

Quantitative Comparison of Activity Calculation Methods for the Selection of Most Reliable Radionuclide Inventory Estimation

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Abstract

It is important to know the accurate radionuclide inventory of radioactive waste for the reliable management. However, estimation of radionuclide concentrations in drummed radioactive waste is difficult and unreliable because of difficulties of direct detection, high cost, and radiation exposure of sampling personnel. In order to overcome these difficulties, scaling factors (SFs) have been used to assess the activities of radionuclides that could not be directly analyzed. A radionuclide assay system has been operated at KORI site since 1996 and consolidated scaling factor method has played a dominant role in determination of radionuclides concentrations. However, some problems are still remained such as uncertainty of estimated scaling factor values, inaccuracy of analyzed sample values, and disparity between the actual and ideal correlation pairs and the others. Therefore, it needs to improve the accuracy of scaling factor values. The scope of this paper is focused on the improvement of accuracy and representativeness of calculated scaling factor values based on statistical techniques. For the selection of reliable activity determination method, the accuracy of estimated SF values for each activity determination method is compared. From the comparison of each activity determination methods, it is recommended that SF determination method should be changed from the arithmetic mean to the geometrical mean for more reliable estimation of radionuclide activity. Arithmetic mean method and geometric mean method are compared based on the data set in KORI system.

1. Introduction

The measurement of radionuclide inventory contained in radwaste drum is very important for the

reliable management of radioactive waste in NPP. However, many radionuclides of which the declaration is required do not emit gamma radiation and must be analyzed by complicated radiochemical analysis. An established waste characterization program in KORI site measures the concentrations of gamma-emitting nuclides directly and estimates the concentrations of other relevant nuclides indirectly by relating Difficult-To-Measure (DTM) radionuclides to other Easy-To-Measure (ETM) radionuclides. Scaling factors are generated by use of sample data that are gathered from the radiochemical analysis of waste samples collected from different waste stream. The activity is determined by radionuclide assay system and scaling factor method.

However, it needs to collect the more number of samplings and to use reliable sampling procedures for the improvement of reliability. Furthermore, it needs to improve the accuracy of estimated SF values based on proper selection of activity calculation method. For that reason, research is in progress to improve the scaling factors for updating the radionuclide assay system, based on the statistical approach. Accuracy of each applicable activity determination method is compared using a foreign data set, and then the most reliable activity determination method is selected. The selected method and previous method used in KORI system is compared by the use of the sample-analyzed data set in KORI system.

2. Activity Determination Methods

Applicable activity determination methods are summarized in Table 1. Statistical techniques are used in these methods such as the arithmetical mean, geometrical mean, linear regression, and logarithmic regression [1, 2, and 3]. However, there is not any definition for the most reliable activity determination method. For that reason, each country uses its own preferred scaling factor method. In general, arithmetic mean is not used for the calculation of SF value in the activity determination method. However, arithmetical mean was used for the activity determination methods in KORI site because it has a little sample-analyzed data. In this study, two set of input data were used for the comparison of each method. At first, foreign data set is used for the comparison of four activity determination methods [3]. Next, KORI sample-analyzed data set is used for the comparison of arithmetic mean method and selected most reliable method [4]. For the comparison of each method, proper data set of key/ DTM nuclides in a specific waste type or all waste type was used. Detailed information of data set is summarized in Table 2.

Table. 1. Activity determination methods using key nuclides

Method		Mathematical expression	Coefficient	Activity determination
Arithmetic mean	Linear relation	$A_{RN}=a \cdot A_{KN}$	a= Average ratio [SF]	Arithmetic mean of SFs
Linear regression		$A_{RN}=a+b \cdot A_{KN}$	a, b=const.	Linear Regression of key & DTM nuclides

Geometric mean	Linear relation of Logarithm	$A_{RN}=c*A_{KN}$	c=Average ratio [SF]	Geometric mean of SFs
Linear regression of logarithms		$\text{Log}(A_{RN})=c'+d*\text{log}(A_{KN})$ $A_{RN}=c*(A_{KN})^d$	c, c', d =const.	Logarithmic linear regression of Key & DTM nuclides

Table. 2. Information of the input data set

Data set	(1) EPRI-5077	(2) KORI data set
Waste type (# of data set)	Spent Resin (139) (Excluding 7 extreme data)	A) Spent filter (4) B) Concentrate bottom (4) C) Spent resin (4) D) DAW (12)
Key nuclide	Co-60	Co-60, Cs-137
DTM nuclide	Ni-63	H-3, C-14, Ni-63 Sr-90, Tc-99, Gross alpha

3. Results and Discussion

The resulting plots of measured and estimated concentrations for each activity determination methods are shown in Figure 1. In arithmetic mean method, each activity and total activity are overestimated. Linear regression method is not proper for activity determination because it shows large disparity between the measured and estimated activities. The linear regression of logarithms has a characteristic to underestimate the total activity. The reason is that it overestimates the activities in low activity region and underestimates the activities in high activity region. Therefore, this method is under-conservative. In the geometric mean method, the estimated activity is very close to the measured one and total activity is conservatively estimated at the reasonable level. These comments could be confirmed through the comparison of measured and estimated concentrations of each activity determination method in Figure 1. More detailed comparison results of each activity determination method are summarized in Table 3. From the ideal case (ideal regression = reference line), we can establish the conservative conditions such as the following.

a) Individual activity

(Ideal case: A=1, B=0)

1) Estimated concentration \geq Measured concentration

2) $A \geq 1$ & $B \geq 0$

b) Ratio of total radioactivity

(Ideal case: Ratio=1; Estimated total activity = Measured total activity)

1) Ratio ≥ 1 (Estimated total activity \geq Measured total activity)

From the overall comparison of activity determination methods, it is concluded that geometric mean method is the most reliable activity determination method. Geometric mean and arithmetic mean are evaluated and compared by the use of the sample-analyzed data set in KORI system. The ratio values of arithmetic mean and geometric mean for each waste type and radionuclide are illustrated in Figure 2. The ratio values are higher than 1 for all waste types and pairs of DTM/Key radionuclides, which are corresponding to the viewpoint of statistics. This is particularly high in resin and DAW.

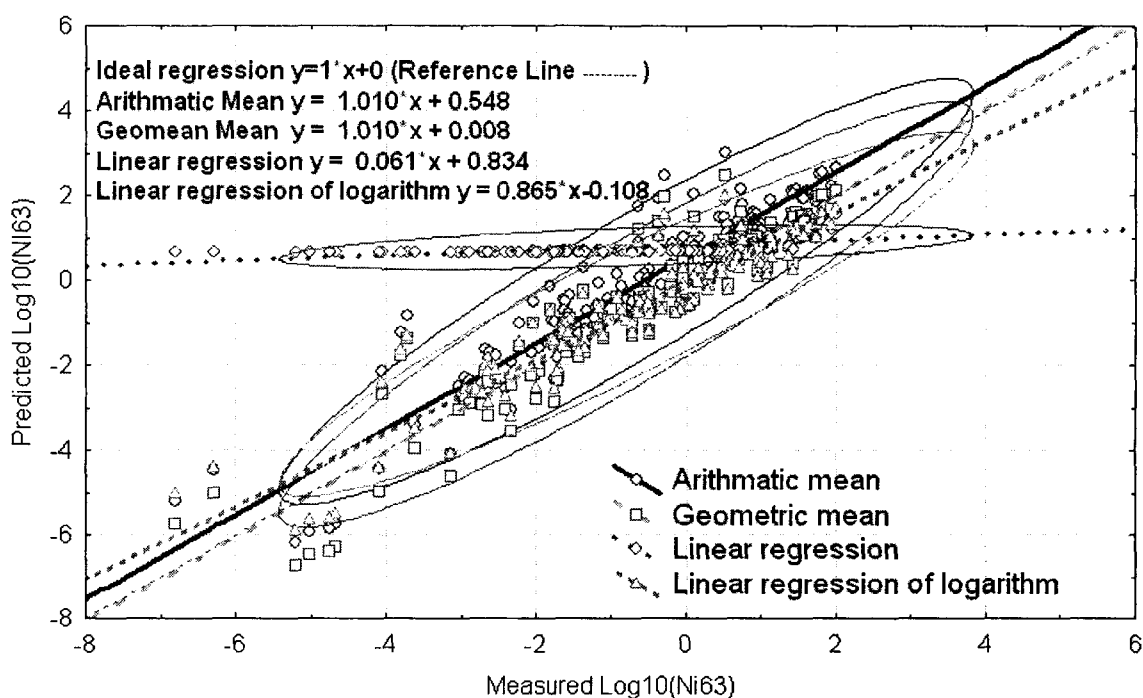


Fig. 1. Comparison of measured and estimated concentrations for each activity determination method

Table. 3. Comparison results of each activity determination methods

Method	Estimated (Y) Vs Measured (X) $Y=A*X+B$	Ratio of total radioactivity [Estimated /Measured]	Accuracy of estimation
Arithmetic mean	A=1.0101 B=0.5481	5.20	Overestimation of each activity and total activity
Linear regression	A=0.0607 B=0.8343	1.00	Large disparity between measured activity and estimated activity

Geometric mean	A=1.0101 B=0.0081	1.50	Proximity of each activity Conservative estimation of total activity
Linear regression of logarithms	A=0.8648 B=- 0.1080	0.65	In a low activity region : Overestimation In a high activity region : Underestimation Underestimation of total activity (Under-conservative)

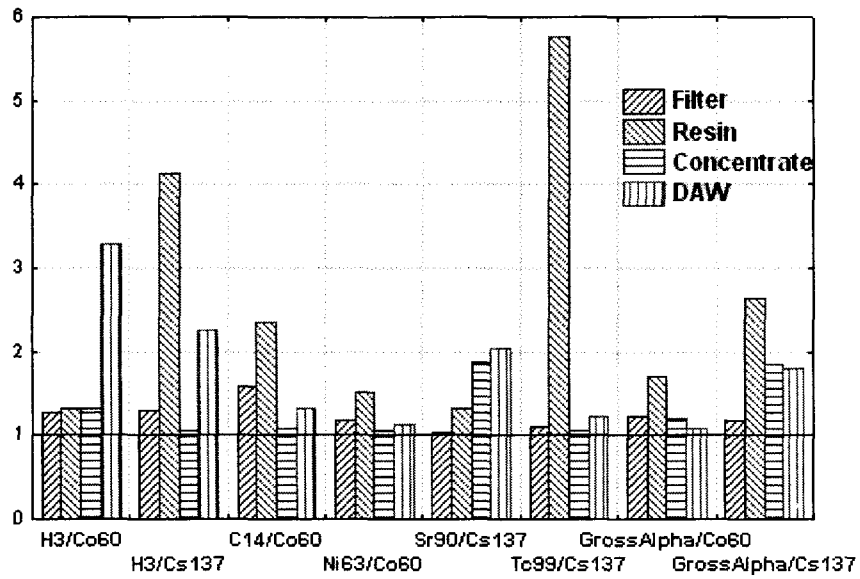


Fig. 2. Ratios of arithmetic mean to geometric mean for each waste type and pairs of radionuclides

4. Conclusion

For the evaluation of accuracy for each activity determination method, foreign and KORI data set were used. Inter-comparison was conducted in a viewpoint of accuracy and conservation of estimation. From the comparison of each activity determination method, it is concluded that geometric mean method is the most reliable activity determination method. Also, it is recommended that SF determination method should be changed from the arithmetic mean to the geometrical mean for the improvement of accuracy and reasonable conservation in activity determination. From the comparison of geometric and arithmetic means based on the sample-analyzed data set in KORI system, arithmetic mean is higher than geometric mean for all waste types and pairs of DTM/Key radionuclides. This is corresponded to a viewpoint of statistics. In particular, SF values in resin and DAW is higher than ones in other waste types.

An additional and frequent sampling procedure is in progress to update the performance of Korean nuclear waste management. As this study goes on, it is possible to get more accurate and reliable predictions for the information of radioactive waste based upon Korean analyzed database.

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