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Mass Attenuation Coefficients of IAEA Soil Standards at Different Gamma-ray Energies

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ABSTRACT

Self-attenuation correction is very important in gamma-ray spectrometry of large samples. This paper presents the measurement of mass attenuation coefficients for three IAEA soil samples (IAEA-447, IAEA-TEL-2011-03, and IAEA-TEL-2012-03). It has been shown that the transmission method is important when the chemical composition of a sample is not known. For samples with established chemical composition, NIST offers WinXCom which provides mass attenuation coefficients based on the photon interaction data. WinXCom was applied and mass attenuation coefficients for the world average soil and IC-2018-01 soil were estimated. The mass attenuation coefficients required for the self-attenuation correction of soil samples for the determination of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs have also been presented.

1. Introduction

Gamma-ray spectrometry is the most commonly used technique for the determination of both natural and anthropogenic radionuclides, qualitatively as well as quantitatively. For activity measurement, nature and counting geometry of the sample play vital roles. The nature of the sample includes its chemical composition, particle size, and density. Counting geometry means sample size, its dimensions, sample to detector distance, and other factors. Choosing suitable geometry depends upon the level of activity present in a sample. The counting geometry means whether the source is point-like or volumetric and if it is counted at close or far distance. For low-level activity measurements, the choice of large sample size is unavoidable which leads to correction for self-attenuation of gamma-rays. There are two approaches for activity measurements: absolute and relative method. In the relative approach, self-attenuation correction of gamma-rays can be avoided if sample and calibration standard both have similar matrix and are counted under identical geometries. In practical situation making similar geometry is easy, however finding standard having similar composition is difficult. This difficulty is caused by the variation in elemental compositions and densities of sample and standard. Nevertheless, these differences will require the self-attenuation coefficients to correct the final activities. If this correction is not applied, then at low energy it may lead to an error of more than ten percent [1].

Correction for self-attenuation requires mass attenuation coefficient values at different energies. Mass attenuation coefficient is a measure of the probability of interaction that occurs between incident photons and matter of unit mass per unit area. In gamma-ray spectrometry, the most common applications span 50 keV to 2000 keV energy range. Gamma rays are attenuated while passing through an absorber. The degree of attenuation depends on the scattering and various other absorption processes, which include photoelectric absorption, Compton scattering and pair production. In photoelectric absorption, a photon is completely absorbed and transfers its energy to an electron. The probability of photoelectric absorption is high at lower energies. In Compton scattering, the photon transfers a part of its energy to the electron termed as recoil electron and the photon itself is scattered through a certain angle. Compton scattering is important in the intermediate energy range. In pair production, the gamma-ray photon disappears and is replaced by an electron-positron pair. The probability of this type of interaction is relatively higher at high energy [2]. In samples having matrices with low atomic mass (Z) such as soil, vegetation and water, the major modes of interaction of gamma rays are Compton scattering and photoelectric absorption. The knowledge of the mass attenuation coefficients of photons in biological and other materials such as soils or alloys is of significant interest for industrial, biological, agricultural and medical applications. Mass attenuation coefficients are determined either experimentally or by mathematical modeling of the physical interactions of gamma-rays with a material.

To obtain self-attenuation correction factors, various techniques such as experimental [3], theoretical [4-6], semi-empirical [7-10] and Monte Carlo [11] have been used by different workers. Experimentally, the attenuation coefficients are determined by using the ratio of transmitted and incident intensities of a well-collimated beam of gamma-rays. Otherwise, if the chemical composition of a sample is known, the attenuation coefficients can be calculated using elementary data. One such provision of elementary data is

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XCOM [12, 13]. It was created by National Institute of Standards and Technology (NIST). XCOM can compute the mass attenuation coefficients or photon interaction cross-sections for many elements, compounds or mixtures in the energy range 1 keV – 100 GeV. The newest version of XCOM is known as WinXCom [14] and operates under the Windows operating system. It has a graphical user interface and provides output in the text form [15].

Radionuclides are used in many scientific studies such as ^{210}Pb is used for determining the sedimentation rate in lakes, estuaries and coastal marine environment. Similarly, ^{234}Th is employed as a natural tracer in the study of sediment chronology [16]. Gamma spectrometry laboratory at PINSTECH has long been involved in the determination of gamma-rays in environmental samples. These samples included soils [17], sediments [18], biological materials [19] and water samples [20]. The current study presents the transmission and computational methods for the determination of mass-attenuation coefficients for four reference soils, commonly used as calibration standards in gamma-ray spectrometry.

1.1 Methodology

In gamma-ray spectrometry, activity (A) of a radionuclide in a sample is calculated as:

$$A = \frac{N}{l\,\varepsilon\,I_\gamma} C_1 C_2 \ldots C_i \quad (1)$$

where N is the net count rate at energy E, l is the counting time, ε is the detector efficiency at the E, I_γ is the gamma-ray emission probability and $C_i s$ are the correction factors for self-attenuation, decay time, and other corrections.

1.2 Transmission Method

If an incident gamma-ray of energy E has intensity I_0 , then after passing through a medium of thickness t its intensity will become I. The mathematical relation holding for incident and transmission rays is Beer-Lambert's law, which is written as:

$$I = I_0\, e^{-\mu t} \quad (2)$$

The transmission of gamma photon is dependent on the thickness t of the medium and linear attenuation coefficient μ at energy E. Linear attenuation coefficient is function of density of the medium, atomic number of the constituent elements and energy of the incident beam. This relationship can only be used for good geometry conditions which means a thin absorber and well collimated beam of gamma-rays [2]. Eq. (2) can be modified in terms of mass attenuation coefficient, which is independent of density. The relation is given as:

$$\mu_m = \frac{\mu}{\rho} = \frac{1}{\rho t} \ln\left(\frac{I_0}{I}\right) \quad (3)$$

where $\mu_m$is the mass attenuation coefficient and ρ is the density of the material. The products ρt has units of (g cm^{-2}). For cylindrical sources, ρt can be calculated with the mass of the source divided by the area of the base of the source. For sample having many elements, its mass attenuation coefficient is calculated by the mixture rule:

$$\mu_m = \sum w_i\, (\mu_m)_i \quad (4)$$

where w_i is the weight fraction of the ith element [2].

Once the linear attenuation coefficient or mass attenuation coefficient is known. The measured count rate of a radioactive sample for a cylindrical geometry is given as:

$$I = I_0(1 - e^{-\mu t})/\mu t \quad (5)$$

Finally, the attenuation correction factor becomes:

$$c_{att} = (1 - e^{-\mu t})/\mu t \quad (6)$$

The correction factor for self-attenuation in relative quantification is:

$$c = \frac{(\mu t/(1-e^{-\mu t}))_{sam}}{(\mu t/(1-e^{-\mu t}))_{std}} \quad (7)$$

For many matrices, one could estimate the correction factor using a similar material.

1.3 WinXCom

If the composition of a sample is reasonably well established, another option is to estimate the mass attenuation coefficient (μ/ρ) for the composite material using the component parts $(\mu/\rho)_i$. This approach is useful but seems unwieldy when every sample is of different composition and useless when the sample composition is unknown. The maximum number of elements, WinXCom can handle is less than or equal to 28.

2. Experimental

2.1 Samples

This study included three IAEA reference soils that included IAEA-447 (moss-soil), IAEA-TEL-2011-03 (soil) and IAEA-TEL-2012-03 (soil) and one intercomparison soil prepared at PINSTECH (IC-2018-01). All IAEA soils were once distributed in different proficiency test exercises [21] by the IAEA. Some of these soils are now available as reference materials.

2.2 Experimental Setup

In the transmission method, ^{152}Eu point calibration source was used, which covered 121–1408 keV energy range. All measurements were made through a high resolution 20% n-type HPGe coaxial detector with full width at half maximum value of 2.0 keV at 1332 keV. The detector was shielded by circular lead bricks. For all measurements, the dead time was kept below 5% and source to detector distance was more than 10 cm. Soil samples were packed in cylindrical polyethylene containers of size 58 mm diameter and 69 mm height. All samples were packed in such a way to get homogenous density and uniform thickness. Lead collimator was used to make parallel beam of gamma-rays.

3. Results and Discussion

The study was performed in two steps: in the first step, methods were validated by using three metal foils (Goodfellow, UK): tin 5 mm, iron 1.35 mm and copper 2.5 mm. All foils were of 99.99 % purity. Density of the tin foil was 7.265 g cm^{-3}, iron foil was 7.874 g cm^{-3} and copper foil was 8.96 g cm^{-3}. In transmission method, peak areas at different gamma-ray energies of ^{152}Eu were measured with and without metal foils and mass attenuation coefficients (cm^2 g^{-1}) were calculated using Eq. (3). WinXCom was then employed and mass attenuation coefficients were determined for the same three foils. The comparison of mass attenuation coefficients by transmission method and WinXCom is presented in Fig. 1. The coefficients by the two methods were used and self-attenuation correction factors as given in Eq. (7) were calculated. The difference between the two methods in the form of percent root mean square error was 3%, 1% and 3% for tin, iron and copper, respectively. It seems a reasonable agreement of the measured and theoretical values. Error introduced in the measurements involved the counting statistics, non-uniformity of the foils, impurities in the metals and scattered photons.

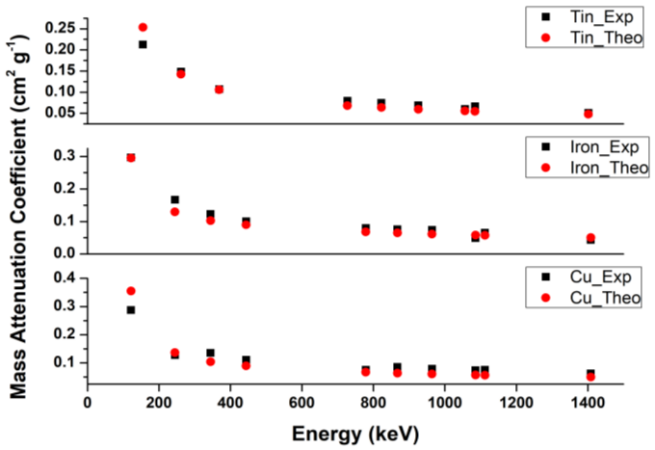

Fig. 1: Experimental and theoretical (WinXCom) mass attenuation coefficients for tin, iron and copper foils.

After validating the method as given above, the mass attenuation coefficients and attenuation correction factors were determined for three different IAEA reference soils by the transmission method. Since the elemental compositions of the three soils were not available, therefore the calculation of their mass attenuation coefficients by WinXCom was not possible. However, to give an idea about the mass attenuation coefficients for soils, WinXCom was applied for the world average soil using the composition given by Mason and Moore [22]. The elemental concentrations for the world average soil for 28 elements are presented in Table 1. This data include only those elements having concentration more than or equal to 0.003%. The results of the measured and calculated coefficients are presented in Fig. 2. It can be seen that the values measured for the IAEA soils and the world average soil are not very different. However, when mass attenuation coefficients were calculated for IC-2018-01 (soil)

Table 1: Elemental composition of average earth crust composition (Mason data) [22] and IC-2018-01 soil [24].

Element	Weight fraction	
	World average soil [21]	Soil IC-2018-01
O	0.466	-
Si	0.2772	0.27
Al	0.0813	0.0376
Fe	0.05	0.0332
Ca	0.0363	0.022
Na	0.0283	0.00218
K	0.0259	0.0137
Mg	0.0209	0.0123
Ti	0.0044	0.0031
H	0.0014	-
P	0.00105	0.00076
Mn	0.00095	0.00073
F	0.000625	-
Ba	0.000425	0.037
Sr	0.000375	0.012
S	0.00026	0.0011
C	0.000200	-
Zr	0.000165	-
V	0.000135	0.0111
Cl	0.000130	-
Cr	0.000100	0.0054
Rb	0.000090	0.00724
Ni	0.000075	0.002
Zn	0.000070	0.00837
Ce	0.000060	0.0065
Cu	0.000055	0.0040
Y	0.000033	-
La	0.000030	0.0023

by employing WinXCom and the concentration data given in Table 1, it showed a marked difference with the world average soil in the 40-200 keV region. This difference becomes insignificant after 200 keV and negligible after 500 keV as shown in Fig. 3. It appears that the Compton scattering dominates the absorption of gamma-rays over much of the 40-3000 keV range and is independent of atomic number. This means that, even if the composition of the sample is only approximately known, a reasonable self-absorption correction can be made with the major elements. However, this comfortable situation breaks down at low energy, where self-absorption is particularly severe, because of the strong dependence of photoelectric absorption on atomic number.

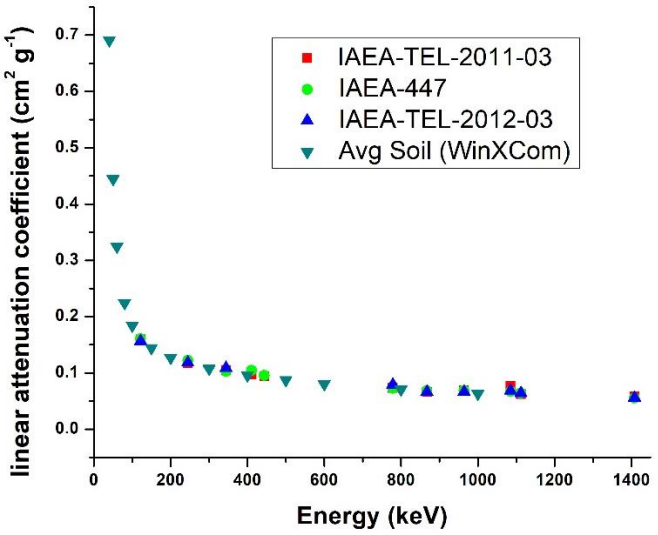

Fig. 2: Experimental and theoretical (WinXCom) mass attenuation coefficients of three IAEA soils and world average soil.

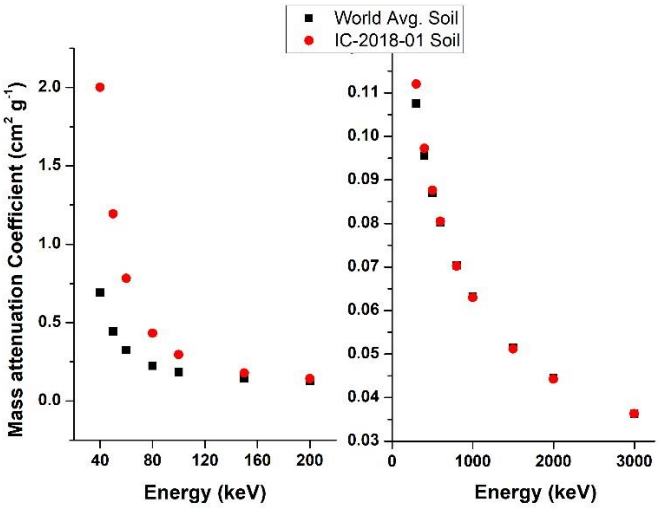

Fig. 3: Comparison of mass attenuation coefficients calculated by WinXCom for the world average soil and IC-2018-01 soil. Left Plot shows the difference of mass attenuation coefficients in the low energy range (40-200).

Exponential functions were fitted to the mass attenuation profiles of world average soil and IC-2018-01 soil in two energy ranges separately. These functions are presented as:

World average soil (40 - 200 keV): $\mu_m = 23.607E^{-1.02}$

IC-2018-01 soil (40 - 200 keV): $\mu_m = 737.93E^{-1.656}$

World average soil (200 - 3000 keV): $\mu_m = 1.5406E^{-0.465}$

IC-2018-01 soil (200 - 3000 keV): $\mu_m = 1.929E^{-0.465}$

It can be seen that the fitted functions are similar in 200 - 3000 keV for most type of the soils but different below 200 keV.

In soil samples, usually the concentration activities of ^{226}Ra, ^{232}Th and ^{40}K are measured [23]. The ^{226}Ra is estimated from γ-lines of ^{214}Bi (609.3 and 1764.5 keV) and ^{214}Pb (295.2 and 351.9 keV). The activity of ^{232}Th is estimated from γ-line of ^{212}Pb at 238.6 keV, of ^{208}Tl at 583.2 keV and of ^{228}Ac at 911.2 keV. The activities of ^{40}K and ^{137}Cs are measured using γ-lines at 1460.8 and 661.62 keV, respectively. Since, the difference between the calculated and the experimental mass attenuation coefficients after 200 keV is low; therefore, the values calculated for world average soil can be used for soils not mentioned here without risking too much accuracy. The mass attenuation coefficients at energies used in the determination of ^{226}Ra, ^{232}Th and ^{40}K and for the world average soil are presented in Table 2. If the chemical composition and density of an unknown soil sample and the calibration soil are similar then its attenuation correction factor will be approximately equal to one. However, if densities of both soils are different then the correction for self-attenuation has to apply.

Table 2: Mass attenuation coefficients calculated by WinXCom using world average soil data for radionuclides commonly observed.

Energy (keV)	Radionuclide	Parent radionuclide	mass attenuation coefficient (cm^2 g^{-1})
238.6	^{212}Pb	^{232}Th	0.1208
295.2	^{214}Pb	^{226}Ra	0.1094
351.9	^{214}Pb	^{226}Ra	0.1008
583.2	^{208}Tl	^{232}Th	0.0797
609.3	^{214}Bi	^{226}Ra	0.0781
661.7	^{137}Cs	^{137}Cs	0.0752
911.2	^{228}Ac	^{232}Th	0.0648
1460.8	^{40}K	^{40}K	0.0520
1764.5	^{214}Bi	^{226}Ra	0.0476

4. Conclusions

This study presentes the mass attenuation coefficients for three IAEA intercomparison soils determined by the transmission method. The results were compared with the mass attenuation coefficents estimated by WinXCom for the world average soil and IC-2018-01 soil. The data were fitted with the exponential fuctions and it has been shown that the self-attenuation correction factors can be estimated using the world average soil composition. This study also presents the mass attenuation coefficients required in the energy range 238.6 – 1764.5 keV for the determination of ^{212}Pb, ^{214}Pb, ^{208}Tl, ^{214}Bi, ^{137}Cs, ^{228}Ac, ^{40}K and ^{214}Bi in soil samples.

References

[1] K. Satoh, N. Ohashi, H. Higuchi and M. Noguchi, "Determination of attenuation coefficient for self-absorption correction in routine gamma ray spectrometry of environmental bulk sample", J. Radioanal. Nucl. Chem., Articles, vol. 84, pp. 431-440, 1984.

[2] G.F. Knoll, "Radiation Detection and Measurement (4th Edition)", John Wiley & Sons, Inc., New York, USA, 2010.

[3] N.H. Cutshall, I.L. Larsen and C.R. Olsen, "Direct analysis of ^{210}Pb in sediment samples: self-absorption corrections", Nucl. Instrum. Methods, vol. 206, pp. 309-312, 1983.

[4] M.S. Badawi, M.M. Gouda, S.S. Nafee, A.M. El-Khatib and E.A. El-Mallah, "New analytical approach to calibrate the co-axial HPGe

detectors including correction for source matrix self-attenuation", Appl. Radiat. Isot., vol. 70, pp. 2661-2668, 2012.

[5] M.I. Abbas, Y.S. Selim and M. Bassiouni, "HPGe detector photopeak efficiency calculation including self-absorption and coincidence corrections for cylindrical sources using compact analytical expressions", Rad. Phys. Chem., vol. 61, pp. 429-431, 2001.

[6] A.A. El-Sayed, "Evaluation of Compton scattering and self-attenuation coefficient after γ-ray analysis of naturally occurring radioactive elements in environmental samples", J. Radioanal. Nucl. Chem., vol. 274, pp. 379-387, 2007.

[7] J.P. Bolivar, "On self-attenuation corrections in gamma-ray spectrometry", Appl. Radiat. Isot., vol. 48, pp. 1125-1126, 1997.

[8] M.S. Al-Masri, M. Hasan, A. Al-Hamwi, Y. Amin and A.W. Doubal, "Mass attenuation coefficients of soil and sediment samples using gamma energies from 46.5 to 1332 keV", J. Environ. Radioact., vol. 116, pp. 28-33, 2013.

[9] K. Haddad and H. Suman, "Determination of the gamma self-attenuation correction factors using intensity ratios", J. Radioanal. Nucl. Chem., vol. 268, pp. 109-112, 2006.

[10] F. Hernández and F. El-Daoushy, "Semi-empirical method for self-absorption correction of photons with energies as low as 10 keV in environmental samples", Nucl. Instrum. Methods Phys. Res. A, vol. 484, pp. 625-641, 2002.

[11] J. Saegusa, K. Kawasaki, A. Mihara, M. Ito and M. Yoshida, "Determination of detection efficiency curves of HPGe detectors on radioactivity measurement of volume samples", Appl. Radiat. Isot., vol. 61, pp. 1383-1390, 2004.

[12] J.H. Hubbell, "Photon mass attenuation and energy-absorption coefficients from 1 keV to 20 MeV", Appl. Radiat. Isot., vol. 33, pp. 1269 - 1290, 1982.

[13] M.J. Berger, J.H. Hubbell, S.M. Seltzer, J. Chang, J.S. Coursey, R. Sukumar, D.S. Zucker and K. Olsen, XCOM: Photon Cross Section Database (online: http://physics.nist.gov/xcom National Institute of Standards and Technology, Gaithersburg, MD, USA, 2010.

[14] L. Gerward, N. Guilbert, K.B. Jensen and H. Levring, "WinXCom - A program for calculating X-ray attenuation coefficients", Rad. Phys. Chem., vol. 71, pp. 653-654, 2004.

[15] NIST, XCOM (online): https://physics.nist.gov/PhysRefData/Xcom/html/xcom1.html, 2020.

[16] C.C. Fuller, A. van Geen, M. Baskaran and R. Anima, "Sediment chronology in San Francisco Bay, California, defined by ^{210}Pb, ^{234}Th, ^{137}Cs, and ^{239,240}Pu", Mar. Chem., vol. 64, pp. 7-27, 1999.

[17] M. Ali, M. Wasim, M. Arif, J.H. Zaidi, Y. Anwar and F. Saif, "Determination of the natural and anthropogenic radioactivity in the soil of Gilgit - a town in the foothills of Hindukush range", Health Phys., vol. 98 (Supplement 2), pp. S69-S75, 2010.

[18] M. Wasim, S. Iqbal, M. Arif and M. Ali, "Determination of elements in Hunza River sediment by k 0 instrumental neutron activation analysis", J. Radioanal. Nucl. Chem., vol. 298, pp. 563-570, 2013.

[19] S. Iqbal, M. Wasim, M. Arif and Y. Anwar, "Methodology development for the determination of gamma emitting radionuclides in moss-soil (IAEA-447) and statistical evaluation of the proficiency test results submitted to the IAEA", Nucleus, vol. 48, pp. 231-236, 2011.

[20] I. Fatima, J.H. Zaidi and M. Arif, "Measurement of natural radioactivity in bottled drinking water in Pakistan and consequent dose estimates", Radiat. Prot. Dosimetry, vol. 123, pp. 234-240, 2007.

[21] IAEA, "Analytical Quality in Nuclear Applications, Worldwide Open Proficiency Test: Determination of Natural and Artificial Radionuclides in Moss-Soil and Water IAEA-CU-2009-03", IAEA Analytical Quality in Nuclear Applications Series No. IAEA/AQ/22, IAEA, Vienna, 2012.

[22] B. Mason and C.B. Moore, "Principles of Geochemistry", John Wiley & Sons, New York, 1982.

[23] M. Wasim, M. Ali and S. Iqbal, "Assessment of the risk associated with the gamma-emitting radionuclides from the soil of two cities in Central Karakorum", J. Radioanal. Nucl. Chem., vol. 303, pp. 985-991, 2015.

[24] M. Wasim, "Intercomparison Exercises: Element in soil (IC-2018-01EL), Radionuclides in soil (IC-2018-01RN) and Arsenic in water (IC-2018-02As)", Report No. FOA/Doc-01, Forum of Analysts, Islamabad, 2019.