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Mass attenuation coefficients of environmental samples for gamma-ray energies from 46.5 keV to 1408 keV

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▶ ***Original article***

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ABSTRACT

Background: In gamma-ray spectrometry for the radioactivity analysis of bulk samples, self-attenuation correction is necessary when the attenuation rates in test sample differ from the one in the efficiency calibration source. Thus, the mass attenuation coefficient of test samples is an important factor in gamma-ray spectrometry. This study estimates the mass attenuation coefficients for seven kinds of environmental samples. ***Materials and Methods:*** An uncollimated transmission system with a high purity germanium detector system was used to measure the gamma-ray transmission rates with and without test samples. The system was calibrated using C (activated charcoal), H_2O, MnO_2, NaCl, Na_2CO_3, and $(NH_4)_2SO_4$ as reference materials. Sea sediment, surface soil, fish, seaweed, Chinese cabbage, milk, and pine needles were selected, and ten identical samples for each sample type were tested using the system. ***Results:*** The calibration of the uncollimated transmission system was validated with good agreement within 4% between linear attenuation coefficients by experiment and calculation for K_2CrO_4 and SiO_2. The standard deviation of the mass attenuation coefficients for each kind of sample was estimated as less than 5% above around 100 keV. ***Conclusion:*** Mass attenuation coefficient does not significantly depend on the sample type for gamma-ray energy higher than around 100 keV, but mass attenuation coefficient for the lower gamma-ray energy should be considered even with similar kinds of sample. The mass attenuation coefficients tabulated in this paper can be used as a reference or comparable value in gamma-ray spectrometry for environmental samples.

Keywords: *Mass attenuation coefficient, gamma-ray spectrometry, self-attenuation correction.*

INTRODUCTION

Gamma-ray spectrometry has been widely used for environmental radioactivity analyses because it has the advantage of analyzing various gamma-ray emitting nuclides simultaneously with a relatively rapid and simple pretreatment compared to a radiochemical analysis. In general, detection efficiency calibrations of gamma-ray spectrometers are performed using standard mixed gamma-ray sources with a density of approximately 1 g cm^{-3}. Environmental samples can have a variety of chemical compositions and densities, and even the types of samples can differ. Therefore, the self-attenuation effects between the calibration sources and environmental samples may also be different, and detection efficient correction may be needed to prevent the radioactivity from being over

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estimated or under estimated. Various methods to obtain the self-attenuation correction factor (SCF) have been published and it can be obtained by equation (1):

$$\mathrm{SCF} = \frac{\varepsilon(\mu_{l\_s})}{\varepsilon(\mu_{l\_c})} \quad (1)$$

where ε is the detection efficiency and μ_{l_s} and μ_{l_c} are linear attenuation coefficients of the sample and calibration sources, respectively. The linear attenuation coefficient (μ_l) is calculated by multiplying the mass attenuation coefficient μ_m by the sample density. The μ_m of environmental samples can be determined using an analytical calculation with elementary data and a chemical composition analysis or using the transmission method with collimated or uncollimated transmission methods (1-3). In general, the density of the soil of environmental samples tends to be larger than the calibration standard gamma-ray source with a density of about 1 g cm^{-3}, and radioactivity analysis results without the self-attenuation correction may be underestimated due to $\mathcal{E}(\mu_{l\_s})<\mathcal{E}(\mu_{l\_c})$. For this reason, the μ_m for various soil and sediment samples has been studied (4-6), and it can be useful as a reference for self-attenuation correction in gamma-ray spectrometry. However, in addition to the soil, there are various type of samples including biological samples for environmental radioactivity monitoring. In contrast to soil samples, usual biological samples tend to have a lower density than standard sources, and the radioactivity analysis results can be overestimated due to $\mathcal{E}(\mu_{l\_s})>\mathcal{E}(\mu_{l\_c})$ if the self-attenuation effect is not corrected. There have been a few studies including μ_m for biological samples, for example, grass for self-attenuation consideration in gamma-ray spectrometry (7, 8). However, the μ_m of land or marine biological samples such as vegetable, milk, plant or fish used for environmental monitoring has not been comprehensively reported yet. Therefore, for more accurate gamma-ray spectrometry for environmental samples, μ_m for more various samples should be studied as well as soil samples.

The purpose of this study is to estimate the μ_m of major samples for environmental radioactivity monitoring and to establish a database for it. μ_m for seven types of samples in an energy range from 46.5 keV to 1408 keV was studied. This paper introduces the determination process of μ_m for environmental samples with unknown chemical composition and discusses characteristics of μ_m according to sample type.

MATERIALS AND METHODS

For this study, we used the CT method with an uncollimated beam to determine the μ_m for environmental samples, with C (activated charcoal), H_2O, MnO_2, NaCl, Na_2CO_3, and $(NH_4)_2SO_4$ as reference materials for the calibration of the transmission system (3). The processing steps of the CT method are summarized as follow:

1) Measurement system calibration

i) Preparing reference materials with known chemical composition that has the same geometry as a test sample including the sample bottle.

ii) Obtaining the photon transmission ratio (I_m/I_o) of full energy peak count rates with (I_m) and without (I_o) reference materials using gamma-ray sources with a target energy range. (The measurement system arrangement is shown in figure 1)

iii) Determining the regression equation (2) with fitting parameters a and b through linear fitting of the data sets of the measured I_m/I_o and calculated μ_l with the chemical composition of reference materials:

$$\mu_l = a + b \ln (I_m/I_o) \quad (2)$$

2) Determination of μ_m for test samples

i) Preparing test samples that have the same geometry as the calibration reference materials including the sample bottle.

ii) Obtaining I_m/I_o for the test samples in the same manner as for the system calibration.

iii) Calculating μ_l by substituting I_m/I_o into equation (1), and μ_m by dividing μ_l by the apparent density of the measured test sample.

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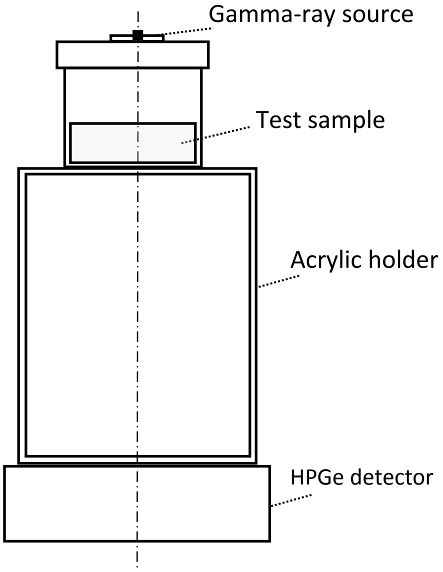

Figure 1. Experimental arrangement.

Each reference material was dried and sieved using standard 200 μm mesh nets for a homogeneous density distribution in a sample bottle and then filled to a height of 20 mm in a cylindrical acrylic container with a diameter of 60 mm and a height of 40 mm. The apparent densities of C, H_2O, MnO_2, NaCl, Na_2CO_3, and $(NH_4)_2SO_4$ were measured as $(0.637 \pm 6.72 \times 10^{-5})$ g cm^{-3}, $(1.00 \pm 2.96 \times 10^{-5})$ g cm^{-3}, $(2.77 \pm 1.10 \times 10^{-4})$ g cm^{-3}, $(1.34 \pm 1.08 \times 10^{-4})$ g cm^{-3}, $(1.34 \pm 8.66 \times 10^{-5})$ g cm^{-3}, and $(1.07 \pm 2.89 \times 10^{-6})$ g cm^{-3}, respectively.

To determine the mass attenuation coefficients in a broad energy range, we used the gamma-ray standard sources of ^{210}Pb (46.5 keV), ^{241}Am (59.5 keV), ^{152}Eu (121.8, 244.7, 344.3, 444, 778.9 and 1408 keV), ^{137}Cs (661.7 keV), and ^{60}Co (1173.2 and 1332.5 keV). The calibration sources were surrounded by an acrylic capsule with a height of 3.2 mm and a diameter of 24.5 mm. ^{210}Pb and other sources had radioactivity levels of approximately 37 kBq and 370 kBq, respectively. A Broad Energy Germanium (BEGe, Mirion Technologies Inc.) detector with a 60% relative efficiency was used to measure the transmitted gamma-rays. Each gamma-ray source was placed on the central axis of the front face of the detector end cap. Figure 1 shows a schematic drawing of the experimental arrangement. The distance from the center of the source to the external front face of the detector was set to 200 mm to ignore the true coincidence summing effect. In these measurement conditions, we calculated the summing effects for the calibration sources used in this study to be less than 1%. For each of the five standard gamma-ray sources, the count rates of the full energy peak with an empty bottle and the reference material were below 1% of the relative standard deviation for the energy of interest. The measured gamma-ray energy spectra was analyzed to obtain full energy peak counts using the Genie2K software (Mirion Technologies Inc.).

To validate the transmission system, we measured the μ_m values of K_2CrO_4 ($(1.92 \pm 9.92 \times 10^{-5})$ g cm^{-3}) and SiO_2 ($(1.47 \pm 6.37 \times 10^{-5})$ g cm^{-3}) and compared them to the reference values calculated by the XCOM (Photon Cross Sections Database) with a certified chemical composition (9). In this study, we collected from different areas in Korea seven kinds of environmental samples - sea sediment, surface soil, fish, seaweed, Chinese cabbage, milk, and pine needles - constituting the most common samples for environmental radioactivity monitoring (10, 11). Each sample was prepared via the general pretreatment methods used in an environmental radioactivity analysis. The sea sediment and surface soil samples were dried at 105 ˚C while the other samples were completely combusted after drying. All samples were then ground to less than 200 μm and filled in the bottles with the same geometric configuration as the calibration samples. Table 1 shows the sample types, the number of samples used, and the measurement conditions.

For each environmental sample, the count rates of the full energy peak were measured in the same manner as the reference materials for the calibration of the system and each sample was measured without a transmission source to measure the sample background due to the likely detectable anthropogenic gamma-rays from the sample. The μ_m was determined through a linear fit with the derived μ_l values for each sample by equation (3), as follows:

$$\mu_m(E) = (a + b\ln(R(E)))/\rho \qquad (3)$$

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Where R(E) is the gamma-ray transmission ratio (I_m/I_o) at energy (E), ρ is the apparent density of the measured sample. The SigmaPlot (Systat Software Inc.) software was used for statistical analysis and graphing.

Table 1. Information of the environmental samples used in this study.

Sample Material	Apparent density (g cm^{-3})	Number of Samples	Measurement Condition
Sea sediment	1.01 - 1.79	10	Dry
Surface soil	0.980 - 1.48	10	Dry
Fish	0.668 - 0.986	10	Ash
Seaweed	0.602 - 0.944	10	Ash
Chinese Cabbage	0.596 - 0.798	10	Ash
Milk	0.572 - 0.779	10	Ash
Pine needle	0.416 - 0.660	10	Ash

RESULTS

Table 2 shows the ratio (I_m/I_o) of the transmitted full energy peak count rates (I_m) with each calibration material and the full energy peak count rates (I_o) with a blank bottle for gamma-ray energies from 46.5 keV to 1408 keV. To obtain the regression equation with the fitting parameters a and b of equation (1), the μ_l values for C, H_2O, $(NH_4)_2SO_4$, NaCl, Na_2CO_3 and MnO_2 were plotted together and fitted by the linear least squares method, as shown in figure 2. The fitted parameters for μ_m in Eq. (2) were assigned as follows: a = (0.002008 ± 0.000495) cm^{-1} and b = (-0.4953 ± 0.0013) cm^{-1} with the coefficient of determination R^2 = 0.9995. For K_2CrO_4 and SiO_2 that were used to validate the calibrated system, the μ_m values according to an experiment and calculation using the XCOM were in good agreement within 4%, as shown in table 3.

Figure 3 shows the I_m/I_o ratios and μ_m values with apparent densities recorded experimentally at different energy levels for 70 samples; the μ_m for each sample was determined using equation (2). The data set of the gamma-ray energy and transmission ratio was fitted with a sixth- degree polynomial.

Table 2. The ratio (I_m/ I_0) of count rates with and without material for C, H_2O, $(NH_4)_2SO_4$, NaCl, Na_2CO_3 and MnO_2.

Energy (keV)	I_m/ I_0					
	C	H_2O	$(NH_4)_2SO_4$	NaCl	Na_2CO_3	MnO_2
46.5	0.789	0.652	0.520	0.242	0.577	0.0277
59.5	0.808	0.684	0.615	0.427	0.615	0.0199
121.8	0.840	0.730	0.721	0.668	0.688	0.345
244.7	0.872	0.780	0.777	0.753	0.747	0.534
344.3	0.885	0.800	0.795	0.769	0.770	0.581
444	0.892	0.816	0.817	0.795	0.785	0.619
661.7	0.905	0.838	0.841	0.817	0.820	0.664
778.9	0.917	0.851	0.853	0.832	0.829	0.683
1173.2	0.933	0.881	0.876	0.863	0.859	0.732
1332.5	0.934	0.885	0.883	0.868	0.866	0.745
1408	0.936	0.891	0.892	0.870	0.868	0.764

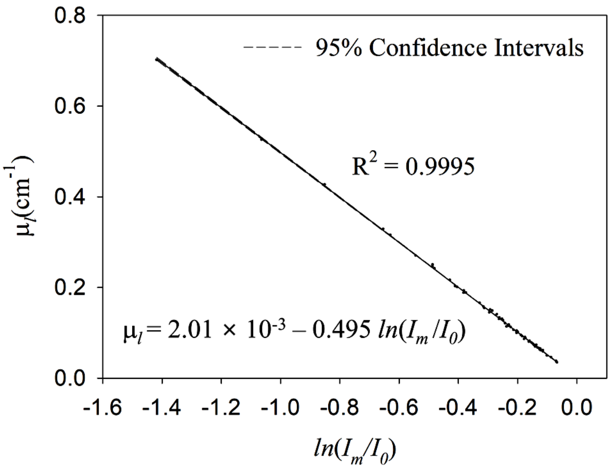

Figure 2. The linear attenuation coefficients (μ_l) for C, H_2O, $(NH_4)_2SO_4$, NaCl, Na_2CO_3 and MnO_2 versus the transmission ratio (I_m/I_0).

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Table 3. Comparison of μ_l by the CT method and the XCOM for K_2CrO_4 and SiO_2.

Energy (keV)	Nuclide	K_2CrO_4			SiO_2		
		CT method (cm^{-1})	XCOM calculation (cm^{-1})	Difference (%)	CT method (cm^{-1})	XCOM calculation (cm^{-1})	Difference (%)
46.5	^{210}Pb	1.76	1.77	-0.3	0.467	0.462	0.9
59.5	^{241}Am	0.961	0.974	-1.3	0.328	0.333	-1.6
121.8	^{152}Eu	0.330	0.330	0.1	0.217	0.214	1.4
244.7	^{152}Eu	0.212	0.219	-3.1	0.169	0.168	0.7
344.3	^{152}Eu	0.188	0.190	-1.2	0.149	0.148	0.4
444	^{152}Eu	0.170	0.171	-0.8	0.135	0.134	0.9
661.7	^{137}Cs	0.142	0.144	-1.4	0.116	0.113	2.5
778.9	^{152}Eu	0.132	0.133	-1.0	0.108	0.105	2.7
1173.2	^{60}Co	0.111	0.109	1.6	0.088	0.086	1.5
1332.5	^{60}Co	0.102	0.103	-0.9	0.083	0.081	2.0
1408	^{152}Eu	0.096	0.100	-3.6	0.081	0.079	2.8

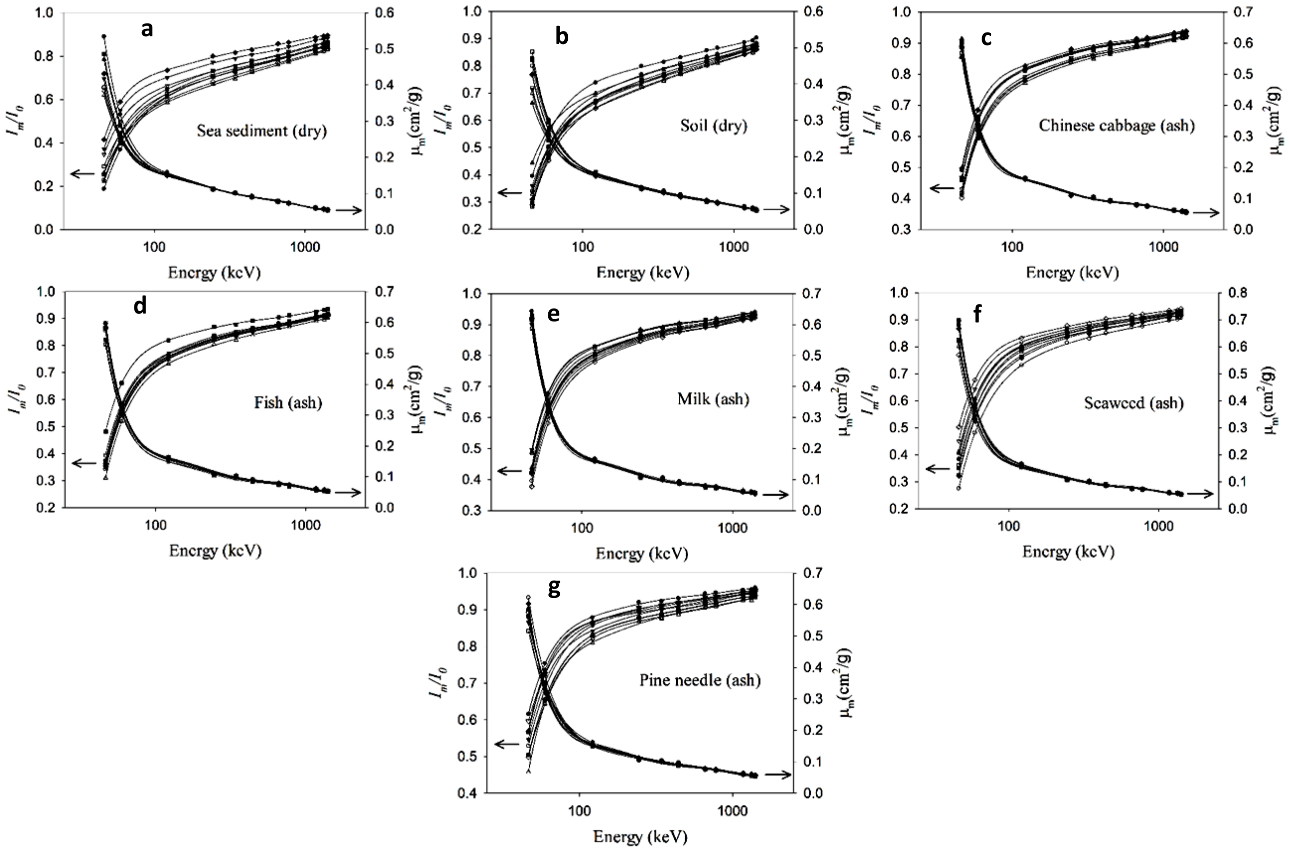

Figure 3. The mass attenuation coefficients (μ_m) and the ratio (I_m/I_0) versus the energy for seventy environmental samples; (a) sea sediment, (b) soil, (c) Chinese cabbage, (d) fish, (e) milk, (f) seaweed and (g) pine needle.

DISCUSSION

As shown in figure 4 and table 4, the standard deviation of μ_m for each sample matrix over most of the samples tends to decrease with increasing gamma-ray energy, especially for the dried soil samples compared to the combusted biological samples, due to differences in the chemical composition. This phenomenon occurs because low-energy gamma-rays are typically attenuated by photoelectric absorption in the medium of the interaction. In other words, since

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photoelectric absorption is proportional to $Z^{4\sim5}$ (12), the μ_m for low energy can vary significantly if the effective atomic numbers differ from one another, even if they are of the same type. In addition, in combusted biological samples, most of the organic matter is removed and the same kind of samples have a similar effective atomic number, so there may be less relative standard deviation than in soil containing various minerals. In this study, the μ_m for the sea sediment, surface soil, seaweed and pine needles was estimated with a relative standard deviation of 5% above around 100 keV, and the μ_m for the other samples tended to have lower relative standard deviations even in the low gamma-ray energy range. This is because Compton scattering with linearly increasing probability with Z is dominant in the energy region. This result is consistent with the ones reported by several researchers (4, 13-15). They concluded that the variations of μ_m for building materials, soils and biological materials in the medium energy region between 100 keV (or 300 keV) to 3 MeV are very small, through Monte Carlo simulation, calculation using the XCOM, or real measurements.

Soil is usually distinguished as minerals and organic constituents of variable depths which differ from the parent materials in terms of the physical constitutions, morphologies, biological characteristics, chemical properties and compositions (16, 17). For that reason, the sea sediment and surface soil samples, which did not undergo a combusted pretreatment, can have higher relative standard deviations of μ_m at low gamma-ray energy levels compared to the other types of samples. In biological samples, each sample can have different constituents due to enrichment or the selective uptake of inorganic substances (18). However, after combustion only a few elements with a high vaporization point remain in the minerals. This also explains the low relative standard deviation of μ_m in the combusted biological samples even for low energy levels.

The μ_m values with the relative standard deviation for the environmental samples as presented in table 4 will be able to be used as a reference in the determination of μ_m for self-attenuation correction within allowable uncertainties according to the purpose of gamma-ray spectrometry.

Table 4. The mean mass attenuation coefficient of environmental samples in an energy range of 46.5 to 1408 keV.

Energy (keV)	Sea Sediment (Dry)	Surface Soil (Dry)	Seaweed (Ash)	Pine Needle (Ash)	Fish (Ash)	Chinese Cabbage (Ash)	Milk (Ash)
46.5	0.437 (12.49)	0.423 (11.13)	0.645 (6.82)	0.574 (5.32)	0.576 (3.88)	0.583 (3.38)	0.616 (2.47)
59.5	0.273 (8.57)	0.273 (7.91)	0.360 (6.05)	0.341 (4.45)	0.327 (3.52)	0.332 (2.50)	0.342 (2.31)
121.8	0.153 (1.97)	0.153 (2.22)	0.159 (3.39)	0.156 (2.70)	0.159 (3.10)	0.164 (1.09)	0.162 (1.80)
244.7	0.111 (0.74)	0.113 (1.45)	0.109 (1.65)	0.109 (2.34)	0.109 (2.75)	0.111 (1.25)	0.109 (2.11)
344.3	0.101 (0.55)	0.102 (2.02)	0.100 (2.02)	0.102 (1.74)	0.101 (2.76)	0.103 (1.31)	0.101 (2.18)
444	0.091 (1.42)	0.091 (2.43)	0.088 (2.55)	0.092 (2.92)	0.089 (2.74)	0.092 (1.48)	0.090 (2.63)
661.7	0.078 (1.82)	0.078 (2.08)	0.075 (2.30)	0.077 (2.17)	0.077 (2.72)	0.079 (1.56)	0.077 (2.88)
778.9	0.073 (1.18)	0.073 (2.00)	0.072 (2.13)	0.074 (2.38)	0.073 (2.45)	0.075 (1.22)	0.074 (2.48)
1173.2	0.060 (1.09)	0.060 (2.22)	0.060 (1.78)	0.061 (2.81)	0.060 (2.71)	0.062 (1.53)	0.061 (2.97)
1332.5	0.056 (1.36)	0.057 (1.97)	0.058 (1.70)	0.059 (2.73)	0.057 (2.82)	0.059 (1.98)	0.059 (2.29)
1408	0.054 (1.72)	0.053 (2.22)	0.053 (1.74)	0.055 (2.70)	0.053 (2.71)	0.056 (1.94)	0.056 (2.27)

Relative standard deviations (%) are given in brackets in each column.

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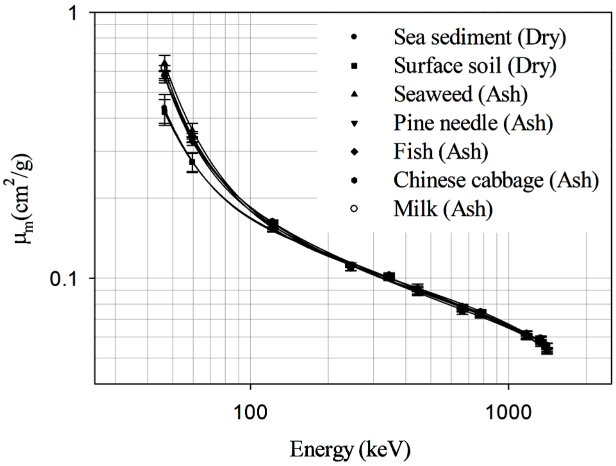

Figure 4. The mean attenuation coefficients (μ_m) versus the energy for each type of environmental sample.

CONCLUSION

The μ_m of seven kinds of environmental samples was estimated for gamma-ray energies ranging from 46.5 keV to 1408 keV. The standard deviation of μ_m for the same kinds of samples tended to decrease with an increasing gamma-ray energy. This showed that μ_m dose not significantly depend on the sample type for gamma-ray energy higher than around 100 keV for the studied environmental samples. In contrast, μ_m for the lower gamma-ray energy should be considered even with a similar sample type. Although analytical samples differ from those tested in this study, μ_m as presented here will be useful as a reference or comparable value for other analysts. Therefore, the μ_m database established through this study is expected to be used as reference data in the process of determining self-attenuation correction factors.

Conflicts of interest: *Declared none.*

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