ALKÜ Fen Bilimleri Dergisi 2019, Özel Sayı (NSP 2018): 165-173

Estimation of Measurement Uncertainty for ^{230}Th Radioisotope Analysis in Soil

Meryem Seferinoğlu[1,2*], Pınar Esra Erden[1,3]
[1]*Turkish Atomic Energy Authority Sarayköy Nuclear Research and Training Centre, Turkey.*
[2]*Sinop University Faculty of Engineering and Architecture, Department of Nuclear Energy Engineering, Turkey.*
[3]*Ankara Hacı Bayram Veli University Polatlı Faculty of Science and Arts, Department of Chemistry, Turkey.*
**mseferinoglu@sinop.edu.tr*
pinarerden@hotmail.com

Abstract

The uncertainty of a measurement is the interval of values within which the true value is believed to lie with a stated probability when all sources of error have been taken into account and is therefore represents the reliability of a measurement. In this study, we evaluate the measurement uncertainty in the determination of 230Th in the soil sample using a radiochemical separation procedure and alpha spectrometry. All the sources in the analysis method that contribute to the combined uncertainty were identified. The uncertainty associated with a certain parameter was used to determine its partial contribution to the combined standard uncertainty of the measurand. The spreadsheet and full matematical approach was used to present the combined standard uncertainty. slakta, ALKÜ Fen Bilimleri Dergisi sistemine değerlendirme için yüklenecek olan makaleler için yazım formatı tanımlanmıştır. Metin biçimlendirmesi, tablo ve resim başlıkları ve referanslar ile ilgili bilgilere bu şablon üzerinden erişebilirsiniz. MS Word formatındaki orijinal araştırma makaleleri ve derleme makaleleri bu talimatlara uygun olarak yazılacaktır.

Keywords: Measurement Uncertainty, Thorium Isotopes, Alpha Spectrometry

1. INTRODUCTION

All measurements of radioactivity are subject to uncertainty, and the result of a measurement is only valid if it contains a measured value accompanied by an expression of the associated a realistic uncertainty [1]. The uncertainty in the radiometric measurement has generally been defined based on the standard deviation of repeated measurements. The standard deviation given by the square root of the number of counts reflects only the statistical uncertainty of counting. The uncertainties associated with many other sources is essential to be taken into account the assessment of the measurement results [2]. Nowadays, assessment of the measurement uncertainty and design of an uncertainty budget are receiving increasing attention. However, quantifying measurement uncertainty and emphasizing it in a true form are not straightforward tasks and there is no common method for the estimation of measurement uncertainty. The Guide to the Expression of Uncertainty in Measurement (GUM), published by ISO, provides general rules for evaluating and expressing uncertainty in measurement [3]. On the other hand, some European documents report further details on uncertainty calculations in quantitative chemical analysis. There are different methods to calculate uncertainty such as the analytical approach which is only applicable in relatively simple assess,

the GUM uncertainty approach which is an approximation to the analytical method and the Monte Carlo simulation approach which proposes a practical solution for complicated models [4]. However, the application of these procedures to environmental radioactivity analysis involving radiochemistry does not as intelligible as it must be, because the estimation of uncertainty requires the analyst to take into account all steps of the analytical procedure and all possible sources of uncertainty.

Routinely radionuclides monitoring is not only very important in the nuclear industry but also in the non-nuclear industry for protection of worker's health and safety issues related with harmful radiation doses exposure to the employees. There are several non-nuclear industries where employees are exposed to radioactive materials such as thorium and its daughter nuclides [5]. So the accurate and precise measurement of the activity concentration of Th isotopes is of great interest due to the radiotoxic effect of these isotopes [6]. The aim of this work is to investigate a procedure for the estimation of the measurement uncertainty associate with activity concentration of ^{230}Th in soil determined by alpha spectrometry with radiochemical separation procedure. For this purpose, the steps in the radiochemical procedure that contribute considerably to the combined measurement uncertainty were defined. The uncertainty associated with a certain parameter was used to determine its partial contribution to the combined total uncertainty. Finally, the uncertainty budget was expressed in a clear and simple manner.

2. EXPERIMENTAL

The analytical procedure for soil involves; i) sample preparation step contains ashing, acid digestion and dissolution, ii) radiochemical separation step for purification of thorium, iii) alpha source preparation step for transferring the purified thorium fraction to solid phase and iv) counting of alpha source.

2.1 Sample preparation

The analytical procedure was performed using proficiency test exercise soil sample (TAEK RMB-2015-01) of TAEK-2015 proficiency test. 2.0 g of soil was weighed into a porcelain capsule with a microbalance calibrated with SI traceable weights. A weighed amount of ^{229}Th tracer was incorporated into the soil sample at the beginning of the analysis to determine the radiochemical yield throughout the alpha source preparation method. The sample was ashed at 600 ºC in a microwave furnace for 16 h to destroy the organic matter. Ashed samples were transferred into a Teflon beaker and dissolved with 20 mL concentrated HNO_3 and 10 mL concentrated HCl by stirring and heating up to 225 ºC until the solution became almost clear. The mixture was boiled and stirred for 3.0 h, let to cool and centrifuged. The solid residue after centrifugation was added 10 mL concentrated HNO_3 and 15 mL concentrated HF and heated up to 250 ºC for 1 h, let to cool and centrifuged. 10 mL concentrated HF was added to the residue, boiled for 3 h and centrifuged. The supernatant fractions of the three centrifugations were combined into a clean Teflon beaker and evaporated near to dryness. 5 mL of concentrated HNO_3 was added to the residue and evaporated to dryness. Finally, 10 mL 3 M HNO_3/1 M $Al(NO_3)_3$ was used to re-dissolve the residue.

Radiochemical separation is necessary for isolation of thorium isotopes from all other interfering radioactive and non-radioactive elements existing in the soil sample. Radiochemical separation step is mainly based on the isolation of thorium from other radionuclides such as U, Pu, Am and Np in the soil sample using TEVA (100-150 µm particle size) resin prior to the counting by alpha spectrometry. The Eichrom technologies ACW10 VBS analytical procedure used for thorium separation in water was modified in order to measure the thorium activity in soil [7]. The schematic diagram of radiochemical separation is given in Fig. 1. The TEVA column was placed on the vacuum box and adjust the vacuum to achieve the flow rate of 1.0 mL/min. After precondition of column by 5 mL of 3 M HNO_3 solution, the sample solution was poured into the column and allowed the solution pass through the TEVA resin. Thorium is retained on the resin while uranium, americium and neptunium (v) are removed with load solution and 3 M HNO_3 rinses. The column was washed with 30 mL of 3 M HNO_3 and finally 15 mL of 9 M HCl solution was load into column to strip thorium fraction. The purified thorium solution was evaporated near to dryness.

The alpha source was prepared on the stainless steel discs using electrodeposition method described by the Eichrom ACW03 VBS (Eichrom, 2005) analytical procedure [8].

The alpha source was counted by alpha spectrometer with Passivated Implanted Planar Silicon (PIPS) detector with active surface area of 450 mm2. The energy calibration was carried out using an electroplated mixed standard alpha source consists of ^{238}U, 1.67 Bq (± 5.2%), ^{234}U, 1.63 Bq (± 4.9%), ^{239}Pu, 1.91 Bq (± 5.5%), and ^{241}Am, 1.81 Bq (± 5.5%) supplied by Eckert & Ziegler Isotope Products.

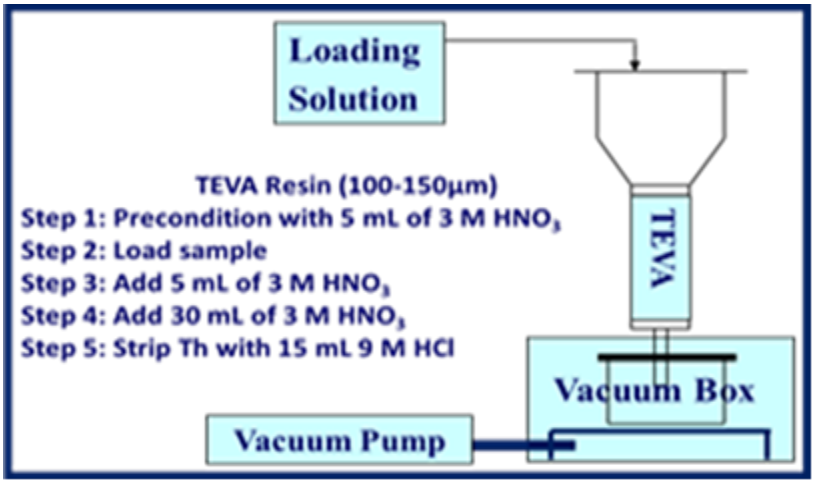

FIG. 1. Radiochemical separation of thorium isotopes using vacuum box system

3. RESULTS AND DISCUSSION

3.1 Qualification of measurement uncertainty associated with ^{230}Th activity concentration

The total measurement uncertainty is evaluated based on Equation 1 combined with Equation 2 which are describe the relationship of ^{230}Th activity concentration with variables considered in the accurate measurement. While A_A shows that the amount of activity on the electroplate source, a_A shows the activity concentration of ^{230}Th isotopes in the soil. The activity concentration of ^{230}Th isotope in the soil is calculated using equation 1[9].

$$a_A = \frac{A_A}{m_s} f_1 f_2 f_3 f_4 \qquad in \ \frac{Bq}{Kg} \tag{1}$$

where a_A is activity concentration of ^{230}Th referred to the sampling date in Bq kg^{-1}, A^A is the amount of activity of ^{230Th} on the electroplated source in Bq, m_s is the mass of sample, f_1 is correction for decay of ^{230}Th in the time interval from the end of sampling to the beginning of the measurement, f_2 is correction for decay of ^{230}Th during the counting interval (t_G), f_3 are correction for decay of ^{229}Th in the time interval from the calibration date of tracer to the beginning of the measurement and f_4 is correction for decay of ^{229}Th during the counting interval (t_G). f correction is generally very close to unity in case of ^{230}Th ($T_{1/2}$ = (7.538 ± 0.030) x 10^4 a) and ^{229}Th ($T_{1/2}$ = (7.34 ± 0.16) x 10^{3}a).

The activity concentration of ^{230Th} isotope on the electroplated source is calculated with following equation;

$$A_A = C_T m_T \left(\frac{R_{GA} - R_{BA}}{R_{GT} - R_{BT}} - q_1\right) \left(\frac{P_\alpha T}{P_\alpha A}\right) \qquad in \ Bq \tag{2}$$

where C_T is the activity concentration of tracer (^{229}Th) solution, m_T is the mass of added tracer, R_{GA} is the gross counting rate of ^{230}Th and R_{GT} is the gross counting rate of ^{229}Th, R_{BA} blank counting rate of ^{230}Th, R_{BT} is the black counting rate of ^{229}Th, q_1 is the isotopic impurity ratio of ^{230}Th isotopes in the tracer, $P_\alpha T$ and P_α A sum of α-emission probabilities of the α-lines of ^{229}Th and ^{230}Th. The activity concentration of thorium-230 isotope in the soil was calculated as 2200.4 Bq Kg^{-1}.

3.1.1. Uncertainty components associate with ^{230}Th activity on the electroplated source U(A_A)

The uncertainty of activity on the electroplate source includes the uncertainties of chemical recovery, measured counting rate of analyte and tracer, blank counting rate of analyte and tracer, isotopic impurities of analyte in standard tracer solution and sum of emission probabilities of analyte and tracer.

Uncertainty connected with chemical recovery contains the uncertainties based on the trace activity of the standard reference solution taken from the certificate of reference material supplied by the producer ($U(C_T)$=0.00072 Bq g^{-1} (0.75%), the coverage factor, k=1) and mass of tracer. The uncertainty of mass of tracer is calculated using the equation $U(w_i)$= 8.16 x 10^{-5} + 5.35 x 10^{-6} R supplied by the calibration certificate of the analytical balance, in which R is the weight of standard ^{229}Th solution.

Uncertainty associated with measured gross counting rates of analyte peak and tracer peak in the sample alpha energy spectrum for acquisition time t_G are estimated by following equations, respectively.

$$U(R_{GA}) = \sqrt{\left(\frac{R_{GA}}{t_G} + \frac{R_{bgd(Th-230)}}{t_B}\right)}, \quad U(R_{GT}) = \sqrt{\left(\frac{R_{GT}}{t_G} + \frac{R_{bgd(Th-229)}}{t_B}\right)} \qquad (3)$$

In our case, the spectral resolution was enough to use simple peak area integration of gross channel counts of analyte and tracer. The sample alpha energy spectrum is given in Figure 2.

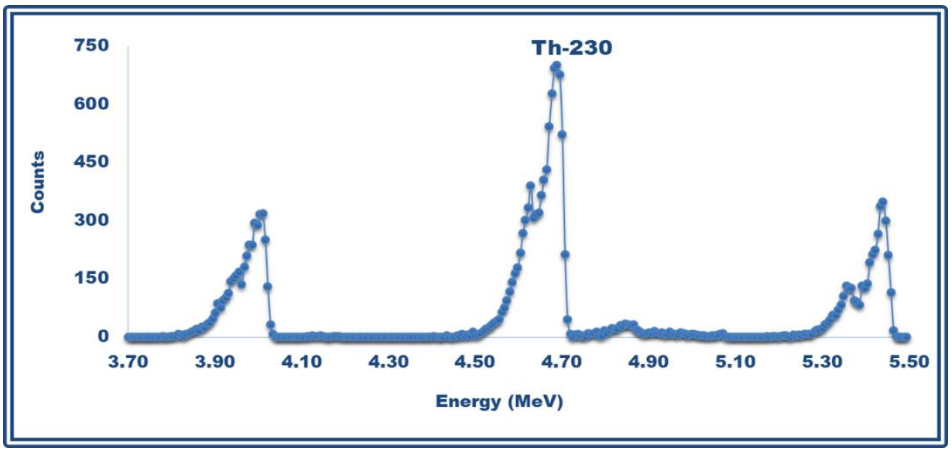

FIG. 2. The alpha energy spectrum of soil sample obtained by alpha spectrometry

Black counting rates, RBT of 230Th and 229Th radioisotopes and their uncertainties, U(RBT) are estimated from measurements of a series of n prepared blank sources and calculated using below equations.

$$R_{BT} = \sqrt{\frac{\sum_{i=1}^{n} R_{BT,i}}{n}}, \qquad U(R_{BT}) = \sqrt{\frac{\sum_{i=1}^{n}(R_{BT,i} - R_{B(Th-229)})^2}{n-1}} \tag{4}$$

Isotopic impurity of ^{230}Th isotopes in the tracer standard solution (q1) is obtained from measurements of a few separately electroplated tracer sources. Thus, the isotopic impurity is determined as an average ratio of the net count rate of ^{230}Th to net count rate of ^{229}Th. The uncertainty due to isotopic impurity is estimated by repeated measurements and is calculated as 2.4 x 10^{-4} Bq g^{-1}. Uncertainty of the sum of emission probabilities of radionuclides are found as U(P□T)/P□T = 0.0032 for ^{229}Th and U(P□A)/P□A = 0.0049 for ^{230}Th taken from IAEA recommended data.

3.1.2. Uncertainty components associate with the activity concentration of ^{230Th} U(a_A)

The uncertainty components associated with activity concentration of ^{230}Th can be expressed as a function of definition weighting uncertainty and decay correction factors uncertainties. Uncertainty associated with sample mass is calculated as 5.8 x 10^{-8} kg used the equation taken from balance calibration certificate. Uncertainties of decay correction factors of ^{230}Th and ^{229}Th isotopes depending on the half-live of radionuclide are calculated by using equation 5 and 6.

$$U(f_1) = f_1(t_S - t_E)U(\lambda_A), \qquad U(f_2) = f_2(1 - f_2 \exp(-\lambda_A t_G))\frac{U(\lambda_A)}{\lambda_A} \tag{5}$$

$$U(f_3) = f_3(t_S - t_C)U(\lambda_T), \qquad U(f_4) = f_4(1 - f_4 \exp(-\lambda_T t_G))\frac{U(\lambda_T)}{\lambda_T} \tag{6}$$

where t_s is the beginning of measurement time, t_E is the end of sampling time, t_C is calibration time of tracer radionuclide solution and t_G represent the counting period. λ_A and u(λ_A) are the decay constant of ^{230}Th and its uncertainty, respectively, λ_T and $U(\lambda_T)$ are the decay constant of ^{229}Th and its uncertainty, respectively. Table 1 shows quantities and individual uncertainties of all parameter in the equation 1 combine with equation 2.

TABLE I. Quantities and individual uncertainties of all parameter in the equation 1 combine with equation 2

Parameters	***Value***	***Standard Uncertainty***
m_s (kg)	0.001	5.8 x 10^{-8}
f_1	1.000	6.1 x 10^{-9}
f_2	1.000	3.7 x 10^{-11}
f_3	0.999	1.4 x 10^{-5}
f_4	1.000	2.1 x 10^{-9}
C_T (Bq g^{-1})	0.096	7.2 x 10^{-4}
m_T (g)	2.020	3.3 x 10^{-5}
R_{GA} (cps)	1.40 x 10^{-6}	1.5 x 10^{-3}
R_{BA} (cps)	4.63 x 10^{-6}	3.5 x 10^{-6}
R_{GT} (cps)	1.06 x 10^{-2}	4.1 x 10^{-4}
R_{BT} (cps)	1.02 x 10^{-5}	4.8 x 10^{-6}
q_1	1.02 x 10^{-3}	2.4 x 10^{-4}
$P_\alpha T$	0.889	5.0 x 10^{-3}
$P_\alpha A$	0.997	3.2 x 10^{-3}

3.1.3. Combined standard measurement uncertainty

The combined standard uncertainty associated with ^{230}Th activity concentration in the soil can be calculated with full mathematical approach or spreadsheet approach.
The combine standard measurement uncertainty associated with activity concentration of ^{230}Th isotope in the soil was calculated as 95.9 Bq kg^{-1} (4.36 %) with the following equations (Eq. 7 - 9).

$$U(A_{230_{Th}}) = A_{230_{Th}}\left[\left(\frac{u(C_T)}{C_T}\right)^2 + \left(\frac{u(m_T)}{m_T}\right)^2 + \left(\frac{U(y)}{y}\right)^2 + \left(\frac{u(P_\propto A)}{P_\propto A}\right)^2 + \left(\frac{u(P_\propto T)}{P_\propto T}\right)^2\right]^{1/2} \quad (7)$$

$$y = \frac{R_{GA} - R_{BA}}{R_{GT} - R_{BT}} - q_1$$

The uncertainty of y value related with counting rates is evaluated with below equation.

$$U(y) = \left[\left(\frac{R_{GA} - R_{BA}}{R_{GT} - R_{BT}}\right)\left[\left(\frac{U^2(R_{GA}) + U^2(R_{BA})}{(R_{GA} - R_{BA})^2}\right) + \left(\frac{U^2(R_{GT}) + U^2(R_{BT})}{(R_{GT} - R_{BT})^2}\right)\right] + U^2(q_1)\right]^{1/2} \quad (8)$$

The combined measurement uncertainty related with activity concentration of ^{230} Th is calculated by equation 9.

$$U(a_{230_{Th}}) = a_{230_{Th}} \left[\left(\frac{u(A_{230_{Th}})}{A_{230_{Th}}} \right)^2 + \left(\frac{u(m_s)}{m_s} \right)^2 + \left(\frac{U(f_1)}{f_1} \right)^2 + \left(\frac{U(f_2)}{f_2} \right)^2 + \left(\frac{U(f_3)}{f_3} \right)^2 + \left(\frac{U(f_4)}{f_4} \right)^2 \right]^{1/2} \quad (9)$$

The combined standard measurement uncertainty associated with activity concentration of ^{230}Th isotope in the soil was also calculated as 88.3 Bq kg^{-1} (4.01 %) according with spreadsheet approach [10]. The calculation of the combined standard measurement uncertainty by the spreadsheet approach is given in Table 2. Table 2 shows that this approach gives a good estimation on the total uncertainty.

The percentage contribution of each components into the overall uncertainty can be also calculated by spreadsheet method. The method allows to observe the correlations between the parameters and percentage contribution of different uncertainty components all in one table. The percentage contribution of each parameter into the total uncertainty on the activity concentration of ^{230}Th is given in Table 3. Consequently, the main uncertain sources in the total uncertainty value associated with activity concentration of thorium isotopes is uncertainties due to the counting rates of ^{230}Th and ^{229}Th. Especially the uncertainty due to the tracer counting rate (^{229}Th) is the biggest contributor on the overall standard uncertainty.

TABLE 2. Combined standard measurement uncertainty calculation associated with the activity concentration of ^{230}Th isotope in the soil sample.

a_A (Bq/Kg)=2200.4	m_s	f_1	f_2	f_3	f_4	C_T	m_T	R_{GA}	R_{BA}	R_{GT}	R_{BT}	q_I	$P_{\alpha T}$	$P_{\alpha A}$
	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001	0.001
	1.000	**1.000**	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
	1.000	1.000	**1.000**	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
	0.999	0.999	0.999	**0.999**	0.999	0.999	0.999	0.999	0.999	0.999	0.999	0.999	0.999	0.999
	1.000	1.000	1.000	1.000	**1.000**	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000	1.000
	0.096	0.096	0.096	0.096	0.096	**0.097**	0.096	0.096	0.096	0.096	0.096	0.096	0.096	0.096
	2.020	2.020	2.020	2.020	2.020	2.020	**2.020**	2.020	2.020	2.020	2.020	2.020	2.020	2.020
	0.140	0.140	0.140	0.140	0.140	0.140	0.140	**0.142**	0.140	0.140	0.140	0.140	0.140	0.140
	4.6E-06	4.6E-06	4.6E-06	4.6E-06	4.6E-06	4.6E-06	4.6E-06	4.6E-06	**8.1E-06**	4.6E-06	4.6E-06	4.6E-06	4.6E-06	4.6E-06
	1.1E-02	1.1E-02	1.1E-02	1.1E-02	1.1E-02	1.1E-02	1.1E-02	1.1E-02	1.1E-02	**1.1E-02**	1.1E-02	1.1E-02	1.1E-02	1.1E-02
	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	1.0E-05	**1.5E-05**	1.0E-05	1.0E-05	1.0E-05
	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	1.0E-03	**1.3E-03**	1.0E-03	1.0E-03
	0.889	0.889	0.889	0.889	0.889	0.889	0.889	0.889	0.889	0.889	0.889	0.889	**0.894**	0.889
	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	0.997	**1.000**
Varied Results	2200.3	2200.4	2200.4	2200.4	2200.4	2216.9	2200.5	2223.8	2200.4	2118.1	2201.4	2200.4	2212.8	2193.4
Residuals	1.2E-01	-1.3E-05	-8.2E-08	-3.2E-02	-4.5E-06	-1.7E+01	-3.6E-02	-2.3E+01	5.4E-02	8.2E+01	-1.0E+00	3.9E-02	-1.2E+01	7.0E+00
Square of Residuals	1.5E-02	1.8E-10	6.7E-15	1.0E-03	2.0E-11	2.7E+02	1.3E-03	5.5E+02	2.9E-03	6.8E+03	1.0E+00	1.6E-03	1.5E+02	4.9E+01
Sum of Squares	7.8E+03													
Unc.	**88.3**	**4.01%**		**By Spreadshete Approch**			**Variation**							
Unc.	**95.9**	**4.36%**		**By Mathematical Formula**			**0.34%**							

TABLE 3. The percentage contribution of each component on the overall measurement uncertainty of Th isotope activity concentration in the soil.

Parameters	***% Contribution***
m_s	0.0
f_1	0.0
f_2	0.0
f_3	0.0
f_4	0.0
C_T	3.5
m_T	0.002
R_{GA}	7.0
R_{BA}	0.0
R_{GT}	86.9
R_{BT}	0.01
q_1	0.0
$P_\alpha T$	1.95
$P_\alpha A$	0.62

4. CONCLUSION

The evaluation procedure of measurement uncertainty associated with the activity concentration of Th-230 isotope in soil are discussed. The application of alpha spectrometry with radiochemical procedure to environmental radioactivity analysis and the related uncertainties do not as intelligible as it must be, because the estimation of uncertainty requires the analyst to take into account all steps of the analytical procedure and all possible sources of uncertainty.

In this study, the uncertainty components contribute to the total standard measurement uncertainty of Th-230 activity on soil are identified and quantified for each steps in the analytical analysis procedure. The spreadsheet approach and full mathematical approach are used for calculation of the combine standard measurement uncertainty.

The application of spreadsheet approach is easier method than mathematical approach and gives a good agreement within the value obtained from full mathematical approach. Main uncertainty sources were identified as uncertainties associated with the counting rates which are very difficult to reduce.

5. ACKNOWLEDGEMENTS

The authors would like to thank to Turkish Atomic Energy Authority, Radiation Metrology laboratory for supplying soil sample.

REFERENCES

[1] Vreček P., Benedik L., "Evaluation of measurement uncertainty in the determination of 210Pb and 210Po using beta counting and alpha spectrometry" Accred. Qual. Assur. 8, 134-137 (2003).

[2] Heydorn, K., "Evaluation of the uncertainty of environmental measurements of radioactivity" Journal of Radioanalytical and Nuclear Chemistry. 262, No. 1 249-253 (2004).

[3] ISO, 1995. "ISO Guide to the expression of uncertainty in measurement" International Organization for Standardization, Geneva,ISBN:92-67r-r10188-9 (Available online from http://www.bipm.org/en/publications/guides/gum.html)

[4] Guimarães Couto, P. R., Damasceno, C. D., and Pinheiro de Oliveira, S., "Monte Carlo Simulations Applied to Uncertainty in Measurement" Theory and Applications of Monte Carlo Simulations. Chapter 2, 27-51, DOI: 10.5772/53014.

[5] Modna, D. K., Jerome, S. M., White, M. A., Woods, M. J., "Thorium in workplace measurement intercomparison". Applied Radiation isotopes, 53, 265-271 (2000).

[6] Jia, G., Torri, G., Ocone, R., Di Lullo, A., De Angelis, A., & Boschetto, R., "Determination of thorium isotopes in mineral and environmental water and soil samples by α-spectrometry and the fate of thorium in water" Applied Radiation and Isotopes, 66 (10), 1478-1487, (2008).

[7] Eichrom Technologies, LLC Method No: ACW10VBS Analytical Procedure Revision: 1.1 May 1, 2014

[8] Eichrom Technologies, LLC Method No: ACW03 Analytical Procedure Revision: 2.2 May 1, 2014

[9] Kanisch, G., "Alpha-spectrometric analysis of environmental samples" (IAEA-TEC-DOC-1401). Quantifying Uncertainty in Nuclear Analytical Measurements. International Atomic Energy Agency, Vienna, pp. 127–139 (2004).

[10] Holmes, L., "Alpha Spectrometric Analysis of Environmental Samples Spreadsheet approach" (IAEA-TEC-DOC-1401) Quantifying Uncertainty in Nuclear Analytical Measurements. International Atomic Energy Agency, Vienna, pp. 114–149 (2004).