Measurement of radiological baseline data of the hazard radiation in bottled drinking water samples in Basrah/Iraq

Riyadh Mnade Ramadhan¹, Abdalrahman Al-Salihi², Alaa Heider Khalaf¹
1. Department of Physics, College of Science, University of Basrah, Basrah, Iraq
2. Department of Basic Sciences, College of Dentistry, University of Basrah, Basrah, Iraq
Corresponding Author Email: rmrsphd@yahoo.com

Abstract
An investigation was carried out on several radiological hazard indices in twenty three bottled drinking water samples that were collected from various locations in Basrah/Iraq. The specific activity of 238U, 232Th, 40K and 137Cs isotopes was measured using the surveillance and measurement (SAM) device, model 940-2G. The specific activity values of U-238, Th-232, K-40 and Cs-137 ranged as (0.029±0.001-3.017±0.003) Bq/l, (0.025±0.002-2.326±0.001) Bq/l, (4.706±0.002-161.560±0.001) Bq/l and (0.040±0.003-0.953±0.001) Bq/l, respectively. The calculation of many risk indices have been carried out for all drinking water samples. All results have agreed with those reported in published studies and all these obtained findings have been recognized to be below the worldwide limit values. Therefore, there is no significant radiological risk in drinking bottled water brands in Basrah governorate, Iraq.

Keywords: Radioactivity; SAM940; Bottled Drinking Water; Basrah Governorate
1. Introduction

The assessment of naturally occurring radioactive materials (NORM) in drinking water provides significant information about the quality of drinking water. This type of health studies permits the calculation of population exposure to radioactivity by the drinking of healthy water [1-6]. Uranium decay chain, thorium decay chain and potassium are classified as naturally occurring radioactive materials (NORM). These isotopes can be easily detected by gamma ray spectroscopy because of their abundant in the natural atmosphere [7, 8]. The world population is subjected to the numerous types of radiation sources including artificial radiation (15%) and natural radiation (85%) in which foodstuffs and drinks contain 11%. This may give a chance to the contamination of radioactive materials [9]. Drinking water can be affected by man-made radiation caesium-137 ($^{137}$Cs) which made through nuclear accidents and processes is an example of anthropogenic radionuclides [10]. The average of doses to many of organs of the human body also represent a significant way for long-term health conditions [11]. Most of people are subjected to these sorts of radiation sources every day and in anywhere. Water is one of the basic constituents of the human diet [12]. Over recent years, bottled drinking water has widely consumed around the world [2]. Thus, a great deal of research has been carried out about the radioactivity of bottled drinking water in different countries [1-7, 13, 14]. For a systematic methodology, this research concentrated on bottled drinking water that is widely consumed by various age groups in Basrah, Iraq. It is very necessary of Iraqi government to control local bottled drinking water to check that are uncontaminated with isotopes. This study is critical in determining the radiation hazard on human and essential in creating procedures and rules involving to radiation protection. It is critical for calculating the radiation levels that affect Iraqi population. That is because the excessive exposure of the radiation may cause major health problems such as carcinogenesis [9]. That is why this study is significant to be done. The measurement of radioactivity in bottled drinking water is very important for monitoring radiation hazards on human health [2, 15]. This scientific research is aimed to create radiological baseline data of the hazard radiation in bottled drinking water samples in Basrah/Iraq. This aim to be achieved the levels of radioactivity and radiation risk indices of consumed bottled drinking water types in Basrah, Iraq are essential to be calculated and investigated.
2. Materials and Methods

Twenty three brands of bottled drinking water were selected and then all samples were bought from local markets in Basrah governorate as showing in

Table 1. The sample collection was made between April and May of 2019. This study was conducted over a four-month period (from April 2019 to July 2019).

| Sample code | Sample commercial name | Sample origin City |
|-------------|------------------------|--------------------|
| W1          | Ayoon                  | Baghdad            |
| W2          | Durat Al-khaleej       | Basrah             |
| W3          | Ala                    | Baghdad            |
| W4          | Alam                   | Basrah             |
| W5          | Al-bakera              | Babylon            |
| W6          | Aljnaaen               | Basrah             |
| W7          | Miah Aljanoob          | Basrah             |
| W8          | Alkafal                | Karbala            |
| W9          | Alkotheer              | Basrah             |
| W10         | Nabee-Alkwthar         | Baghdad            |
| W11         | Alrawase               | Basrah             |
| W12         | Al-Ssad                | Baghdad            |
| W13         | Aquasil                | Basrah             |
| W14         | Barada                 | Basrah             |
| W15         | Hani                   | Baghdad            |
| W16         | Life                   | Sulaymaniayah      |
| W17         | Mowj                   | Muthanna           |
| W18         | Nawar                  | Basrah             |
| W19         | Salsal                 | Basrah             |
| W20         | Al-rawia               | Baghdad            |
| W21         | Veneza                 | Baghdad            |
| W22         | Wadi Mina              | Basrah             |
Each 500 ml of water sample was weighed and put in 500 ml polyethylene plastic Marinelli beakers (pail) and properly stored. The storage period of labeled samples was for 30 days to reach equilibrium between parents and their daughters [16]. The measurements of all samples were carried out by SAM940-2G device operating with NaI(Tl) gamma-ray detector. Surveillance and Measurement (SAM940-2G) operating with BNC 2”x2” gamma-ray NaI(Tl) detector has 256 channels, voltage operation of 600 Volts, coarse gain=1 and fine gain=1.1386. The energy calibration, resolution calibration and efficiency calibration of a BNC 2”x2” NaI (Tl) detector were determined experimentally for (32.90, 661.7, 31.63, 80.90, 356.01, 1173.20 and 1332.50) keV. The calculation of the activity level and presence of $^{238}$U and $^{232}$Th in all samples was derived by arithmetical average of activities took from the peaks of their daughters in the water spectrum. $^{238}$U derived from $^{214}$Bi (609.32 keV) and $^{214}$Pb (295.21 and 351.92 keV). $^{232}$Th derived from $^{212}$Pb, $^{208}$Tl and $^{228}$AC at energies of 238.63, 583.19 and 911.16 keV respectively. The activity values of $^{40}$K in all samples were determined from the single peak of potassium at 1461 keV. The present work has determined the activity values and existence of caesium-137 ($^{137}$Cs) in all samples at energy of 661.61 keV. The measurement time for each sample was 1800 seconds.

The specific activity ($A_s$) was measured applying the following expression [17-19]:

$$A_s = \frac{N}{\varepsilon_f \times P_\gamma \times m \times t_s}$$

where, $N = \text{count per second (cps)}$, $\varepsilon_f = \text{the efficiency at the peak energy}$, $t_s = \text{the live time of the spectrum of sample (1800 seconds)}$, $m = \text{volume (0.5 l)}$ and $P_\gamma = \text{the probability of gamma-ray emission}$.

The rate of absorbed dose which is measured by (nGy/h) is obtained using the indicated relation [20]:

$$D(\frac{nGy}{h}) = 0.461A_U + 0.623A_{Th} + 0.0414A_K$$

where, $A_U$ is the specific activity of $^{238}$U, $A_{Th}$ is the specific activity of $^{232}$Th, and $A_K$ is the specific activity of $^{40}$K.

$^{238}$U ($^{226}$Ra), $^{232}$Th, and $^{40}$K are used to obtain the annual effective dose equivalent as showing in the following equations [20]:

$$\text{AEDE}_{outdoor} (\frac{mSV}{y}) = D \times 8760 \times 0.7 \times 0.2 \times 10^{-6}$$
where, D is the rate of absorbed dose measured in nGy/h. The number of 0.2 refers to outdoor occupancy factor, 0.8 is indoor occupancy factor, 0.7 Sv/Gy is conversion factor. The cancer risk due to gamma radiation effects which is called Excess Lifetime Cancer Risk can be considered as following [19, 21]:

\[ ELCR = AEDE \times DL \times RF \]  

(5)

where, \( AEDE, DL \) and \( RF \) are the equivalent of annual effective dose, the average of human age (70 years) and the factor of risk respectively. The value of risk factor in the public is 0.05 per Sievert as recommended by ICRP for stochastic effects [10, 21].

The activity levels of \(^{238}\)U, \(^{232}\)Th and \(^{40}\)K are inhomogeneous distributed in the water samples. Hence, all samples would be examined by radium equivalent activity. The \( Ra_{eq} \) which measured in Bq l\(^{-1}\) can be measured by the following formula [17, 19, 22]:

\[ Ra_{eq} \left( \frac{Bq}{l} \right) = A_U + 1.43A_{Th} + 0.077A_K \]  

(6)

The internal \( (H_{in}) \) and external hazard \( (H_{ex}) \) indices to gamma ray radiation in water samples were measured applying the following equations [17, 19, 22]:

\[ H_{in} = \frac{A_U}{185} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \]  

(7)

\[ H_{ex} = \frac{A_U}{370} + \frac{A_{Th}}{259} + \frac{A_K}{4810} \]  

(8)

The radiological risk of gamma index \( (I_\gamma) \) is calculated through the following formula [17, 19]:

\[ I_\gamma = \frac{A_U}{300} + \frac{A_{Th}}{200} + \frac{A_K}{3000} \]  

(9)

Alpha index was measured by applying the following relation [17, 19] :

\[ I_\alpha = \frac{A_{Ra}}{200} \]  

(10)

where \( A_{Ra} \) are the specific activity of \(^{226}\)Ra supposed in equilibrium with the specific activity of \(^{238}\)U.

3. Results and Discussion

The specific activity of \(^{238}\)U, \(^{232}\)Th, \(^{40}\)K and \(^{137}\)Cs in different types of water samples has been measured using equation 1 and their results are reported in Table 2. The specific activity
of $^{238}\text{U}$ was ranged from (0.029±0.001) Bq l$^{-1}$ to (3.017±0.003) Bq l$^{-1}$. As for $^{232}\text{Th}$, was ranged from (0.025±0.002) Bq l$^{-1}$ to (2.326±0.001) Bq l$^{-1}$. For $^{40}\text{K}$ was ranged from (4.706±0.003) Bq l$^{-1}$ to (161.560±0.001) Bq l$^{-1}$. As for $^{137}\text{Cs}$, it was ranged from (0.040±0.003) Bq l$^{-1}$ to (0.953±0.001) Bq l$^{-1}$. The average specific activity of $^{40}\text{K}$ is higher than those of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{137}\text{Cs}$. The world average limit specific activity values of $^{238}\text{U}$, $^{232}\text{Th}$, $^{40}\text{K}$ and $^{137}\text{Cs}$ are 33 Bq l$^{-1}$, 45 Bq l$^{-1}$, 412 Bq l$^{-1}$ and 101 Bq l$^{-1}$, respectively [23, 24].

The gamma absorbed dose rates which calculated by using equation 2 for all samples in this study were ranged between 0.098 nGy/h and 7.610 nGy/h as shown in Table 3. The outcomes obtained for absorbed dose rates appeared lower than the international limit value of 58 nGy/h reported in UNSCEAR 2000 [20]. Taking into account a 20% outdoor and 80% indoor occupancy factors, the AEDE$_{\text{outdoor}}$ and AEDE$_{\text{indoor}}$ values were measured for all water samples in this work as shown in Table 3. The mathematical calculations of these quantities were achieved using equations 3 and 4. The lowest value of both AEDE$_{\text{outdoor}}$ and AEDE$_{\text{indoor}}$ was 0.000 mSv/y whereas the highest values of AEDE$_{\text{outdoor}}$ and AEDE$_{\text{indoor}}$ were 0.009 mSv/y and 0.037 mSv/y, respectively as shown in Table 3. The achieved results demonstrate that the AEDE in all samples do not appear higher than the world average annual effective dose equivalent. The estimated world average AEDE$_{\text{outdoor}}$ and AEDE$_{\text{indoor}}$ are 0.07 mSv/y and 0.34 mSv/y respectively, as recommended by UNSCEAR 2000 [20]. The outdoor and indoor excess lifetime cancer risk (ELCR) values were measured for all samples as shown in Table 3. The mathematical calculations of these quantities were done using equation 5. The consequences obtained show that the ELCR in all water samples appeared below the world average excess lifetime cancer risk. The estimated world average ELCR$_{\text{outdoor}}$ of $0.29 \times 10^{-3}$ and ELCR$_{\text{indoor}}$ of $1.4 \times 10^{-3}$ reported in UNSCEAR 2000 [10, 25]. The radium equivalent activity, internal and external radiation hazard indices, the gamma index and alpha index were calculated by applying the equations 6, 7, 8, 9 and 10 respectively. There is a variation in the values of these radiation hazard indices in all samples as reported in Table 4.

Fig. 1 and Fig. 2. The results of all risk indices are not higher than world limit values.
Table 2: Specific activity results of $^{238}$U, $^{232}$Th, $^{40}$K and $^{137}$Cs in water samples

| Sample code | Specific activity ($A_s$) in (Bq l$^{-1}$) ($\pm$ Uncertainty) |
|-------------|---------------------------------------------------------------|
|             | $^{238}$U | $^{232}$Th | $^{40}$K | $^{137}$Cs |
| W1          | ND       | 0.489±0.004 | ND       | ND        |
| W2          | ND       | 2.326±0.001 | 34.508±0.002 | ND        |
| W3          | 0.211±0.001 | 0.914±0.002 | ND       | ND        |
| W4          | 1.084±0.001 | 1.593±0.002 | 4.706±0.002 | ND        |
| W5          | 0.211±0.001 | 0.914±0.002 | ND       | ND        |
| W6          | 1.242±0.008 | 0.790±0.003 | 10.196±0.002 | ND        |
| W7          | 0.404±0.001 | 0.025±0.002 | ND       | ND        |
| W8          | 0.052±0.005 | 1.441±0.003 | 161.560±0.001 | ND        |
| W9          | 0.211±0.002 | 0.914±0.002 | ND       | ND        |
| W10         | ND       | 0.157±0.005 | ND       | 0.179±0.003 |
| W11         | ND       | 0.157±0.005 | ND       | 0.179±0.003 |
| W12         | 0.948±0.001 | 1.921±0.002 | 71.369±0.001 | 0.953±0.001 |
| W13         | 0.424±0.009 | 0.727±0.002 | 9.411±0.002 | 0.258±0.003 |
| W14         | 0.029±0.001 | 0.482±0.002 | 30.587±0.002 | 0.357±0.002 |
| W15         | 0.200±0.001 | 0.402±0.001 | ND       | ND        |
| W16         | 0.701±0.001 | 1.301±0.003 | 43.919±0.002 | ND        |
| W17         | 0.029±0.004 | 0.482±0.001 | 30.587±0.002 | 0.357±0.003 |
| W18         | 3.017±0.003 | 1.307±0.003 | 36.861±0.002 | ND        |
| W19         | 0.245±0.002 | 0.941±0.002 | 21.960±0.002 | 0.119±0.004 |
| W20         | 0.029±0.007 | 0.482±0.002 | 30.587±0.002 | 0.357±0.003 |
| W21         | 1.150±0.004 | 0.672±0.001 | 79.996±0.007 | 0.040±0.003 |
| W22         | ND       | 0.620±0.002 | ND       | ND        |
| W23         | 0.178±0.003 | 1.118±0.004 | 6.274±0.002 | 0.179±0.003 |
| Minimum     | 0.029±0.001 | 0.025±0.002 | 4.706±0.002 | 0.040±0.003 |
| Maximum     | 3.017±0.003 | 2.326±0.001 | 161.560±0.001 | 0.953±0.001 |

*ND: Not detected
Table 3: The results of gamma absorbed dose rates (D), annual effective dose equivalent values (AEDE) and excess lifetime cancer risk values (ELCR) in all samples

| Sample code | D (nGy/h) | AEDE\textsubscript{Outdoor} (mSv/y) | AEDE\textsubscript{Indoor} (mSv/y) | ELCR\textsubscript{Outdoor} | ELCR\textsubscript{Indoor} |
|-------------|-----------|------------------------------------|-----------------------------------|-----------------------------|-----------------------------|
| W1          | 0.305     | 0.000                              | 0.001                             | 0.001                       | 0.005                       |
| W2          | 2.878     | 0.004                              | 0.014                             | 0.012                       | 0.049                       |
| W3          | 0.667     | 0.001                              | 0.003                             | 0.003                       | 0.011                       |
| W4          | 1.687     | 0.002                              | 0.008                             | 0.007                       | 0.029                       |
| W5          | 0.667     | 0.001                              | 0.003                             | 0.003                       | 0.011                       |
| W6          | 1.487     | 0.002                              | 0.007                             | 0.006                       | 0.026                       |
| W7          | 0.202     | 0.000                              | 0.001                             | 0.001                       | 0.003                       |
| W8          | 7.610     | 0.009                              | 0.037                             | 0.033                       | 0.131                       |
| W9          | 0.667     | 0.001                              | 0.003                             | 0.003                       | 0.011                       |
| W10         | 0.098     | 0.000                              | 0.000                             | 0.000                       | 0.002                       |
| W11         | 0.098     | 0.000                              | 0.000                             | 0.000                       | 0.002                       |
| W12         | 4.589     | 0.006                              | 0.023                             | 0.020                       | 0.079                       |
| W13         | 1.038     | 0.001                              | 0.005                             | 0.004                       | 0.018                       |
| W14         | 1.580     | 0.002                              | 0.008                             | 0.007                       | 0.027                       |
| W15         | 0.342     | 0.000                              | 0.002                             | 0.001                       | 0.006                       |
| W16         | 2.952     | 0.004                              | 0.014                             | 0.013                       | 0.051                       |
| W17         | 1.580     | 0.002                              | 0.008                             | 0.007                       | 0.027                       |
| W18         | 3.731     | 0.005                              | 0.018                             | 0.016                       | 0.064                       |
| W19         | 1.608     | 0.002                              | 0.008                             | 0.007                       | 0.028                       |
| W20         | 1.580     | 0.002                              | 0.008                             | 0.007                       | 0.027                       |
| W21         | 4.261     | 0.005                              | 0.021                             | 0.018                       | 0.073                       |
| W22         | 0.386     | 0.000                              | 0.002                             | 0.002                       | 0.007                       |
| W23         | 1.038     | 0.001                              | 0.005                             | 0.004                       | 0.018                       |
| Minimum     | 0.098     | 0.000                              | 0.000                             | 0.000                       | 0.002                       |
| Maximum     | 7.610     | 0.009                              | 0.037                             | 0.033                       | 0.131                       |
Table 4: The results of radium equivalent activity, radiation hazard (internal, external, gamma and alpha) indices in all samples

| Sample code | Ra$_{eq}$ (Bq l$^{-1}$) | $H_{in}$ | $H_{ex}$ | $I_{\gamma}$ | $I_{\alpha}$ |
|-------------|------------------------|---------|---------|-------------|-------------|
| W1          | 0.700                  | 0.002   | 0.002   | 0.002       | 0.000       |
| W2          | 5.984                  | 0.016   | 0.016   | 0.023       | 0.000       |
| W3          | 1.518                  | 0.005   | 0.004   | 0.005       | 0.001       |
| W4          | 3.723                  | 0.013   | 0.010   | 0.013       | 0.005       |
| W5          | 1.518                  | 0.005   | 0.004   | 0.005       | 0.001       |
| W6          | 3.157                  | 0.012   | 0.009   | 0.011       | 0.006       |
| W7          | 0.439                  | 0.002   | 0.001   | 0.001       | 0.002       |
| W8          | 14.553                 | 0.039   | 0.039   | 0.061       | 0.000       |
| W9          | 1.518                  | 0.005   | 0.004   | 0.005       | 0.001       |
| W10         | 0.225                  | 0.001   | 0.001   | 0.001       | 0.000       |
| W11         | 0.225                  | 0.001   | 0.001   | 0.001       | 0.000       |
| W12         | 9.191                  | 0.027   | 0.025   | 0.037       | 0.005       |
| W13         | 2.188                  | 0.007   | 0.006   | 0.008       | 0.002       |
| W14         | 3.074                  | 0.008   | 0.008   | 0.013       | 0.000       |
| W15         | 0.774                  | 0.003   | 0.002   | 0.003       | 0.001       |
| W16         | 5.944                  | 0.018   | 0.016   | 0.023       | 0.004       |
| W17         | 3.074                  | 0.008   | 0.008   | 0.013       | 0.000       |
| W18         | 7.724                  | 0.029   | 0.021   | 0.029       | 0.015       |
| W19         | 3.281                  | 0.010   | 0.009   | 0.013       | 0.001       |
| W20         | 3.074                  | 0.008   | 0.008   | 0.013       | 0.000       |
| W21         | 8.271                  | 0.025   | 0.022   | 0.034       | 0.006       |
| W22         | 0.887                  | 0.002   | 0.002   | 0.003       | 0.000       |
| W23         | 2.260                  | 0.007   | 0.006   | 0.008       | 0.001       |
| Minimum     | **0.225**              | **0.001** | **0.001** | **0.001** | **0.000** |
| Maximum     | **14.553**             | **0.039** | **0.039** | **0.061** | **0.015** |
The findings show that the specific activity of $^{238}\text{U}$, $^{232}\text{Th}$, $^{40}\text{K}$ and $^{137}\text{Cs}$ in all water samples appeared lower than the world average specific activity values. The higher average specific activity of $^{40}\text{K}$ compared with the average activity of $^{238}\text{U}$, $^{232}\text{Th}$ and $^{137}\text{Cs}$ was expected because of its natural presence [26]. The levels of background and the detection limits of technique may conceal minor peaks of $^{238}\text{U}$, $^{232}\text{Th}$ [27]. Previous studies reported that the detection of $^{238}\text{U}$ and $^{232}\text{Th}$ is not necessary to be found in all food samples [28, 29].
existence of $^{137}$Cs in water samples may because of the Chernobyl accident fallout, the usage of contaminated water bottles [8].

The variability in the unique behaviour of isotopes in the water samples of different Iraqi cities may be due to their geographical and geological conditions.

3. Conclusions

This study investigates the calculation of dose levels and determines the specific activity of $^{238}$U, $^{232}$Th, $^{40}$K and $^{137}$Cs in all samples of drinking water that were collected from various locations in Basrah / Iraq. Therefore, it can be concluded that there is no significant radiological risk in drinking bottled water brands in Basrah governorate, Iraq.
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قياس النشاط الإشعاعي للمياه المعبأة بالقناني في البصرة/ العراق

رياض منادي رمضان1، عبد الرحمن الصالحية2، علاء حيدر خلف2

1قسم الفيزياء، كلية العلوم، جامعة البصرة، البصرة، العراق
2قسم العلوم الأساسية، كلية طب الأسنان، جامعة البصرة، البصرة، العراق

المستخلص

الهاتف من هذه الدراسة قياس النشاط الإشعاعي في قناتي مياه الشرب المستهلكة في البصرة/ العراق، وحساب العديد من مؤشرات الخطر الإشعاعي. جمعت 23 عينة من من قناتي مياه الشرب من الأماكن المحلية لمختلف المناطق في محافظة البصرة. حدد النشاط الإشعاعي (بيكيل / لتر) لنظامي البودوم 238 و البودوم 232 والسبيروم 40 والسبيروم 137 في عينات مياه الشرب المعبأة بواسطة جهاز قياس وترابي الإشعاع (SAM940). يشغيل برامج حاسوبية خاصة يتم من خلالها التحكم بالجهاز عن بعد وإجراء جميع التحليلات للأطوار. النشاط الإشعاعي (بيكيل / لتر) لنظامي البودوم 238 و البودوم 232 والسبيروم 40 والسبيروم 137 هي (0.003±0.003-0.001±0.008) بيكيل / لتر، (0.025±0.002-2.325±0.001) بيكيل / لتر، (0.029±0.001-3.017±0.003) بيكيل / لتر، (4.706±0.002-161.560±0.001) بيكيل / لتر. 

حسب القدرة هي تتمثل في حساب حساسية الطرحة للماء، والتي تم اجرائها على تكسير الذرات في حساسية الإشعاع، حيث أن التكلفة من مؤشرات الخطر الإشعاعي لجميع العينات في هذه الدراسة. تمّت مقارنة نتائج الدراسة الحالية بالدراسات السابقة وقيمت الرد الأقصى في جميع أنحاء العالم. وقد اتفقت جميع النتائج مع تلك الموصوفة في الدراسات السابقة، وتبين أن جميع النتائج ضمن الحدود المسموح بها عالمياً. ومن ثمّ، فإنّ الاتجاهات من المواد الغذائية هي أهم يدلا الاستهلاك في محافظة البصرة. يمكن استخدام نتائج هذه الدراسة في توفير قاعدة بيانات أساسية لمعدلات الإشعاع الطبيعي والصناعي في المواد الغذائية الأساسية المستهلكة في البصرة/ العراق.

كلمات مفتاحية: النشاط الإشعاعي، جهاز قياس وترابي الإشعاع (SAM940)، قناتي مياه الشرب، محافظة البصرة