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A New Approach for Determination of Volumetric Water Content in Soil Samples by Gamma-Ray Transmission

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Abstract A new method was presented for the purpose of volumetric water content determination in any soil sample by gamma-ray transmission. Monte Carlo (MC) simulation technique was used to determine the functional behavior of the linear attenuation coefficient of soil samples having different water contents for three soil samples. Using this functional behavior, the linear attenuation coefficients of dry soil and water were obtained from the intercept and the slope, respectively. It was experimentally shown that the mass attenuation coefficients of soil samples were not sensitive to the chemical composition but only to the physical density. This independence was exploited in this study to obtain the linear attenuation coefficient of a completely dry soil which was found to be 0.1409, 0.1274, and 0.1657 cm^{-1} for Gumushane, Ardahan, and Trabzon soil, respectively. The linear attenuation coefficient of water was determined to be 0.09 cm^{-1}. Then, the volumetric water contents were obtained by measuring the gamma-ray intensities passed through three wet soil samples. The results were found to be 0.186, 0.182, and 0.214 $cm^3\ cm^{-3}$ for Gumushane, Ardahan, and Trabzon soil, respectively. The results obtained by the method introduced were compared with the results obtained using gravimetric method. A very good agreement was observed.

Keywords Mass attenuation coefficient · Soil-water content · Monte Carlo simulation

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1 Introduction

Industrial activities, agricultural chemicals, and improper disposal of wastes are among the factors which are responsible for soil and ground water pollution. These chemical pollutants may possess serious health risks for man and other living organisms. Therefore, the behavior and the fate of these pollutants are of interest and tried to be understood for the last tens of years. Among other factors, soil-water content has been shown to play an important role on distribution, transport, and dissipation of these pollutants (Koo et al. 1990; Cho et al. 2005; Passeport et al. 2011; Vallee et al. 2016). Therefore, a number of methods have been introduced in order to determine soil-water content. The most popular ones are gravimetric, electromagnetic, tensiometric, hydrometric, and nuclear methods. A review article (Schmugge et al. 1979) explains in detail these techniques. In nuclear techniques, generally two types of radiation are used for water content determination: (*i*) particle radiation (neutrons) and (*ii*) electromagnetic radiation (gamma-rays or X-rays). Recently, fast neutrons produced by cosmic rays in the upper atmosphere are utilized to determine water content in soil (Hawdon et al. 2013). However, this method is restricted to be used in areas with a high altitude and the areas of low water content (Bogena et al. 2013).

Being an electromagnetic radiation type, gamma-ray transmission has found increasing applications in the soil density measurements and soil-water system in general (Cesareo et al. 1994; Pires et al. 2003; Appoloni and Pottker 2004). A number of authors reported the determination of volumetric water content in soil samples with the gamma-ray transmission method (Baytaş and Akbal 2002; Demir et al. 2008; Ün et al. 2011). Time-domain reflectometry (TDR) was used to determine unfrozen and ice content in a frozen soil with gamma attenuation (Zhou et al. 2014).

In all these methods involving gamma-ray transmission, volumetric water content cannot be determined unless the soil sample is dried. However, drying is a time consuming and grueling process. An alternative way of being able to determine the volumetric water content without drying process would be appreciated. If the functional behavior of the linear attenuation coefficient of a soil-water system is known, the volumetric water content can be determined without drying. This could be experimentally achieved by introducing a known amount of water into the system step by step. However, as the homogeneous distribution of water into the soil after adding water (unlike real soil samples) cannot be ensured, this may cause systematic errors which may deteriorate the quality of data. Monte Carlo simulations could be a great help for this purpose. In a previous publication (Celik and Cevik 2010), the water concentration effect in a soil sample on full energy peak efficiency in gamma-ray spectrometry was determined. In this process, a known amount of water was added into a soil sample step by step in order to determine the functional behavior of the full energy peak (FEP) efficiency. The same procedure can be used to determine the linear attenuation coefficient of a dry soil.

The main objective of this study is to present a new methodology to determine soil-water content in three soil samples having different physical properties. The methodology exploits the independence of linear attenuation coefficients of soil samples on chemical composition at energies higher than 100 keV which enables us to be able to determine the soil-water content without the drying process. The introduced method is believed to improve the analyses of soil-water content.

2 Theory

The attenuation of gamma-rays in a medium is expressed by

$$I = I_0 \exp(-\mu x) \tag{1}$$

where I_0 is the initial intensity of gamma-rays and I is the intensity after attenuation through a media of length x in cm, μ (cm^{-1}) is the linear attenuation coefficient of the material. If the quantity μ is divided by the density ρ, the mass attenuation coefficient, μ/ρ (cm^2 g^{-1}), is obtained. When a completely dry soil is considered, Eq. (1) is written as

$$I_{\mathrm{ds}} = I_0 \exp(-\mu_{\mathrm{ds}} x) \tag{2}$$

where I_{ds} is the intensity of gamma-ray passed through a completely dry soil and x is the thickness of the dry soil. When a soil having a certain amount of water is considered on the other hand, the following can be written:

$$I_{\mathrm{sw}} = I_0 \exp[-(\mu_{\mathrm{ds}} x + \mu_{\mathrm{w}} x_{\mathrm{w}})] \tag{3}$$

where x_{w} is the effective thickness of water which is spread in the soil system of thickness x. If the volumetric water content $\left(\theta_{\mathrm{w}} = \left(\frac{x_{\mathrm{w}}}{x}\right)\right)$ is inserted in Eq. (3), the following relation is obtained.

$$I_{\mathrm{sw}} = I_0 \exp[-x(\mu_{\mathrm{ds}} + \theta_{\mathrm{w}} \mu_{\mathrm{w}})] \tag{4}$$

Combining Eq. (2) and Eq. (4) yields the volumetric water content (cm^3 cm^{-3}) in a wet soil as in the following.

$$\theta_{\mathrm{w}} = \frac{1}{x \mu_{\mathrm{w}}} \ln\left(\frac{I_{\mathrm{ds}}}{I_{\mathrm{sw}}}\right) \tag{5}$$

In Eq. (5), volumetric water content cannot be determined unless knowing I_{ds} which is the gamma-ray intensity passing through a completely dry soil. In the current paper, however, we introduced a method by which one does not have to dry the soil sample in order to determine the volumetric water content. The method used in the current study is presented by starting the rearrangement of Eq. (4) as in the following.

$$\frac{1}{x} \ln\left(\frac{I_0}{I_{\mathrm{sw}}}\right) = \mu_{\mathrm{ds}} + \theta_{\mathrm{w}} \mu_{\mathrm{w}} \tag{6}$$

Solving Eq. (6) for volumetric water content gives the following relation.

$$\theta_{\mathrm{w}} = \frac{1}{\mu_{\mathrm{w}}} \left[\frac{1}{x} \ln\left(\frac{I_0}{I_{\mathrm{sw}}}\right) - \mu_{\mathrm{ds}}\right] \tag{7}$$

In this relation μ_{ds} and μ_w are linear attenuation coefficients of dry soil and water which were determined with Monte Carlo simulation. For the same photon energy, sample thickness and chemical composition, μ_{ds} and μ_w is constant and hence $\frac{1}{x}\ln\left(\frac{I_0}{I_{sw}}\right)$ can be considered as a function of θ_w in Eq. (6). This functional behavior can be determined by adding the known amount of θ_w to the soil sample and calculating incident (I_0) and attenuated (I_{sw}) photon intensities in each case with Monte Carlo method. When a known amount of water is added into the soil sample, its density as well as chemical composition is changed. This change was implemented in the code and taken into account in simulations. Once μ_{ds} is obtained from the fitting function, one can deduce the volumetric water content (θ_w) using Eq. (7). Once the volumetric water content is obtained, one can calculate the mass of the water in the soil sample using the relation $m_w = V\theta\rho$; here, V is the volume of the wet soil sample; when multiplied by θ, the volume of the water spread into the soil sample is obtained and ρ is the density of the water which is taken to be 1 g cm^{-3}.

2.1 Soil Samples

The soil samples under investigation were collected from undisturbed areas of the Gumushane, Ardahan, and Trabzon provinces in Turkey. The coordinates of the collected samples were given as 40° 25′ N, 39° 29′ E, 1655 m above sea level; 41° 05′ N, 42° 41′ E, 1892 m above sea level; and 40° 56′ N, 39° 41′ E, 407 m above sea level, respectively. The chemical analysis of the soil samples were performed via a 3D optics Epsilon 5 EDXRF Spectrometer at the laboratory of Recep Tayyip Erdogan University in Rize, Turkey, and the results are presented in Table 1. The type of the soil samples and their particle size distribution are presented in Table 2. The samples were selected in such a way that they differ from each other as much as possible with respect to particle size and type.

2.2 Monte Carlo Simulation Software

In order to calculate I_0 and I_{sw}, a detector model has to be constructed. The parameters of the detector are given in Table 3. Detector-sample geometry is given in Fig. 1 which contains the sample and its container, the HPGe detector, Pb collimators, point radioactive source, and lead shield. The diameters of the collimators are 4 mm which provide a narrow enough photon beam.

Table 1 Elemental chemical compositions (in % of mass) of soil samples

Element	Gumushane	Trabzon	Ardahan
Si	17.893	19.352	24.974
O	20.449	22.116	28.542
Al	22.594	22.812	16.068
O	20.083	20.277	14.283
P	–	0.223	0.467
O	–	0.287	0.603
K	2.128	1.106	1.952
O	0.437	0.227	0.401
Ca	4.159	0.809	3.781
O	1.663	0.324	1.512
Ti	0.674	1.455	–
O	0.449	0.970	–
Fe	4.188	6.311	3.094
O	1.795	2.705	1.326
Mg	1.319	–	0.824
O	0.879	–	0.549

The actual detector parameters generally differ from the ones provided by the manufacturer (Hedman et al. 2015) due to limited accuracy of the mounting procedure which causes errors in the distance of crystal to window, misalignment of the axes of crystal and housing, and changes of the sizes of detector components after cooling to cryogenic temperatures. For these reasons, X-ray imaging was performed and allowed to obtain more accurate information on the actual dimensions and placement of the detector components (Fig. 2). No misalignment or tilting of the axes of the components was discovered which might frequently be the case for old detectors.

The simulation process was carried out with the package PRESTA version of EGS4 (Nelson and Hirayama 1985) that perform Monte Carlo simulation of electron-photon showers in arbitrary materials. For EGS4 code, a MORTRAN code implementing a cylindrical model, UCCYSL (Nelson and Hirayama 1985), was used. The process includes the random generation of emission points inside the source. RANLUX random number generator was used with EGS4 code as it was shown to produce relatively better distribution and longer sequence (Gasparro et al. 2008).

Table 2 Particle size distribution of soil types

Type	Particle size (μm)	Gumushane soil	Ardahan soil	Trabzon soil
Clay	<2	7.7	29.3	16.2
Loam	2–50	29.4	56.0	41.8
Sandy	>50	62.9	14.7	42.0

For the model executed with EGS4, the efficiency was divided into 10,010 energy bins, each one having a width of 0.3 keV. This number of bins was chosen in order to have a few bins above the highest energy used in order to check for rounding errors in the calculations.

The simulations do not take cosmic rays or background sources of radioactivity which contribute to peaks that occur in experimental data into account. In addition, the broadening due to electronic noise and the statistics of ion-pair production is not included.

Energies given in the model for simulation were chosen to be always 10 eV below an integer value in order to avoid having an energy lying just at the end of a bin as this gives rounding errors leading to a non-negligible number of events scored in the following channel.

2.3 Experimental Measurements

The experimental set-up is illustrated in Fig. 1. First, the empty container was placed between the radioactive point sources (^{133}Ba, ^{137}Cs, and ^{60}Co) and the detector in order to measure incident photon intensity (I_0). The distance between point sources and the samples was 80 mm. Then, the soil samples under investigation which have a distance of 100 mm to the detector were put in the bottle having 3.778 cm in diameter and placed the same way as for the empty bottle to count photon intensity (I_{sw}) passed through the soil-water media which has a thickness of 70 mm. The counting time for each soil sample was 86,400 s (1 day). Based on measured photon intensities (I_0 and I_{sw}) and using Eq. (6), mass attenuation coefficients of soil-water system were determined for eight photon energies (81, 276, 302, 356, 384, 662, 1173, and 1332 keV).

2.4 Determination of Volumetric Water Contents

Volumetric water contents were determined by using Eq. (7). The photon intensities I_0 and I_{sw} in Eq. (7) were determined experimentally using the experimental geometry given in Fig. 1 for the energy value of 662 keV emitted by ^{137}Cs radioactive point source since, at this energy level, I_{sw} does not depend on the chemical composition. Linear attenuation coefficients of dry soil (μ_{ds}) and water (μ_w) were determined via Monte Carlo simulation. Known amount of water were added to the soil samples, and the simulations were performed in each case in order to determine how $(1/x)\times\ln(I_0/I)$ changes with the water content added to the soil sample. When a known amount of water was added to a soil sample, the chemical composition and the physical density changes. These changes were considered in the code of the program. The simulation results were used to create Fig. 3. The intercept and slope of Fig. 3 give linear attenuation coefficient of completely dry soil and water, respectively. By using this method, one does not need to dry the soil sample in order to determine the linear attenuation coefficient.

3 Results

Experimentally determined mass attenuation coefficients of three soil types are presented in Table 4 with the uncertainties associated with them. The results presented in Table 4 show us that mass attenuation coefficients of soil samples seem to be independent from the chemical composition of the samples especially for the photon energies higher than 81 keV. This means that, as same for the mass attenuation coefficient, linear attenuation coefficients also is not sensitive to the chemical composition of the soil only to the physical density which is determined by dividing the mass by the volume of the bottle which the soil sample was put into. This can be

Table 3 Detector parameters used in simulations

Type	Crystal diameter	Crystal active volume	Crystal to endcap distance	Entrance window thickness	Ge front dead-layer thickness	Relative efficiency
p	7.5 cm	231.1 cm^3	0.4 cm	1 mm (Al)	700 μm	55 %

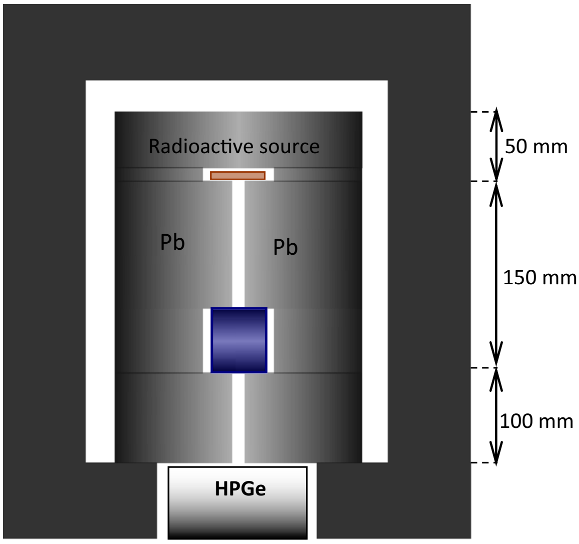

Fig. 1 Detector-sample geometry considered for both Monte Carlo simulations and experimental measurements

explained by the fact that the photoelectric process is the dominant mode of interaction for gamma-rays at relatively low energies (below 100 keV). And the photoelectric

Fig. 2 Radiograph of the detector crystal used in experimental measurements and Monte Carlo simulations

cross section below 100 keV is dominated by the atomic number of the absorbing material which results in dependence of linear attenuation coefficients on chemical composition as given in Eq. (8) (Knoll 2000).

$$\sigma_{\mathrm{ph}} = \mathrm{constant}\frac{Z^n}{E_\gamma^{3.5}} \tag{8}$$

Here, σ_{ph} is the photoelectric cross section, Z is the atomic number of absorbing material, and E_γ is the energy of gamma-ray. The power n varies between 4 and 5. On the other hand, as the energy of gamma-ray increases (above 100 keV), Compton interaction becomes the dominant process and its cross section is given with the Klein-Nishina formula (Knoll 2000) in which gamma-ray energy unlike atomic number of the absorbing materials is the most dominant parameter. Therefore, for the gamma-ray energies higher than 100 keV, linear attenuation coefficients of soil is not sensitive to the chemical composition but to the physical density. This independence at relatively higher energies was also shown earlier (Cesareo et al. 1994).

This independence of mass attenuation coefficient (and hence linear attenuation coefficient) on the chemical composition enables us to determine the linear attenuation coefficient of dry soil using MC simulation. Knowing the average chemical composition of any soil is enough for creating the MC code. The critical parameter here is the physical density of the soil which is easy to determine and makes this method useful and practical.

In order to determine the linear attenuation coefficient of dry soil with MC calculation, 662 keV photon energy emitted from ^{137}Cs radioactive point source was chosen. Linear attenuation coefficients of three soil samples as a function of added volumetric water content (θ_w) were calculated and presented in Fig. 3. Using the fitting function, the linear attenuation coefficients of water (μ_{w}) and dry soil (μ_{ds}) were obtained. As seen from Fig. 3, linear attenuation coefficients of water obtained from the slope for three soil types are in agreement with each other within the relative difference being less than 2 %. On the other hand, linear attenuation coefficients of dry soil obtained from intercepts for three soil types are different from each other indicating the different physical densities of soils.

Using Eq. (7), volumetric water contents were determined for Gumushane, Ardahan, and Trabzon soils. The results are presented in Table 5. As seen from Table 5, 0.186 ± 0.019, 0.182 ± 0.021, and 0.214 ± 0.020 cm^3 cm^{-3}

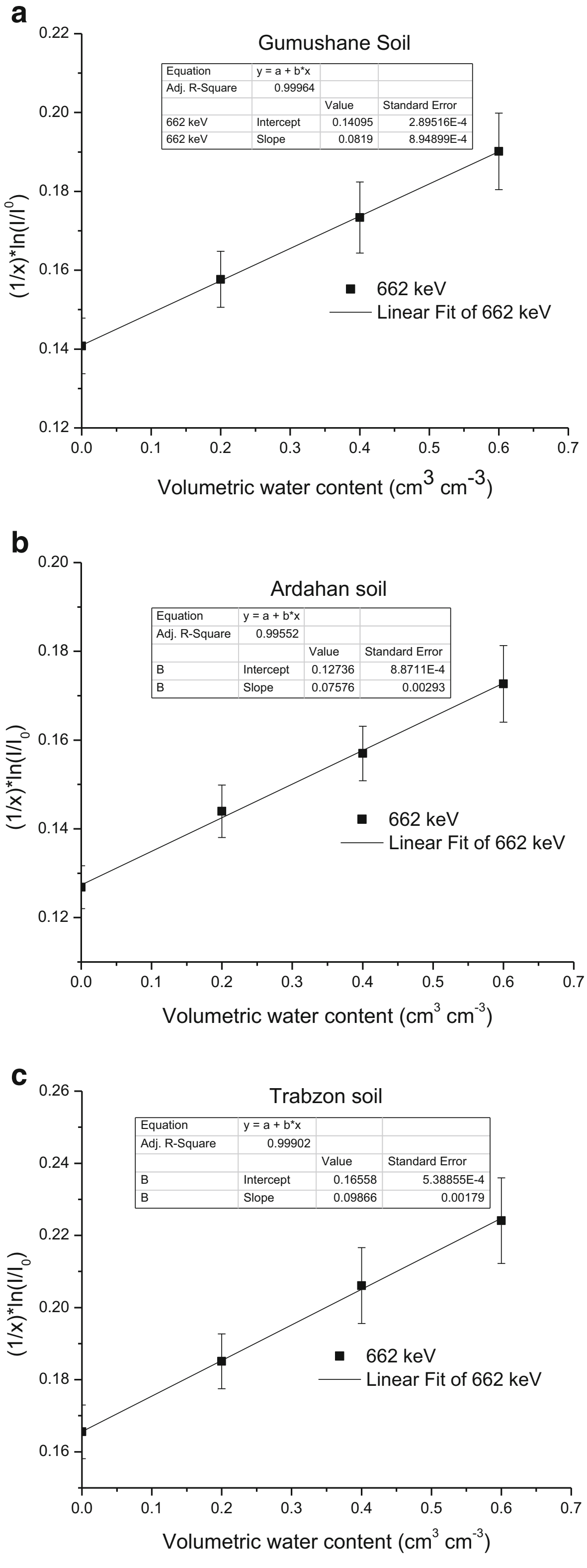

Fig. 3 **a**–**c** Linear attenuation coefficient of soil-water system as a function of volumetric water content

volumetric water contents were obtained for Gumushane, Ardahan, and Trabzon soils, respectively. The obtained volumetric water contents correspond to 22.33 ± 2.79, 21.92 ± 2.49, and 25.76 ± 3.09 g of water for Gumushane, Ardahan, and Trabzon soils, respectively.

3.1 Uncertainty Analysis

The sources of uncertainty associated with simulation results are those of statistical and the cross section used by the program. We chose a sufficient number of photons so that the statistical uncertainty associated to the calculated photon intensities was less than 1 % for the energy considered. It was sufficient to generate primary photons of the order of 10^7. The uncertainty associated to the approximations made in the cross-section data is generally accepted be to be around 2 %.

The biggest contribution to the uncertainties associated with experimentally measured mass attenuation coefficients came mainly from two sources: the uncertainty in counting rate $\left(\sqrt{N}/t\right)$ where t is counting time and N is the number of counts under the full energy peak (FEP) and the uncertainty in determination the thickness of soil samples. Among them, however, the biggest contributor to the uncertainty is the counting rate uncertainty.

The uncertainty analysis was performed by applying the standard error propagation equation, and the results were shown together with the data.

3.2 Verification of the Method

In order to test the validity of the method presented in the current study, we chose the gravimetric method to determine water mass for three soil types and compare the results obtained. After gamma-spectroscopic measurements, the soil samples were taken out of the bottle and weighed. The values of 243.09 g for Gumushane soil, 219.30 g for Ardahan soil, and 286.28 g for Trabzon soil values were obtained and recorded. Then, the soil samples were put in an oven at 105 °C for 1 week in order for them to dry. After 1 week, the soil samples were taken out of the oven and weighed. The masses of water for each soil type were obtained by subtracting the wet

Table 4 Experimentally determined mass attenuation coefficients of three soil types

Radioactive point source	E (keV)	μ/ρ (cm^2 g^{-1})		
		Gumushane	Ardahan	Trabzon
^{133}Ba	81	0.5566 ± 0.0228	0.5459 ± 0.0273	0.5637 ± 0.0311
	276	0.3107 ± 0.0112	0.3112 ± 0.0134	0.3097 ± 0.0149
	302	0.2997 ± 0.0141	0.2993 ± 0.0141	0.3001 ± 0.0141
	356	0.2821 ± 0.0107	0.2823 ± 0.0144	0.2822 ± 0.0138
	384	0.2742 ± 0.0123	0.2743 ± 0.0132	0.2742 ± 0.0115
^{137}Cs	662	0.2174 ± 0.0093	0.2184 ± 0.0093	0.2173 ± 0.0094
^{60}Co	1173	0.1647 ± 0.0064	0.1650 ± 0.0081	0.1647 ± 0.0086
	1332	0.1562 ± 0.0062	0.1562 ± 0.0075	0.1563 ± 0.0073

weight and dry weight of soil samples. The results are presented in Table 5 with the relative differences between the values obtained from current and gravimetric techniques. As seen in Table 5, the agreement between two techniques is about 1 % for Gumushane and Trabzon soil and around 5 % for Ardahan soil.

4 Discussions

Water contents for three different soil types were determined using a gamma-ray transmission experiment. This method is more practical than those of the conventional methods as to avoid the drying process. The result obtained via this technique was compared with the results obtained using gravimetric which takes the subtraction of the mass of dry soil from the wet soil. A good agreement was obtained as shown in Table 5.

One should keep in mind that in order to have reliable data the detector and the source should be collimated well so that the systematic errors due to the scattering can be avoided. The scattering occurs when a photon from the source is scattered by the absorber through a small angle into the detector. The detection of these scattered photons gives a systematic error to the transmission ratio $\ln(I_0/I_{sw})$ observed. For the Compton scattered photons, the change in the energy for small angles is too small for the detector to distinguish from the full energy peak. Elastic scattered photons have nearly identically the same energy as the primary ones. The relative magnitude of this effect was observed to be significant for large collimator diameters (Çelik et al. 2012) and for the detection systems having poor energy resolution as compared to HPGe detector. This effect can be diminished by making narrow beam geometry.

It is found out that Monte Carlo determination of functional behavior of soil-water system is not sensitive to the chemical composition of the soil samples but to the physical density which is easy to determine (mass/volume) and makes this method easy and practical as to avoid the drying process. By this technique that we introduced, one can determine the volumetric water

Table 5 Volumetric water contents and water masses for three soil types

	Gumushane soil	Ardahan soil	Trabzon soil
μ_{ds} (cm^{-1})	0.1409 ± 0.0121	0.1274 ± 0.0116	0.1657 ± 0.0134
μ_w (cm^{-1})	0.0891 ± 0.0068	0.0912 ± 0.0072	0.0907 ± 0.0083
θ_w (cm^3 cm^{-3})	0.186 ± 0.019	0.182 ± 0.021	0.214 ± 0.020
[a]m_w (g)	22.33 ± 2.79	21.92 ± 2.49	25.76 ± 3.09
[b]m_w (g)	22.10 ± 1.28	20.85 ± 1.42	26.03 ± 1.87
Rel diff. (%)	1.05	5.13	−1.04

[a] Water mass obtained from the technique introduced in the current study

[b] Water mass obtained from gravimetric technique

content without drying the soil sample which may take time otherwise. For that, the exact chemical composition of the soil samples need not to be known; only an average chemical composition which any soil sample may have is enough since we have experimentally shown that the mass attenuation coefficient at photon energies higher than 100 keV is not sensitive to the chemical composition. We have successfully validated the current method by comparing the results obtained from the gravimetric method which is probably the most widely used technique for water content determination in soil samples.

In the current study, we determined linear attenuation coefficients of dry soil and water using MC simulation. The reader should be aware that a software like XCOM (Berger et al. 2010) can also be used to achieve the same task.

It should be taken into account that for a good calculation of incident (I_0) gamma-rays and intensity of gamma-rays after passed through the sample medium (I_{sw}), the parameters of the detector (dead-layer thickness, active volume, etc.) should be very well determined. We took the radiograph of the detector as shown in Fig. 2 for correct detector parameters to be implemented in simulations. However, some parameters could not still be viewed by the radiograph like dead-layer thickness. The increase of the dead-layer thickness in time caused the decrease in crystal active volume. We changed the radius of the detector by 10 mm, as a result the incident photon intensity (I_0) at 662 keV changed by 43 %. However, the value $\ln(I_0/I_{sw})$ changed by only 2 % as numerator and denominator changes in the same way in Eq. (6) so that errors were compensated. We used dead-layer thickness tabulated by the manufacturer. As a result of our calculations, we can conclude that even the parameters given by the manufacturer are not the exact parameters as the real ones; our calculations are still valid.

5 Conclusions

The presented method is fast and easy to apply to obtain volumetric water content in any soil sample without drying process which might be grueling and time consuming in some cases especially if the water content is needed to be determined in a short period of time. Experimental measurements showed that the mass attenuation coefficients of soil do not depend on the chemical composition at energies higher than around a hundred kilo-electron volt since the interaction mechanism at this energy level is governed by Compton scattering. This independence of mass attenuation coefficient on chemical composition enabled us to determine the linear attenuation coefficient of completely dry soil samples without drying them by introducing the functional behavior a certain quantity with changing the water content in the sample. For that, the exact chemical composition of the soil sample need not to be known; only the average chemical composition that any soil sample may have is enough. When the gamma-ray intensity passing through the wet soil and the physical density in measured, the soil-water content can easily be determined.

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