Validation testing of radioactive Cesium measurement in Decontaminated waste with the Scattering Gamma Equivalent Method – 17238

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ABSTRACT

Canberra Industries, Inc. [CI], Now Mirion Technologies (Canberra) Inc. [MTC], has developed and validated a new Flexible Container Bag or Feed Roll Assay System 'FRAS' for Interim Storage Facilities [ISF] etc. There is an estimated 22 million cubic meters of soil and vegetation from the decontamination efforts around Fukushima. Removed soil and vegetation waste was put into flexible containers called Super Sacks [SS]. These SSs will be transported to ISF by trucks. These SSs will be divided into three categories according to total Cs radioactivity level (RL). This level is 'higher than 100,000 Bq/kg', '100,000>RL>8,000 Bq/kg' and '8,000 > RL >3,000 Bq/kg', respectively. The main uncertainty for SS measuring is internal inhomogeneity and heterogeneous source distribution. Japan Atomic Energy Agency [JAEA] has developed the Scattering Gamma Equivalent Model [SGE model] for decreasing uncertainty derived from these factors. This method was validated by Mirion Technologies (Canberra) KK [MCKK] and JAEA. According to the validation test, the SGE model can reduce the uncertainty of SS measurement from +/- 25% to +/- less than 15%.

INTRODUCTION

About six years have passed after Fukushima NPS accident. The decontamination of the land has made progress considerable. Removed soil and vegetation waste was put into flexible containers called Super Sacks [SSs] which sizes are 1.1m in Diameter x 1.1m in Height or 1.3m in Diameter x 1.3m in Height.

Now the concentration of SS is measured by a 'Survey meter' in Japan. Survey meter with a collimator placed in close contact with the SS, and generally with multiple other SSs nearby. In this document, this method is called 'Simple method'. This Simple method is computed from surface dose results multiplied by correction factor. The accuracy of this method is often plus or minus biased and has more than +/- 30% uncertainty. The simple method is affected by environmental back ground radiation, heterogeneous source distribution, matrix density inhomogeneity and so on. Therefore developing of new measurement system is desired, for example, it has good accuracy, high thorough put, easy measurable, good durability and so on.

CI (now, MTC) has developed and validated two single SS monitors. One of them is the Feed roll assay system (FRAS). This system can measure Feedroll, removed soil and vegetation in SS accurately. This system is on the bed of a truck and consists

of a large NaI detector. It is 3'x5'x16' in size, the same size as used in the Canberra FastScan Whole Body Counter. The detector is also LED-stabilized, to keep the gain constant under the wide range of temperature variations expected for this outdoor measurement instrument. The detector and the MCA are mounted in a water proof box, which is surrounded on the bottom and all sides by a 15cm thick steel shield. The SS is measured from bottom face. The other system is almost the same as FRAS, except for the detector size (3' x 3').

This type of system is affected by heterogeneous source distribution, matrix density inhomogeneity etc. of the SS.

The JAEA has developed the SGE model for decreasing uncertainty caused by these factors.

Theory

The 'Scattering Gamma Equivalent factor the [SGE factor]' is defined to quantify the influence of gamma ray attenuation of sample itself by the Compton effects. When a hot spot is at a deep position from a face of sample, low energy gamma ray is increased relatively by the Compton effects. In this case, the shape of spectrum is different from that of a homogenous sample, even if the average Cs concentration is the same in the sample. The SGE factor, $X_{geometry}$, is introduced as follows. When 'r' is the distance from source to detector, μ_a and μ_b are the liner attenuation coefficient of different energies of two gamma rays, gamma ray counting rate n_a and n_b are able to be shown $n_a \simeq e^{-\mu ar}/r^2$and $n_b \simeq e^{-ubr}/r^2$, respectively. In case of this, $\ln(n_a/n_b) \simeq -(u_a-u_b)r$, 'r' is able to be shown by logarithm of the ratio of the counting rate. When the density of the sample, in this case a SS, is almost the same, the influence of different positions of sources is mainly the item of '$1/r^2$'. Therefore the efficiency of gamma ray attenuation by Compton effects is able to be shown by '$1/r^2$'. When it substitutes $r \simeq \ln(n_a/n_b)$ for 'r', the $X_{geometry}$ can be written as following equation (1),

$$X_{geometry} = 1 \Big/ \left[ln \left\{ \frac{k}{\frac{n_a}{n_b}} \right\} \right]^2 \qquad (1)$$

where : n_a and n_b are count rate of gamma energy 'a' and gamma energy 'b', respectively.
'k' is emission rate of radio nuclide n_a and n_b, respectively.

When gamma rays from Radioactive Cs are analyzed, the objective radionuclide is Cs-137 because of its relatively long half-life. The objective gamma energy of Cs-137 are 284 keV (0.00058 %) and 662 keV (85.1 %), respectively. For evaluating $X_{geometry}$, different two count rates of gamma energies are needed. Though evaluable gamma ray is only 662 keV of Cs-137, scattering gamma ray area of Compton effects (mainly 300 to 400 keV) is analyzed as different energy gamma ray. This area is called 'SGA'. Figure 2 shows this SGA image. After five years from Fukushima NPS accident, the existence ratio of Cs-137 and Cs-134 was about 1 to

0.2, because The half-life of Cs-134 is about 2.065 years. Cs-134 has several gamma ray energies, for example, 569 keV (15.4%), 605 keV (97.6%), 796 keV (85.5%), 802 keV (8.7%) and so on. When the 662 keV line of Cs-137 was measured with a NaI detector, its spectrum peak includes the 605 keV and 569 keV lines of Cs-134, because of poor resolution. This type of peak is called triplet generally. In addition to this, SGA of Cs-137 includes scattering gamma ray of Cs-134 by Compton effects.

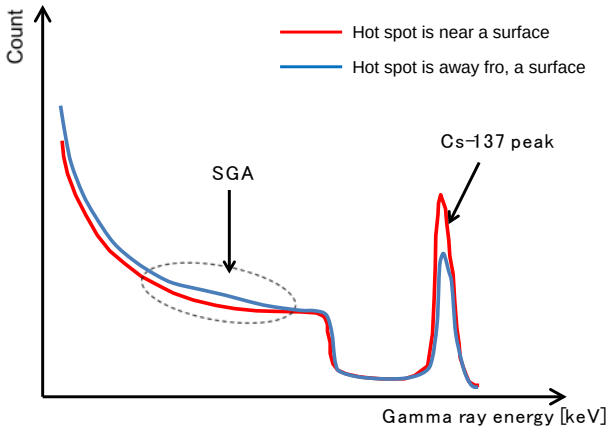


Fig.1 Image of Cs-137 peak and that of scattering gamma ray by Compton effects

The $X_{geometry}$ is calculated by the ratio of different two energy gamma ray. In case of only Cs-137 existence and both Cs-137 and Cs-134, the ratio of SGA of both two cases is approximately equal. Therefore $X_{geometry}$ is evaluated by following equation (2), using 662 keV of Cs-137 including Cs-134 and SGA counting ratio.

$$\frac{n_A}{n_B} = \frac{n_A^o+\delta_A}{n_B^o+\delta_B} = \frac{n_A^o}{n_B^o} \times \left(1+\frac{\delta_A}{n_A^o}\right) \Big/ \left(1+\frac{\delta_A}{n_A^o}\right) \quad (2)$$

where : n_A is the gamma ray count rate around 662 keV peak.
n_B is the count rate of SGA (300 to 400 keV).
n_A^o is net gamma ray count rate, excluded of SGA derives from 796 keV of Cs-134 and so on.
n_B^o is net count rate of SGA (300 to 400 keV), excluded of SGA derived from 796 keV of Cs-134 and so on.
δ_A^o is the count rate of SGA around 662 keV derived from 796 keV of Cs-134 and so on.
δ_B^o is the count rate of SGA (300 to 400 keV) deriverd from 796 keV of Cs-134 and so on.

The 'δ_A/n_A^o' and 'δ_B/n_B^o' have a same gamma count rate of same energy, therefore this value is nearly equal to constant in spite of Cs distribution. The '$(1+\delta_A/n_A^o)/(1+\delta_B/n_B^o)$' is regarded as the constant, so the $X_{geometry}$ can be calculated with the equation (3).
Because the count rate ratio between 662 keV peak and SGA can be regarded as the count rate ratio between the whole peak around 662 keV and SGA (300 to 400 keV).

$$X_{geometry} = 1 \Big/ \left[ln\left\{1\left(\frac{n_A}{n_B}\right)\right\}\right]^2 \quad (3)$$

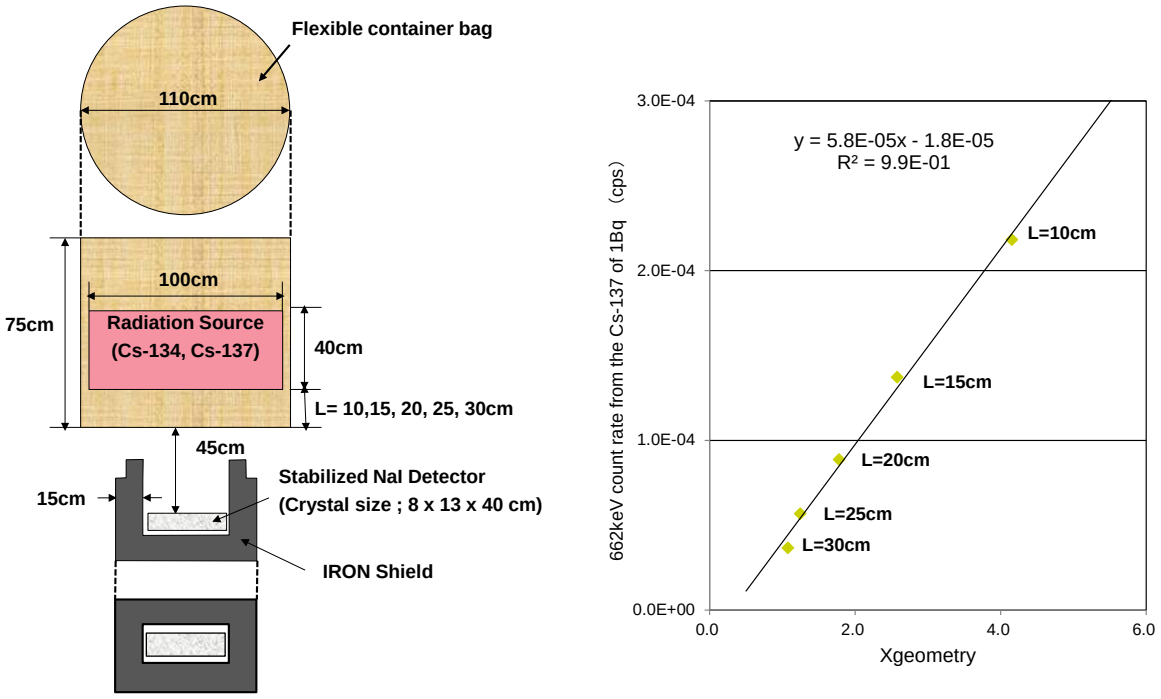
At this time, n_A and n_B in equation (3) are gross count rate.

Factor k in equation (1) needs emission rate of different two gamma ray energy, but the factor k of SGA is not able to define emission rate. Therefore k is nearly equal to 1 in this case.

The $X_{geometry}$ has correlation with the count rate per gamma ray emission. Because the correlation factor can be represented regression curve, it is usable as the callibration curve. When SS containing removed waste is measured, main objective radio nuclide is Cs-137. Therefore Cs-137 gamma ray counting rate of 662 keV is separated by radioactivity ratio of Cs-134/Cs-137. The radio activity ratio of Cs-134/Cs-137 was assumed as 1, when the Fukushima NPS accident occurred. Actual ratio of Cs-134/Cs-137 is calculated by Cs-134 and Cs-137 half-life and elapsed time from the date of Fukushima NPS accident to the measuring date. The Cs-134 content is evaluated by the Cs-137 activity and Cs-134/Cs-137 ratio.

Simulation

The relation between $X_{geometry}$ and gamma ray count rate of 662 keV was evaluated by Monte Carlo Simulation method. In this time, MCNP6 was selected. Inhomogeneous and heterogeneous source distributions of SS were modeled by the MCNP6 for measuring with the FRAS. This model is shown in Fig.2(a).



(a) Calculation model (b) Correlation of $X_{geometry}$ and 662 keV count

Fig.2 Simulation of γ-ray measurement of the SS.

The SS was simulated with the following boundary conditions, which were 110 cm diameter, 75 cm height, 1.0 g/cc and SiO_2 filling. The size of Cesium radiation source was 100 cm diameter and 40 cm height. The distance from the bottom of

the SS was set for 10, 15, 20, 25 and 30 cm. The MCNP6 simulation code needs gamma ray energy from radiation source. Objective radiation energies (and emission rate) of Cs-134 are 563 keV (8.4%), 569 keV (15.4%), 605 keV (97.6%), 796 keV (85.5%) and 802 keV (8.7%), respectively. That of Cs-137 is 662 keV (85.1%).

The value of $X_{geometry}$ was calculated by MCNP6 with following condition. At the equation (3), n_A is the count rate of 563, 569, 605 and 662 keV around 662 keV peak and n_B is the count rate of the SGA (300 keV to 400 keV) from those gamma rays. Figure 2(b) shows the correlation of $X_{geometry}$ and 662 keV count rate calculated by this simulation. The correlation factor was '$y = 5.8x10^{-5} x – 1.8x10^{-5}$ ($R^2 = 0.99$).

Validation Testing Procedure

The validation testing procedure is as follows. First, several SSs with known Cs-134 and Cs-137 concentration were prepared for the SGE model calibration. Second, the measurement data with the FRAS were evaluated with the SGE model. Finally, the measurement data of the FRAS and SGE model were compared with the result of in-situ Ge detector system.

Hardware and software:
Figure 3 shows a photograph of the FRAS setup and drawing of the detector frame and shielding. The shield is made of low background iron and weighs about 2300kg. The shield is 15 cm thickness. The collimator can be arranged in six positions depending upon sample activity and around environmental radiation level. The detector is a large NaI crystal, 3' x 5' x 16' (rectangular). This detector has Canberra's patented LED Stabilization circuitry, which will keep the gain stable over a -20 to +50 degree Centigrade temperature range. The typical resolution is 8% at 662keV. MCA is Canberra's 'Osprey' Digital Signal Analyzer. This compact and low-power MCA allows it to be mounted in the same weatherproof housing as the detector and is powered by a single POE Ethernet cable to the PC. The software is the Japanese version of Canberra Genie. The calibration is computed mathematically with MCNP. Calibrations have been performed every 0.01g/cc. The Genie software computes the net peak area of the Cs-134 and the Cs-137 peaks, using the regions on both sides of the peak as the background. This is possible since the shield effectively removes all Cs peaks from the background spectrum.

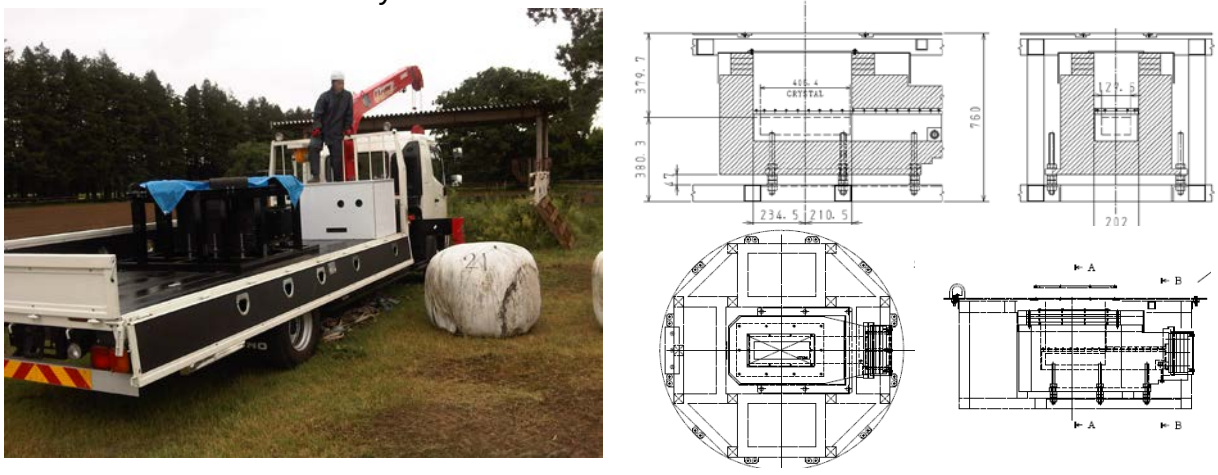


Fig.3 The FRAS photo, detector frame and the drawing of the shield

Preparation of SS for the SGE model calibration:
The Cs concentration of high contaminated soil samples was evaluated as following method. First, this soil was well mixed. Second, about 20 soil samples of 50g were gathered randomly. Third, 11 samples weighing about 1 kg were gathered randomly. Fourth, these 11 samples were measured with the Canberra Ge detector. Finally, mean Cs concentration and 1SD were evaluated from these samples. This high contaminated soil was put into a lot of small bags for easy usable. The Cs concentration of low contaminated soil samples was decided following method. First, bulk soil sample (high contaminated) was well mixed. Second, when soil sample (low contaminated) was put into SS, about 20 soil samples of 50 g were gathered from each shovel randomly. About 10 total 1kg samples were gathered from one SS. Each 1 kg sample was measured with Canberra Ge detector. Table 1 shows the result.

Table 1 The measurement results of gathering from each sample

Sample	Cs-134 (Bq/kg)		Cs-137 (Bq/kg)		Note
	Mean	SD	Mean	SD	
A	3831	165	13000	401	Filled only high contaminated soil
B	1343	103	4626	269	Filled only low contaminated soil
C	1597	143	5098	507	Only low contaminated soil result
D	1350	173	4458	641	Only low contaminated soil result

Figure 3 shows the characterization image of each sample. Sample A was uniform and put into only this high Cs contaminated soil. The weight of Sample A was 761 kg. Sample B was uniform and put into only low Cs contaminated soil. The weight of Sample B was 758 kg. Sample C and D were artificial heterogeneous source distributed SSs. As for sample C, 145 kg high contaminated soil was filled into the bottom layer and 713 kg low contaminated soil was filled into the middle and top layer of SS. As for Sample D, 143 kg high contaminated soil was filled into the middle layer and 778 kg low contaminated soil was filled into the bottom and top layer of SS. Table 2 shows the results of Sample A to D. The Cs concentration of Sample A and B were evaluated from 10 total 1kg (consists of twenty 50 g) samples. Sample C and D were evaluated from weight and mean Cs concentration of low and high contaminated soil in each SS.

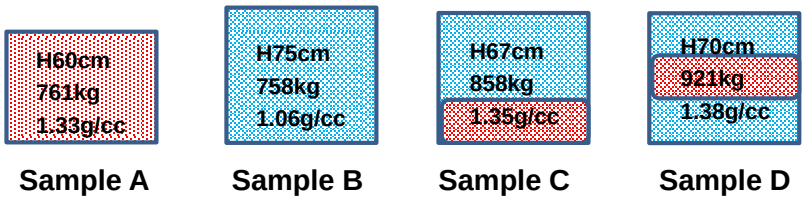


Fig.4 The characterization image of Sample A to Sample D. Red is high contaminated soil. Blue is low contaminated soil.

Table 2 The results and characterization of each sampl

Sample	Fill height	Weight (kg)			Density	Cs-134(Bq/kg)		Cs-137(Bq/kg)	
	(cm)	Total	Low	High	(g/cm^3)	Mean	SD	Mean	SD
A	60	761	0	761	1.33	3831	165	13000	401
B	75	758	758	0	1.06	1343	103	4626	269

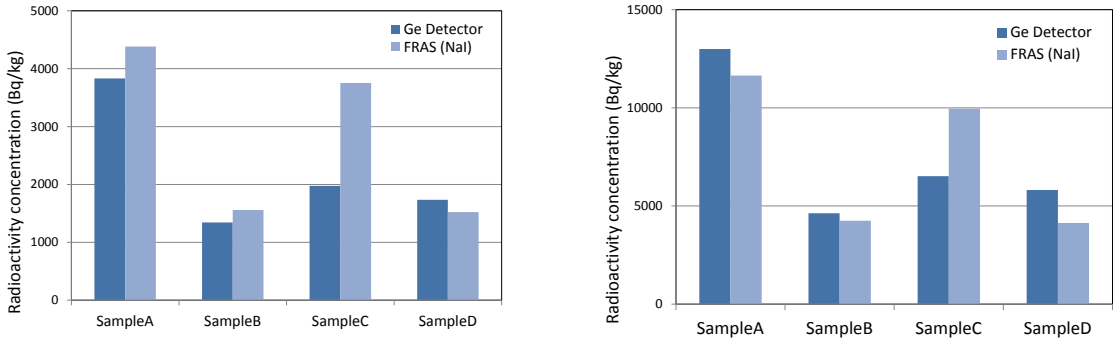
C	67	858	713	146	1.35	1975	122	6516	427
D	70	921	778	143	1.38	1735	148	5818	552

Validation Testing Results

Table 3 shows the results of each sample with th FRAS. Background and sample measurement time were 30 seconds. Figure 5 shows the bar graph of FRAS and Ge measurement results. The results of Sample C and D had a large gap from the results of in-situ Ge system.

Table 3 The measurement results of each sample with the FRAS

Sample	Cs-134 (Bq/kg)		Cs-137 (Bq/kg)	
	Mean	Error	Mean	Error
A	4383	96	11640	318
B	1557	38	4249	121
C	3754	109	9952	294
D	1522	37	4132	118



(a) Mean concentration of Cs-134. (b) Mean concentration of Cs-137.

Fig 5 Comparison of the radioactive Cs of the artificial SS with FRAS and Canberra Ge detector.

Fig ure 6 shows the spectra of BG and Sample A to D. The Compton scattering gamma region is about 300 to 400 keV in these spectra. Figure 7 shows the correlation between $X_{geometory}$ and 662 keV count rate. This $X_{geometry}$ was calculated by equation (3). The vertical axis of Fig. 7 is the 662 keV count rate from Cs-137 of 1 Bq. This count rate was calculated as follows. First, the count rate of Cs-137 of 662 keV was separated from interference peak of several Cs-134 gamma rays [563 keV (8.4%), 569 keV (15.4%) and 605 keV (97.6%)]. Second, separated Cs-137 of 662 keV count rate was divided by Ge measure ment result in

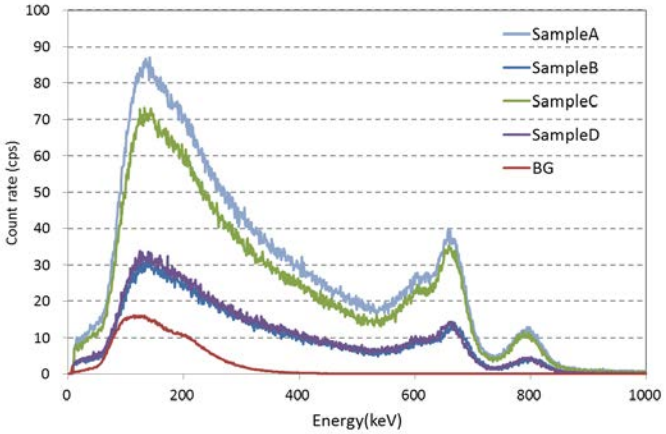


Fig.6 The spectra of BG and Sample A to D.

Table 2. The ratio of Cs-137 / Cs-134 was calculated by each half-life and elapsed time from the date of Fukushima NPS accident. The measurement date was October 8th, 2014. Therefore the elapsed year was about 3.6 years.

The correlation factor between the $X_{geometry}$ and the 662 keV count rate from Cs-137 of 1 Bq was 'y = $4.4x10^{-5}$ x – $4.1x10^{-5}$, R^2 = 0.91'. The decision factor is good and the convergence is relatively good.

The radioactivity of SS is able to be evaluated with the SGE model as follows. First, the calibration curve was calculated by the regression curve between the several conditions' $X_{geometry}$ value and the 662 keV count rate from Cs-137 of 1 Bq. Second, the measured count rate of 662 keV was confirmed where the $X_{geometry}$ was positioned on the regression curve. Though the regression curve of Fig.7 is able to be used as the calibration curve, the obtained measurement data of SS Samples are evaluated with the SG E model. Figure 8 shows the results evaluated with the SGE model.
Equation (4) was defined as 'Relative error: Re'.

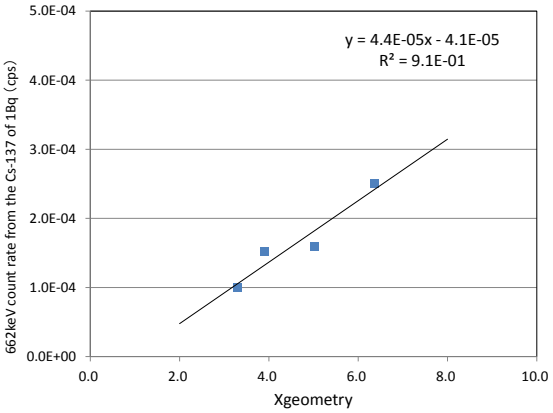


Fig.7 Correlation between $X_{geometry}$ and 662 keV count rate. Sample A and B have almost uniform distribution. Sample C and D have non-uniform Cs distribution. This regression curve is available as calibration curve.

$$R_e = \frac{A_C - A_{Ge}}{A_{Ge}} \qquad (4)$$

where: A_c is a NaI measurement data of each sample.
A_{Ge} is a Ge sampling measurement data as the 'True Value' of each sample.

The Re was evaluated for the FRAS measurement data and the FRAS measurement data with the SGE model. Figure 9 shows the correlation between these two.

The FRAS measurement Res of Sample A and B is about -10 %. The FRAS measurement Res with SGE model is about +/- 15%. The difference between the two was almost none. Because Cs distribution of Sample A and B is almost uniform.

The FRAS measurement Res of Sample C and D were more than +50 % and -30 %, respectively. On the other hand, the FRAS measurement Res with SGE model were only about +/- 5%. This strongly suggests the SGE method can evaluate Cs concentration appropriately, if Cs distribution of sample is heterogeneity.

Discussion

The result of Fig. 9 shows the SGE model can improve the uncertainty of non-uniform sample.

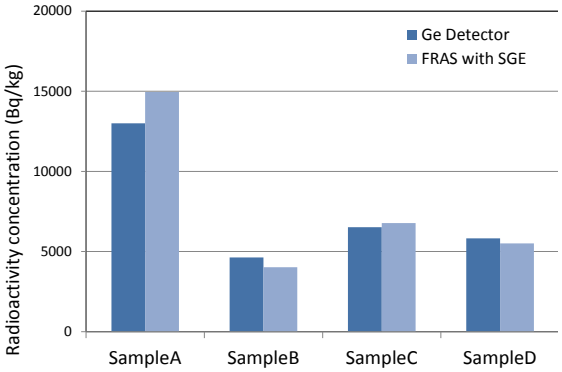


Fig.8 The Cesium-137 results comparison between the Ge measurement and the SGE model.

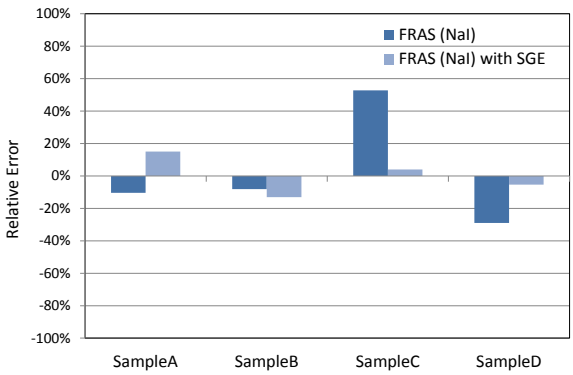


Fig.9 Relative error of the FRAS measurement and the SGE model compared with the Canberra Ge results.

As for the individual $X_{geometry}$ of each sample, those of Sample A and Sample B were relatively equal value. This is because these samples have almost uniform Cs distribution. On the other hand, the $X_{geometry}$ of Sample C was larger value than that of Sample A and B. Because Sample C has high contaminated soil at the bottom layer, the attenuation of gamma ray from high contaminated soil is smaller than that of uniform sample. The $X_{geometry}$ of Sample D was smaller value than that of Sample A and B. Because Sample D has high contaminated soil at the middle layer, the attenuation is larger. In this way, the $X_{geometry}$ shows the Cs distribution of each sample appropriately.

Figure 6 shows the $X_{geometry}$ is different value at different Cs distribution if sample has equal Cs concentration. In this validation testing condition, for example, max difference of the 662 keV count rate from Cs-137 of 1 Bq is about factor 2.5. When the regression curve of Fig.6 is used as calibration curve, the mean Cs concentration of various Cs distributions in a sample is evaluated accurately.

Conclusion

The SGE model is useful for evaluating of non-uniform radioactive sample. According to now evaluating validation testing results, the SGE model can hope to reduce the uncertainty of SS measurements from +/- 25% to +/- less than +/- 15%.

From the beginning of April 2017, the large scale validation testing of the FRAS with the SGE model will be conducted. More accurate evaluation of it will be done.

The SGE model can be applied other Mirion Technologies (Canberra) Inc. product. The evaluation of other products is going to be planned, for example, Box counter, Non-destructive assay system and so on.

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ACKNOWLEDGEMENTS

This research was done with the results of the cooperative research between Kajima Corporation and Canberra Japan KK. We appreciate Kajima Corporation very much.