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Estimating the Activity Concentrations of Difficult-to-Measure Nuclides

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1 Introduction

The gamma-spectroscopic measurement makes it possible to observe nuclides with energies above 60 keV. However, nuclides at low gamma energy and pure beta and alpha active nuclides are not seen. The long-lived pure beta emitters which are present in the waste from nuclear reactors are produced in nuclear reactors either by activation (H-3, C-14, Ni-59, Ni-63, Nb-94) or by fission and transmutation (Sr-90, Tc-99, I-129, Cs-135, U-234, U-235, U-236, U-238, Pu-239, Pu-240, Am-241, Cm-242, Cm-244). If relationships exist between different radionuclides in the waste and if at least one of these radionuclides can be easily measured, waste characterization is simplified. The concentration of crucial (difficult-to-measure) nuclides may be related to some key nuclides. In this case the activity of the key-nuclides can be measured and the total inventory can be calculated using scaling factors. The scaling factors represent the relationship between a key radionuclide and other radionuclides, provided these relationships exist for all relevant radionuclides.

The key nuclides are presently measurable with a good accuracy and representative for activation (^{60}Co) or fission (^{137}Cs) reactions. At the time of waste arising the ratio of concentrations between a crucial nuclide and some key nuclide is constant for any NPP. This constant is called scaling factor. If the scaling factor is known, it can be used to determine concentration of the crucial nuclide and the total radioactivity concentration in a waste package. Scaling factors may vary somewhat between different types of reactors and also between individual reactors of similar type mainly due to fuel leakage. Two sets of scaling factors are used, a primary water related set and a surface contamination related set. The power plants have an extensive bookkeeping for the waste. The scaling factors are updated for various nuclides and waste categories periodically. This makes it

possible to adjust the total activity estimates in the storage, if, for example, it is found out that the previously used scaling factors were erroneous during a certain period.

The true scaling factor is unknown constant. However, for some crucial nuclides it could be estimated through a sample of measurements. Up to now, each country has reported their own scaling factor for the basic difficult-to-measure nuclides.

Scaling Factor Method is an empirical procedure for determining ratio between two nuclide concentrations in low-level waste. If $(K_1, C_1) \dots , (K_n, C_n)$ denote measurements of the concentrations of a key nuclide K and a crucial nuclide C from n random waste packages then

$$\sqrt[n]{\frac{C_1}{K_1}\frac{C_2}{K_2}\cdots\frac{C_n}{K_n}} \tag{1}$$

is often referred as "scaling factor" between C and K.

Scaling factor is used for determining (estimating) values of crucial nuclide concentration corresponding to particular values of key nuclide concentration. Further, it is used for estimating upper bound of radioactivity of the crucial nuclide and total radioactivity in waste.

Note that such a "scaling factor" may vary in different samples while the actual scaling factor between the two nuclides is a constant (for some period of time) for any particular NPP. The quantity (1) is only an estimator of the true scaling factor based on the particular sample and it use instead of the parameter should be justified.

Moreover the crucial nuclide is difficult to be measured so each measurement is subject to a random error. The measurement error will introduce error in the estimation (1) of the scaling factor. The variability of the random errors play an important role when estimating the crucial nuclide using estimated scaling factor. It reflects to the size of deviation of a prediction from an actual value of the crucial nuclide concentration.

In this paper we investigate the reasons of using quantity (1) instead of a scaling factor. We consider an appropriate statistical regression model of the relationship between concentrations of a crucial nuclide and a key nuclide. We suppose that measurements of the key nuclide are without any error while the measurements of the crucial nuclide has some error. The basic assumption is about distribution of the error term. It is naturally to assume that the measurement error of the crucial nuclide is much more higher for large measurements.

We determine interval estimations for the scaling factor based on a sample. The confidence limits for the parameter depend on the sample sizes and on the measurement error of the crucial nuclide. Similar considerations are given in [1] using not clear model. Furthermore, we estimate the confidence limits of the predicted value of the crucial nuclide using the model.

2 Model with Heteroscedastic Error

Suppose we are given a sample of concentration measurements of the two nuclides from n randomly chosen waste packages. We suppose that the key nuclide is measurable without any error while the crucial nuclide is measurable with a random error. Let $(K_1, C_1) \dots , (K_n, C_n)$ be the measurements of the concentration of two nuclides from n randomly chosen waste packages. We suppose that measurements satisfy the model

$$C_i = SF \cdot K_i \cdot e_i, \qquad (i = 1, \dots , n), \tag{2}$$

where SF (scaling factor) is unknown parameter and the random errors e_i follow Lognormal distribution law $LN(0, \sigma^2)$ with some unknown σ^2.

In the model with heteroscedastic error we assume that measurement error is multiplicative. This corresponds to the real situation in which small values of crucial nuclide concentration are more precise while larger values allow large discrepancy.

The model (2) is not linear but we can fit the first-order regression model

$$Y_i = \beta + X_i + \varepsilon_i \tag{3}$$

to the logarithm of the concentration measurements, that is $Y = \ln C$, $X = \ln K$, $\beta = \ln SF$, $\varepsilon = \ln e$.

Since the measurement error e has lognormal distribution, the the additive error $\varepsilon = \ln e$ in (3) has normal distribution with zero mean and σ^2 variance.

The least square estimate for β is then given by

$$\hat{\beta} = \bar{Y} - \bar{X}, \tag{4}$$

where $\bar{X}$ and $\bar{Y}$ are the arithmetic means of the logarithm transformed measurements of K and C, respectively.

Going the reverse transformation in (4) the corresponding estimate for SF is the geometric mean

$$\widehat{SF} = \exp\{\hat{\beta}\} = \exp\left\{\frac{\sum \ln C_i}{n} - \frac{\sum \ln K_i}{n}\right\} = \sqrt[n]{\frac{C_1}{K_1}\frac{C_2}{K_2}\cdots\frac{C_n}{K_n}}. \tag{5}$$

The estimation (5) of the scaling factor is the same as (1). However, the way it was defined has some advantages. First,the assumption about heteroscedastic error is essential in deriving of $\widehat{SF}$. Second, it is clear from the model that $\widehat{SF}$ is unbiased and consistent estimation of the unknown scaling factor. Moreover, for normal distribution it is also the estimator with smallest variance among all estimators of the scaling factor. The last property is essential for design of the experiment.

The assumption about distribution of the error allows to derive interval limits for the scaling factor.

3 Confidence Limits for the Scaling Factor

Each measured value of C is subject to a random error e that enters into the computations of $\hat{\beta}$ and $\widehat{SF}$ and introduces errors in these estimates. The main use of a scaling factor is to determine (estimate) a value $\hat{C}$ of the crucial nuclide concentration corresponding to a particular value K^* of the key nuclide concentration. The estimated value is

$$\hat{C} = \widehat{SF} \cdot K^*, \tag{6}$$

where $\widehat{SF}$ is the estimated scaling factor defined by (5).

This estimated concentration of the crucial nuclide is not exactly equal to the true concentration in the waste due to the fact that $\widehat{SF}$ is not equal to the true scaling factor.

Further, if we use the equation (6) to estimate (predict) some value of C, the random error will affect the estimation. Consequently, the variability of the random errors, measured by σ^2, reflects the estimation of C.

The first step toward acquiring a bound on a prediction error requires that we estimate σ^2, the variance of ε.

The regression method gives

$$s^2 = \frac{1}{n-1}\left[\sum_{i=1}^{n}\left(\log\frac{C_i}{K_i}\right)^2 - n(\log\widehat{SF})^2\right] \tag{7}$$

A level $1-\alpha$ confidence interval for the parameter β is determined by

$$\hat{\beta} \pm t_{\alpha/2,n}\frac{s}{\sqrt{n}},$$

where $t_{\alpha/2,n}$ is the upper $\alpha/2$ critical value for the t-distribution with n degrees of freedom.

The corresponding low and upper confidence limits for SF are determined by

$$\widehat{SF}_{LL} = \widehat{SF} \cdot \exp\left\{-t_{\alpha/2,n}\frac{s}{\sqrt{n}}\right\},\ \widehat{SF}_{UL} = \widehat{SF}.\exp\left\{t_{\alpha/2,n}\frac{s}{\sqrt{n}}\right\}. \tag{8}$$

The interval $\left[\widehat{SF}_{LL};\ \widehat{SF}_{UL}\right]$ covers the true value of the scaling factor with probability $1-\alpha$. It is also the shortest confidence interval with this confidence level since estimator $\widehat{SF}$ has smallest variance among all other estimators of the scaling factor.

In Section 5 we give some examples of estimating scaling factors and confidence limits for the true scaling factors.

A further goal of the estimating is to determine upper bound of radioactivity of the crucial nuclide in waste.

A $1-\alpha$ prediction upper bound for future measurement of the crucial nuclide C for a given concentration K^* of the key nuclide is

$$C \leq \widehat{SF} \cdot K^* \cdot \exp\left(t_{\alpha/2}\frac{s}{\sqrt{n}}\right).$$

The upper estimated bound of C depends on measured concentration K^*. It is much more large for large values of K than for small ones. Increasing the number of measurements in the model (2) will reduce the size of the upper bound.

4 Estimating the Age of Old Waste

Suppose that a waste package has been stored for some unknown time t. If the scaling factor between two nuclides K and C is known, then it can be used to determine the age of the waste. Denote by K_t and C_t the concentrations at the moment t and K_0 and C_0 – the concentrations at the moment of waste arising, Let T_K and T_C are half-life times of the two nuclides, From half-life equations of the two nuclides we get

$$\frac{C_t}{K_t} = \frac{C_0}{K_0}\, 2^{-\lambda t},$$

where and $\lambda = \left(\frac{1}{T_C} - \frac{1}{T_K}\right)$.

Therefore the estimated age of the waste is

$$\hat{t} = \frac{1}{\lambda}\left(\log_2 \widehat{SF} - \log_2 \frac{C_t}{K_t}\right).$$

We can also determine confidence limits for the true age t using confidence limits of the scaling factor:

$$\frac{1}{\lambda}\left(-\log_2 \frac{C_t}{K_t} + \log_2 \widehat{SF}_{LL}\right) \leq t \leq \frac{1}{\lambda}\left(-\log_2 \frac{C_t}{K_t} + \log_2 \widehat{SF}_{UL}\right),$$

where $\widehat{SF}_{LL}$ and $\widehat{SF}_{UL}$ are the low and the upper confidence limits of the scaling factor derived in (8).

5 Example Results

We include results from two data sets of concentration measurements. The first example uses real data coming from various waste streams of Paks NPP [2].

The second example is to illustrate the influence of different measure errors to the confidence limits for the scaling factor. We simulate 200 measurements a

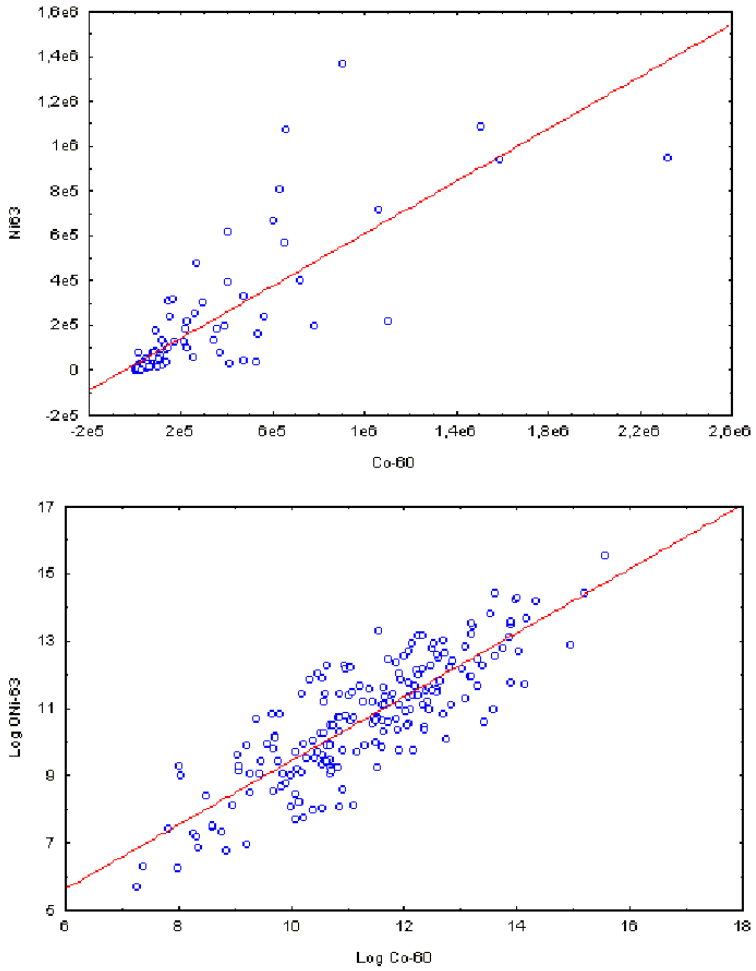

Figure 1. 200 simulated measurements.

crucial and a key nuclides with scaling factor corresponding to the scaling factor between ^{63}Ni and ^{60}Co in some NPP.

Example 1. ^{36}Cl is soft beta emitter with a half-life of 3.01×10^5 years. In niclear power plant ^{36}Cl is formed via neutron activation of ^{35}Cl in the cooling water syatem. Szántó and al. [2] give eight measurements of ^{36}Cl activity measured with Liquid Scintilation Counting method. Key nuclides ^{137}Cs and ^{60}Co were also measured. The measurements are as follow:

	^{36}Cl	^{137}Cs	^{60}Co	^{36}Cl/^{137}Cs	^{36}Cl/^{60}Co
1	1.5	2.97E+04	2.43E+03	5.0E-05	6.1E-04
2	3.5	7.71E+05	1.03E+06	4.5E-06	3.3E-06
3	3.4	4.55E+05	3.09E+06	7.4E-06	1.1E-06
4	4.7	6.82E+05	3.18E+04	6.9E-06	1.5E-04
5	0.4	3.26E+05	1.31E+04	1.3E-06	3.0E-04
6	0.1	6.85E+03	2.06E+04	1.4E-05	4.8E-06
7	1.3	1.78E+06	1.40E+05	7.2E-07	9.2E-07
8	1.8	1.48E+06	4.20E+04	1.2E-06	4.3E-06

Correlation between ^{36}Cl and ^{137}Cs is 0.9663, and between ^{36}Cl and ^{60}Co is 0.8813. Using (5) we estimate the scaling factors of ^{36}Cl relative to ^{137}Cs and ^{60}Co. Then the 0.95% confidence limits for the true scaling factors are calculated using (8):

	low CL	$\widehat{\mathbf{S}F}$	upper CL
^{36}Cl/^{137}Cs	1.27E-06	**4.56E-06**	1.64E-05
^{36}Cl/^{60}Co	2.99E-06	**1.94E-05**	1.26E-04

Confidence limits are calculated using t(0.95,7)= 2.365.

Example 2. One source of the generation of radionuclides in NPPs is the activation of reactor materials and their corrosion products in the reactor core. ^{63}Ni is among the activated corrosion product nuclides whose concentration are limited in most low-level waste disposal facilities. To determine the activity concentration of these difficult-to-measure nuclides, each country usually select ^{60}Co as key nuclide and determines the concentration of ^{60}Co by direct measurements of waste packages. Figure 1 shows the relationship between activity concentration of ^{60}Co and ^{63}Ni. Data is simulated according distributions from [1]. The estimated scaling factor between nuclide concentrations is $\widehat{SF} = 0.76$ and the confidence limits calculated by (8) are $\widehat{SF}_{LL} = 0.72$ and $\widehat{SF}_{UL} = 0.815$.

References

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[2] Zs. Szántó, E. Hertelendi, J. Csongor and J. Gulyás, (1997) Determination of ^{36}Cl in nuclear waste (Institute of Nuclear Research of the Hungarian Academy of Sciences Debrecen) *Hungary Annual Report* (*http://www.atomki.hu/ar97/e/e08.pdf*)