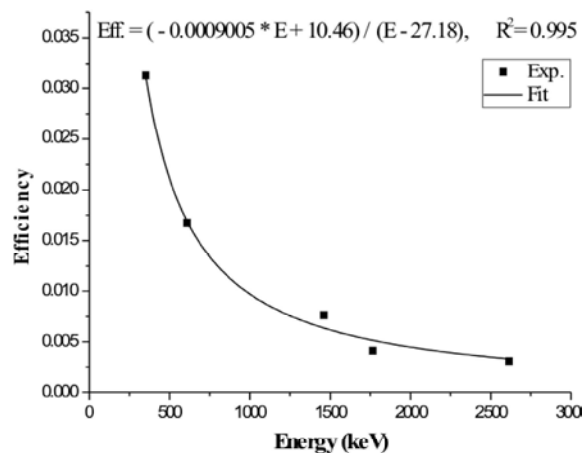


Fig. 5: The absolute efficiency of NaI(Tl) detector as a function of gamma-ray energy from standard samples mixed with Al_2O_3

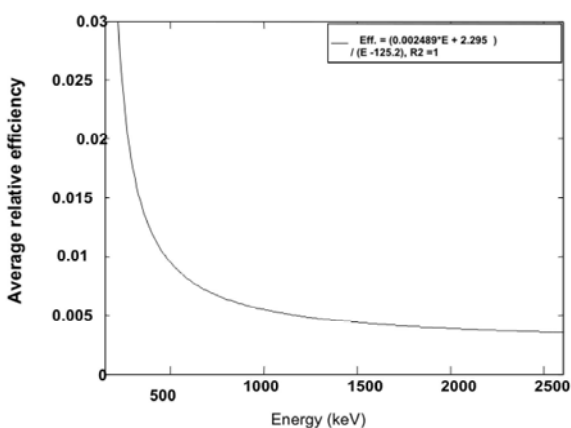


Fig. 3: The average relative efficiency curve as a function of the photon energy

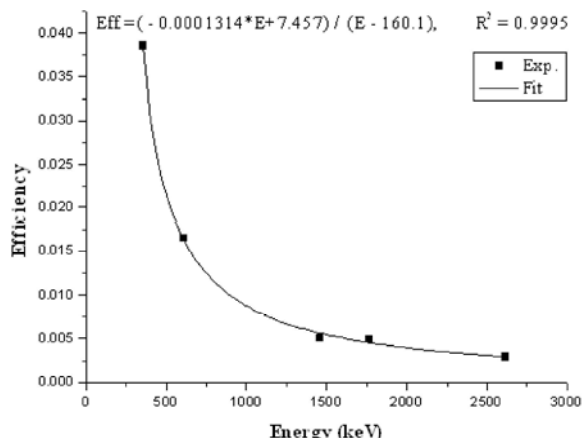


Fig. 6: The absolute efficiency of NaI(Tl) detector as a function of gamma-ray energy from standard samples mixed with sandy soil

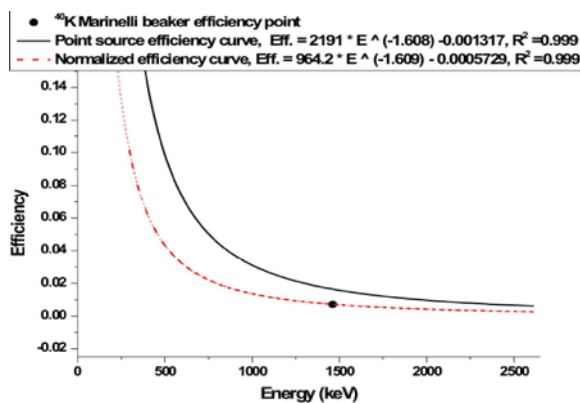


Fig. 4: The efficiency of NaI(Tl) detector as a function of gamma-ray energy from point sources and normalized to the absolute efficiency at 1460.8 keV

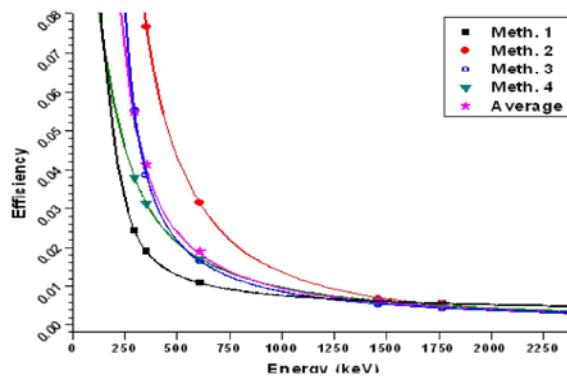


Fig. 7: The absolute efficiency of NaI(Tl) detector as a function of gamma-ray energy from four methods with average

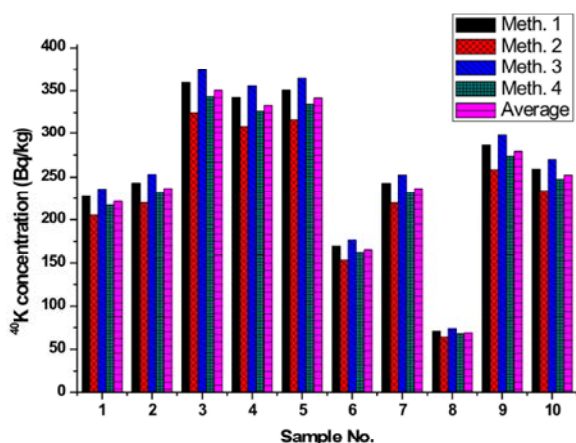


Fig. 8: The distribution of ^{40}K activity concentrations in soil samples from Ain Zalah oil field

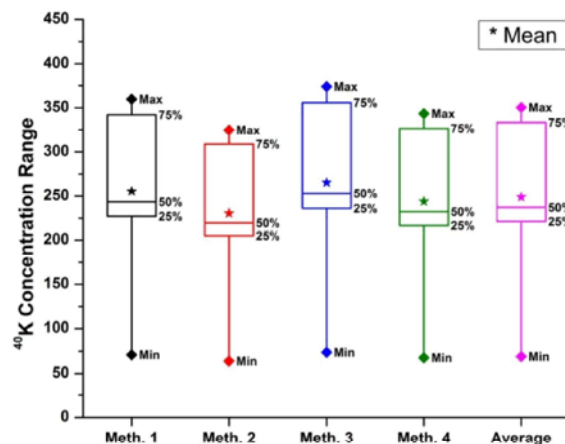


Fig. 11: Box-plot of the activity concentrations for ^{40}K calculated from four methods and compared with the average value

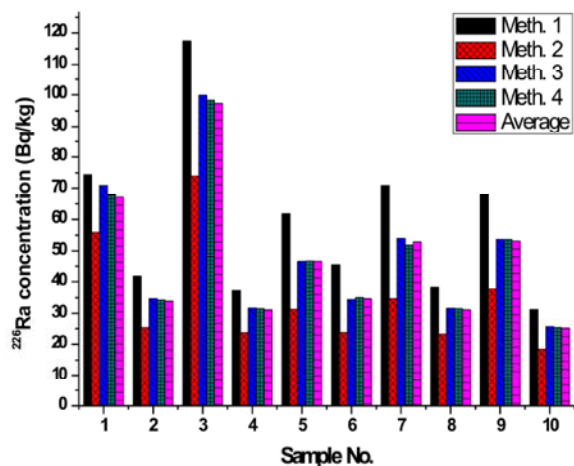


Fig. 9: The distribution of ^{226}Ra activity concentrations in soil samples from Ain Zalah oil field

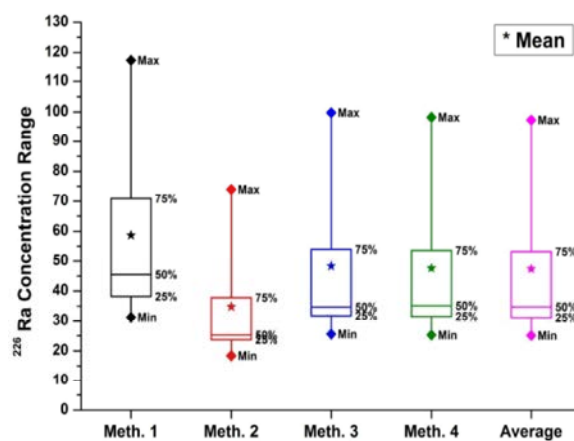


Fig. 12: Box-plot of the activity concentrations for ^{226}Ra calculated from four methods and compared with the average value

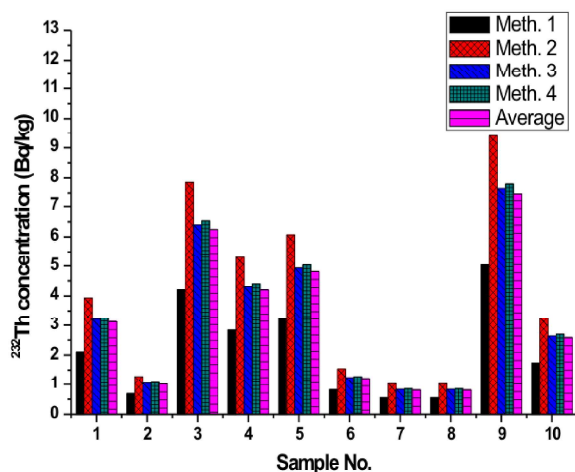


Fig. 10: The distribution of ^{232}Th activity concentrations in soil samples from Ain Zalah oil field

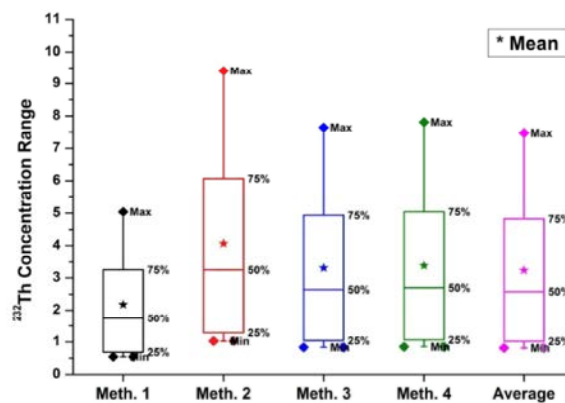


Fig. 13: Box-plot of the activity concentrations for ^{232}Th calculated from four methods and compared with the average value

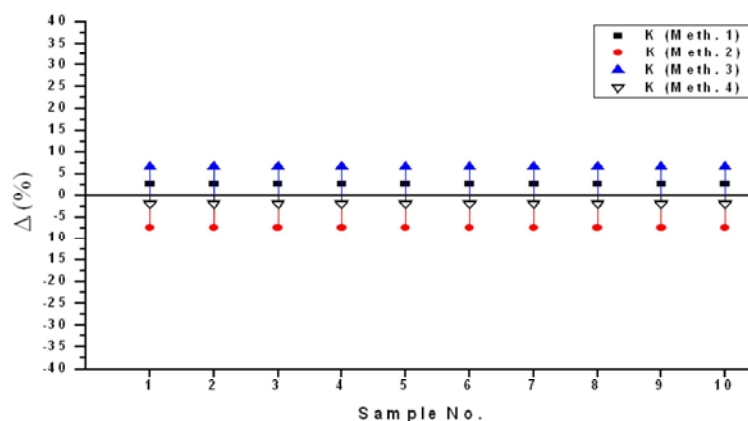


Fig. 14: A comparison between Δ (%) for ^{40}K calculated from four methods in different soil samples

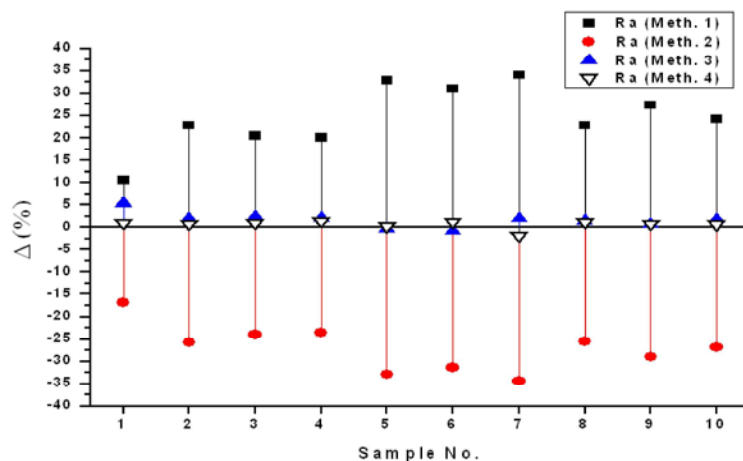


Fig. 15: A comparison between Δ (%) for ^{226}Ra calculated from four methods in different soil samples

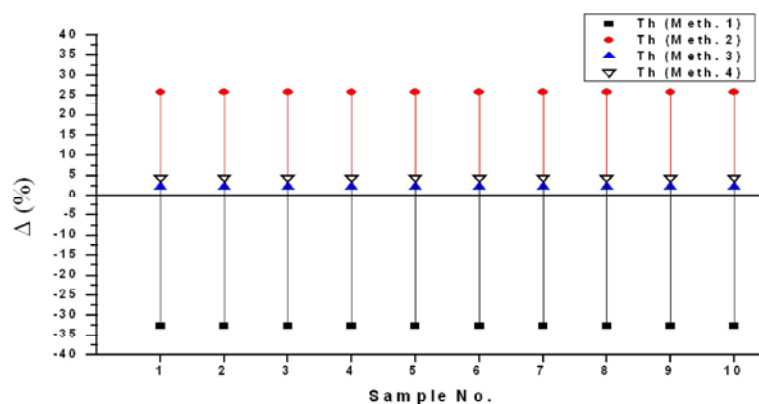


Fig. 16: A comparison between Δ (%) for ^{232}Th calculated from four methods in different soil samples

The distribution of ^{40}K , ^{226}Ra and ^{232}Th activity concentrations in soil samples from Ain Zalah oil field are shown in Figures 8, 9 and 10. A box plots showing the minimum, maximum and average values of activity concentrations for the above isotopes are shown in

Figures 11, 12 and 13. These activity concentrations have been calculated using the detector efficiencies which calculated from four methods. It is obvious from the results shown in Figures 8 and 11 that the ^{40}K concentrations calculated from the four methods were

close to each other and the average values were closer to the values from method 4. A comparison of the ^{226}Ra and ^{232}Th activity concentrations in the soil samples with the average values are shown in Figures 9, 10, 12 and 13. The activity concentration from methods 3 and 4 were closer to the average values, while those from other method were remoter than the average values. The average concentrations of the above isotopes vary between 69.1 Bq/kg (Sample 8) to 350.5 Bq/kg (Sample 3) and from 25.1 Bq/kg (Sample 10) to 97.4 Bq/kg (Sample 3) and from 0.82 Bq/kg (Samples 7 and 8) to 7.5 Bq/kg (Sample 9), respectively. These results were found to be within the recommended values (140 - 850) Bq/kg for ^{40}K , (17 - 60) Bq/kg for ^{226}Ra and (11 - 64) Bq/kg ^{232}Th [23]. The percentage deviations Δ (%) between the calculated C_i and the average activity concentration values C_{Ave} for ^{40}K , ^{226}Ra and ^{232}Th isotopes are calculated by:

$$\Delta (\%) = \frac{C_i - C_{\text{Ave}}}{C_{\text{Ave}}} \quad (7)$$

Figures 14, 15 and 16 show a comparison between Δ (%) for the radioactive isotopes ^{40}K , ^{226}Ra and ^{232}Th in different soil samples from Ain Zalah oil field. The Δ (%) values for ^{40}K isotope calculated from the four methods were close to each other and the average values were closer to the values obtained from method 4 as shown in Figure 14. The higher percentage values Δ (%) for ^{226}Ra and ^{232}Th were found when using methods 1 and 2, while the lower percentage values Δ (%) were found when using methods 3 and 4 as shown in Figures 15 and 16. This may be due to the density of used sandy soil and Al_2O_3 are comparable to studied samples from Ain Zalah oil field. The Δ (%) obtained from methods 1 and 2 are broader than those obtained from methods 3 and 4. It is obvious that methods 3 and 4 are more suitable for efficiency calculations than methods 1 and 2. The Δ (%) obtained for ^{226}Ra and ^{232}Th are broader than those for ^{40}K .

CONCLUSION

Due to the absence of radioactive standard sources (for example IAEA) in our laboratory, we determined the efficiency of NaI(Tl) detector using different methods. The efficiencies obtained from methods 3 and 4 are comparable with the average value. The discrepancy is very pronounced in the low energy region, while they are close to each other in the region of high energies. The Δ

(%) values for ^{40}K isotope calculated from the four methods were close to each other and the average values were closer to the values obtained from method 4. For ^{226}Ra and ^{232}Th isotopes, the lower percentage values Δ (%) were found when using methods 3 and 4. This may be due to the convergence in density between the sandy soil, Al_2O_3 and studied samples from Ain Zalah oil field. Efficiency methods with soil and Al_2O_3 are favorable to those based on point sources, KCl alone. The radioactivity concentration of ^{40}K , ^{226}Ra and ^{232}Th was found to be within the recommended values.

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