Jurnal Sains Nuklear Malaysia, 2019, 31(2): 7 – 13
ISSN: 2232-0946

COMPARISON OF AUTO VERSUS MANUAL CALCULATION OF ^{40}K ACTIVITY CONCENTRATION IN POWDERED MILK MEASURED USING GAMMA SPECTROMETRY SYSTEM

Yii Mei-Wo, Maziah Mahmud and Zal U'yun Wan Mahmood

Radiochemistry and Environment Group, Waste and Environmental Technology Division
Malaysian Nuclear Agency, 43000 Kajang, Malaysia
Correspondence author: yii@nuclearmalaysia.gov.my

ABSTRACT

Gamma spectrometry counting system nowadays equipped with commercial analysis software to calculate the radionuclide activity automatically but manual calculation still a common practice in some laboratories. This paper reported and compared the activity concentration of ^{40}K in 120 powdered milk samples that had been quantified and calculated using both automatic and manual approaches. From software calculation, ^{40}K radioactivity found varies from 212 Bq/kg to 508 Bq/kg with the average of 294 ± 73 Bq/kg and the activity uncertainty associated with samples ranged of 1.9% to 2.8%. Meanwhile, from the manual calculation, ^{40}K radioactivity varies from 206 Bq/kg to 501 Bq/kg with the average of 285 ± 73 Bq/kg and the uncertainty of manual calculations were between 6.8% to 7.0%. Almost all results (except two samples) showed that the auto calculation had activity higher than that obtained from the manual calculation. Overall all samples activities had a difference of less than 9 percent between the auto and manual calculation. However, U-score calculation showing none of the difference between the auto and manual value was significant.

Keywords: Auto and manual activity calculation, gamma spectrometry, potassium-40, powdered milk, U-score

INTRODUCTION

Nowadays, most nuclear analytical instruments are designed very user-friendly and supplied along with the commercial software to help users to perform analysis of the raw data or spectrum and generate reports. Gamma spectrometry counting system is one of such that widely being used to identify gamma-emitting radionuclides quantitatively. A modern gamma spectrometry is normally equipped with commercial gamma-ray spectrum analysis software which can automatically identify and calculate the radionuclide activity. With the advent of computer technology, data analyses become much faster and easier as compared to the time when it was first invented. This computers software has the advantage of resolving overlapping peaks by fitting analytical curves to the collected data and also capable of handling large amounts of data rapidly and accurately. A gamma spectrometry counting system is normally supplied with a multi-channel analyzer (MCA) emulation software, as a standard accessory, and the gamma-ray spectrum analysis software, as an optional accessory (Zaharudin et al., 2007).

The MCA emulation software is the basic software that enables the system to distribute pulses collected in memory base on the distribution of the number of pulses with respect to pulse height. These distributions will usually be displayed as a spectrum. This type of software has limitations as the establishment of energy and efficiency calibration, the analysis of spectrum such as, peak identification, marking of peak areas, and activity calculation, have to be done manually. Whereas, the gamma-ray spectrum analysis software is capable to analyze the γ-spectrum for the photo-peak position and relative intensity of the respective photo-peak is proportional to the measured radionuclides activities. The software then will automatically identify and calculates the activity of the respective radionuclides in the samples based on the ready built-in library. While purchasing a gamma spectrometry system nowadays, both emulation software and gamma-ray spectrum analysis software are supplied together as a standard package (Zaharudin et al., 2007). Despite the advantages and attractive features of the computerized system, the gamma-ray spectrum analysis software also had its limitation such as flexibility in the estimation of the measurement uncertainty and performing density correction.

Potassium is a very significant mineral to the body, important to both cellular and electrical function. It is one of the main blood minerals called "electrolytes" along with the others such as sodium and chloride, and it carries a tiny electrical positive charge (potential) (Elson, 2011). According to Alan (2018), potassium is one of the seven essential macro-minerals to the body. The human body requires at least 100 milligrams of potassium daily to support key processes in the body. Potassium-40 (^{40}K) makes up about 0.012% (120 ppm) of the total amount of potassium found in nature. It is a radioactive isotope of potassium which has a very long half-life of 1.251×10^9 years. Potassium-40 is the largest source of natural radioactivity in the body of animals including humans. For instance, a 70 kg human body contains about 140 grams of potassium, hence about $0.000117 \times 140 = 0.0164$ grams of ^{40}K; whose decay produces about 4,300 disintegrations per second (Becquerel) continuously throughout the life of the body, i.e. equivalent to about 30.7 Bq/g of potassium (Wikipedia, 2018). The study was carried out earlier by Yii et al. (2018), reported that ^{40}K activity about 29.26 ± 1.20 Bq/g of potassium in powdered milk.

Potassium-40 decays to ^{40}Ca by emitting a beta particle with no attendant gamma radiation (89% of the times) and the gas ^{40}Ar by electron capture with the emission of an energetic gamma ray (11% of the times). So, ^{40}K can present both external and internal health hazards. The strong gamma radiation (E_γ = 1.46 MeV) makes the external exposure to this radioisotope a big concern; when inside the body, ^{40}K poses a health hazard from the beta particles emission ($E_{\beta Max}$ = 1.35 MeV) and gamma rays which associate with cell damage and general potential for subsequent cancer induction (Afshari et al., 2009).

Kolapo (2014) reported that many elements or compounds such as metals and metalloids will accumulate along the food chain. Their concentrations in the environment grow with the increase of urban, agricultural, and industrial emissions. The presence of metal aids their entry into the food chain and thereby increases the toxicity effects of the food in humans and animals diet. Milk is one of the most important foods for human nutrition; it is beneficial in the human diet and mostly needed by infant and children during their growing age.

Due to the strong penetrating and discrete gamma radiation energy of ^{40}K, it can be easily measured and quantified using gamma-ray spectrometry. This article reported the activity of ^{40}K found in powdered milk samples estimated by using the commercial software and compared them with the manual calculation value obtained by using the raw data. Differences between these two approaches were discussed to seek the reliability of the commercial software. A total of one hundred twenty powdered milk samples were measured in this study under the same conditions using the same single spectrometry over a year.

MATERIALS AND METHODS

Milk Sample Collection and Preparation

A total of one hundred twenty powdered milk samples were used in this study. For each sample, a portion of the sample (approximately 300 g) was taken and weighted. The powdered milk was dried overnight in an electric scientific oven at 85^{o}C. On the following day, powdered milk was allowed to cool to the room temperature, re-weighted to determine the moisture content, and was transferred and compressed into a 250 ml size marinelli beaker, sealed with thick PVC tape to inhibit radon from escaping (Zal U'yun et al., 2017). The samples weight between 215 – 260 g with a density between 0.58 – 0.87 g/cc. All samples were stored for a period in excessive of 30 days to establish secular equilibrium between ^{226}Ra and ^{228}Ra and their respective radioactive progenies prior to gamma counting (Dowdall and O'Dea, 2002; Yang et al., 2005). [In this paper, only the activity of ^{40}K was reported and discussed but the measurements of gamma emitting radionuclides were carried out simultaneously].

Gamma Spectrometry Counting

A sample spectrum was individually measured with the gamma-ray spectrometry consisting of a high-purity germanium (HPGe) setup and multichannel analyzer of 16,384 channels. The detector used is a coaxial 3 inches diameter closed end, closed facing window geometry with vertical dipstick operated at 1,500 HV bias supplies. The detector is shielded in a chamber made of lead, cadmium, and copper (total thickness 11 cm) to reduce the background radioactivity. This p-type detector is designed to provide 25% relative efficiency with the FWHM resolution of 820 eV at 122 keV gamma-ray line of ^{57}Co and 1.85 keV at 1332 keV gamma-ray line of ^{60}Co. It was calibrated using procedures as reported earlier by Yii et al. (2009) using customized gamma multinuclides standard source comprising of ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co in same counting geometry. Source used was manufactured by Isotope Products Laboratories, USA (source no. 1895-82). A similar geometry container filled with deionized water was measured over every weekend to determine the background counts.

All samples were counted for 54,000 seconds using spectrometer and corrected for density and date of sampling. Counting times are long enough to ensure a 2σ counting error of less than 10%. Previous studies reveal that minimum counting time of 10 hours was sufficient to provide adequate counts under the various gamma-ray peaks (Ahmed and El-Arabi, 2005; Arogunjo et al., 2005; El-Reefy et al., 2006). The activity of ^{40}K and its 1σ uncertainty was calculated directly *via* its energy peak (El-Reefy et al., 2006; Yang et al., 2005) using the commercial software ORTEC Gamma Vision version 6.01. For manual calculation, a peak was marked manually from the centroid to its left-right baseline (Compton) as guide given by Gilmore and Hemingway (1998) and IAEA (1989), total counts were divided with the live counting time to get the peak count rate. Same marking range was performed on the background spectrum, the background counts were divided with the background live counting time for the background count rate. The difference between the peak count rate and the background count rate is the net count rate from which the specific activity of ^{40}K and its 1σ uncertainty was calculated with a spreadsheet using equation as reported in Zaharudin et al. (2007). Minimum detectable activity (MDA) was set at 2 Bq/kg after considering the sample size and counting time of ^{40}K (Zal U'yun et al., 2017).

Jurnal Sains Nuklear Malaysia, 2019, 31(2): 7 – 13
ISSN: 2232-0946

RESULTS AND DISCUSSION

Potassium-40 behaves the same as other potassium isotopes in the environment, being assimilated into tissues of all plants and animals through normal biological processes. It is the predominant radioactive component in human tissues and in most food. For example, milk contains about 74 Bq/L of natural ^{40}K (Baeza et al., 2004; Argonne National Laboratory, 2005). Ingestion of contaminated foods is one of the routes of uptake of potentially dangerous radionuclides for man and dairy products in particular due to importance in human diets (Baeza et al., 2004).

In this study, the moisture contents in the powdered milk were found to vary between 1.4 – 3.9%. Primary count rate under the photopeak of ^{40}K was analyzed and calculated automatically (auto) by the system using standard gamma radioactivity calculation equations of the commercial software. The activity concentration and the associated uncertainty (1σ) of the radionuclide were reported after normalized for one kilogram mass of each sample. For manual calculation, peak area was marked manually from the centroid of the peak to both edge of the peak, activity concentration and its associated uncertainty was calculated using excel worksheet.

The activity concentration of ^{40}K for both auto (blue dot) and manual (orange dot) calculations were presented in Figure 1 below together with its 1σ uncertainty. For software calculation, the values of ^{40}K radioactivity vary from 212 Bq/kg to 508 Bq/kg with an average of 294 ± 73 Bq/kg. The activity uncertainty was different between each sample and it was ranged of 1.9% to 2.8% which mainly due to counting statistic. Meanwhile, from the manual calculation, the values of ^{40}K radioactivity vary from 206 Bq/kg to 501 Bq/kg with an average of 285 ± 73 Bq/kg and the uncertainty of 6.8% to 7.0%. Most of the results showed that auto calculation estimating higher activity value than that from the manual calculation. However, from Figure 1, it found that none of the points had a significant difference as the range of activity concentration of ^{40}K from the auto and manual calculation was overlapping each other. The uncertainty percentage for manual calculation was higher / bigger than that from auto because more sources of uncertainties were included in the manual calculation.

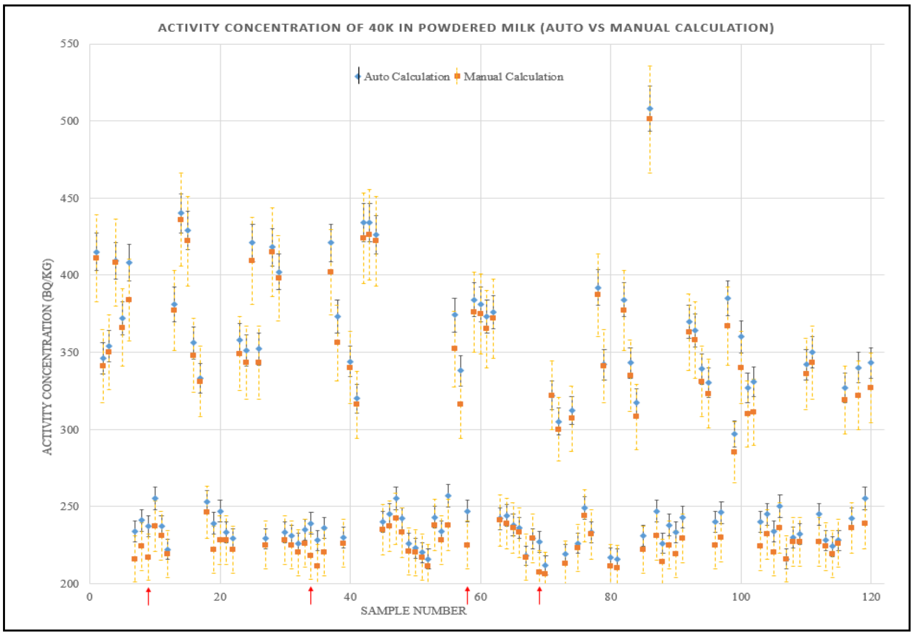

Figure 1: Activity concentration of ^{40}K in powdered milk from software and manual calculation

The activity different (by percentage) between the software and manual calculation were summarized in Table 1. Overall all samples have a difference that not more than 9 percent. Sixty-five percent of the samples had a difference of less than 3 percent between the auto and manual calculation and over 90 percent of the samples had a difference that below 7 percent. There were four samples which have high activity different (by percentage) between the software and manual calculation (points were indicated by red arrow in Figure 1) and the biggest activity different (by percentage) in one of the sample was 8.8 percent where the software reported activity was 247 ± 7 Bq/kg but the manual calculation estimated only 225 ± 16 Bq/kg. In order to evaluate how significant was the difference between these two values, the U-score calculation equation as reported by Zaharudin et al. (2007) was used. It was found that the calculated U-score value was 1.27, which is less than 1.64, indicating that there was no significant difference between these two values as according to the definition given by Khater et al. (2002). The U-score calculation for all data set reveals that U-score values fall between 0.00 – 1.27 showing there was no significant difference among the results. Therefore, it can be concluded that none of the different value obtained between the auto and manual calculation was a significant difference.

Table 1: Percentage different of ^{40}K activity concentration from auto and manual calculation

Percentage Different	**Number of Samples**	**Percentage of Samples (%)**
< 1 %	14	11.7
≥ 1 but < 2%	33	27.5
≥ 2 but < 3%	31	25.8
≥ 3 but < 4%	4	3.3
≥ 4 but < 5%	5	4.2
≥ 5 but < 6%	12	10.0
≥ 6 but < 7%	10	8.3
≥ 7 but < 8%	7	5.8
≥ 8 but < 9%	4	3.3
Total	**120**	**100 %**

CONCLUSIONS

The ^{40}K activity concentration in one hundred twenty powdered milk samples had been quantified and calculated using both commercial (auto) and manually and then be compared. From software calculation, ^{40}K radioactivity varies from 212 Bq/kg to 508 Bq/kg with the average of 294 ± 73 Bq/kg. The activity uncertainty for the samples ranged between the 1.9 to 2.8 percent. Meanwhile, from the manual calculation, ^{40}K radioactivity found varies from 206 Bq/kg to 501 Bq/kg with an average of 285 ± 73 Bq/kg and uncertainty lies between 6.8 to 7.0 percent. Almost all results have auto calculation activity concentration higher than the manual calculation. Overall all samples have a difference that not more than 9 percent and sixty-five percent of the samples have a difference of less than 3 percent between the auto and manual calculation. However, comparing the different values using U-score showing none of the difference between the auto and manual calculation was significant.

ACKNOWLEDGEMENTS

The authors would like to express appreciations to Malaysian Nuclear Agency and also thankful to staff in the group for their kind co-operation and support to implement this study.

REFERENCES

Afshari, N.S., Abbasisiar, F., Abdolmaleki, P. and Nejad, M.G. (2009). Determination of ^{40}K concentration in milk samples consumed in Tehran-Iran and estimation of its annual effective dose, *Iran. J. Radiat. Res*. 7: 159-164.

Ahmed, N.K. and El-Arabi, A.G.M. (2005). Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, upper Egypt, *J. Environ. Radioact.* 84: 51-64.

Alan, C. (2018). Everything you need to know about potassium, access online https://www.medicalnewstoday.com/articles/287212.php on 13th July 2018.

Argonne National Laboratory, EVS. (2005). Human health fact sheet, August 2005.

Arogunjo, A.M., Ofuga, E.E. and Afolabi, M.A. (2005). Levels of natural radionuclides in some Nigerian cereals and tubers, *J. Environ. Radioact.* 82: 1-6.

Baeza, A., Corbacho, J.A. and Miro, C. (2004). Temporal evaluation of natural and man-made radioactivity levels in milk samples: dosimetry implications, *Bull. Environ. Contam. Toxicol*, 72: 547-556.

Dowdall, M. and O'Dea, J. (2002). $^{226}Ra/^{238}U$ disequilibrium in an upland organic soil exhibiting elevated natural radioactivity, *J. Environ. Radioact.* 59: 91-104.

El-Reefy, H.I., Sharshar, T., Zaghloul, R. and Badran, H.M. (2006). Distribution of gamma-ray emitting radionuclides in the environment of Burullus Lake: I. Soils and vegetation, *J. Environ. Radioact.* 87: 148-169.

Elson, M.H. (2011). Role of potassium in maintaining health, access online https://hkpp.org/patients/potassium-health on 13th July 2018.

Gilmore, G. and Hemingway, J. (1998). Practical gamma-ray spectrometry, First Ed. *John Wiley & Sons Ltd*, West Sussex, England. 314p.

IAEA. (1989). Measurement of radionuclides in food and the environment, *Technical Report Series No. 295*, IAEA, Vienna, 61-63.

Khater A.E.M., Ebaid Y.Y., El-Mongy S. and El-Tahawy, M.S. (2002). Determination of ^{238}Pu and $^{239+240}$Pu by destructive assays, Proceedings of an International Conference, Karlshure; Contributed Paper.

Kolapo, A.A. (2014). Assessments of natural radioactivity and heavy metals in commonly consumed milk in Oke-Ogun area, Nigeria and estimation of health risk hazard to the population, *J. Environ. Anal. Toxicol.* 4: 253-257.

Wikipedia (2018). Potassium-40, access online https://en.wikipedia.org/wiki/Potassium-40 on 13th July 2018.

Yang, Y.X., Wu, X.M., Jiang, Z.Y., Wang, W.X., Lu, J.G., Lin, J., Wang, L.M. and Hsia, Y.F. (2005). Radioactivity concentrations in soils of the Xiazhuang granite area, China, *Appl. Radiat. Isot*. 63: 255-259.

Yii, M.W., Zaharudin, A. and Abdul-Kadir, I. (2009). Distribution of naturally occurring radionuclides activity concentration in East Malaysian marine sediment, *Appl. Radiat. Isot*. 67: 630-635.

Yii, M.W., Maziah, M., Dainee, N.F.A.T., Nor-Aza, H., Mohd-Izzat, M.R., Siti, N.H.M.N., Zal U'yun, W.M. (2018). The ^{40}K activity of one gram potassium in milk powder, *in proceeding of Seminar R & D*, Nuklear Malaysia, Bangi.

Zaharudin, A., Yii, M.W. and Abdul-Kadir, I. (2007). Method of software validation for Cs-137 and Ra-226 activity measurements in environmental samples using gamma spectrometry system: In *Validation Procedures of Software Applied in Nuclear Instruments*, IAEA-TECDOC-1565, IAEA, Vienna, 55-63.

Zal U'yun, W.M., Yii, M.W., Mohd-Ashhar, K., Khairuddin, M.K., Abdul-Kadir, I. and Mohd-Abd-Wahab, Y. (2017). Assessment of natural radioactivity level and radiological index in the vicinity of Lynas rare-earth processing plants, *ASEAN J. Sci. Technol. Dev*. 34: 67-78.