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GENERALIZED SCALING FACTOR FOR ESTIMATING THE ACTIVITY CONCENTRATIONS OF DIFFICULT-TO-MEASURE NUCLIDES

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Scaling factors represent the relationship between a crucial radionuclide concentration and other radionuclides concentrations. In this paper a generalization of the Scaling Factor Method is proposed.

1. Introduction

Reactors generate a spectrum of radionuclides some of which are hard to detect. The gamma-spectroscopic measurement makes it possible to observe nuclides with energies above some sensitive level. However, nuclides at low gamma energy and pure beta and alpha active nuclides are not seen. Because these radionuclides never influence the classification status of wastes from nuclear reactors, over the short term, waste generators can be highly conservative about reporting these radionuclides without jeopardizing their ability to ship waste to low level waste disposal facilities. But unfortunately, the total inventories of these radionuclides in disposal facilities tend to strongly influence calculations of impacts caused by possible long-term release of radioactive elements from the disposal facilities.

Scaling Factor Method is an empirical procedure for determining ratio between two nuclide concentrations in low-level waste. If a scaling factor between concentrations is known and at least one of the nuclides is easy to measure, than it can be used to determine concentration of the crucial nuclide and the

total radioactivity concentration in a waste package. This method is generally applicable to reactors that generate a spectrum of radionuclides including hard to detect radionuclides, such as pure beta or alpha emitters. In this method, scaling factors for individual radionuclides are established based on direct measurement of representative samples. Scaling Factors are then applied to ratio hard-to-detect nuclides to easy to detect nuclides based on subsequent analysis for the easy-to-detect nuclides only. The scaling factor is then applied to gross measurement methodology.

In this paper we consider an appropriate statistical regression model of the relationship between concentrations of a crucial nuclide and some key nuclides. We generalize the Scaling Factor Method for the case of several key nuclides. We suppose that measurements of nuclide concentrations include some random errors. The basic assumption is about distribution of the measurement errors. It is naturally to assume that the measurement errors are much more higher for large values of the concentration.

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. Scaling factor procedure for two nuclides is studied in [3]. Similar considerations are given also in [1] using not clear model. Furthermore, we estimate the confidence limits of the predicted value of the crucial nuclide using the model.

2.   Generalized scaling factor

Denote $K_1, \ldots, K_m$ concentrations of m key nuclides that are regularly measured in the waste of any particular NPP. Denote C concentration of a difficult to measure crucial nuclide also presented in the waste. Since C is difficult to measure its concentration is determined using indirect methods. At the time of waste arising the ratio of the crucial nuclide concentration and each of the key nuclide concentrations is constant. This constant is called Scaling Factor.

Suppose that scaling factors between the crucial nuclide and each of the m key nuclide are known. If key nuclide concentrations are measured without any error then the crucial nuclide concentration is related to the key nuclide concentrations by

(1) $$C = f_j K_j, \qquad j = 1, \ldots, m,$$

where f_j is the scaling factor between C and K_j. Thus concentration of the crucial nuclide in a waste package can be determined trough a measurement of the concentration of any key nuclide.

Nevertheless key nuclide concentrations are easy to measure, measurement always includes a random error. The measurement error introduces error in prediction of the crucial nuclide concentration when any of the relationships(1) is used. Different equations from (1) will predict different value for C. In the model bellow we assume that measurement errors are multiplicative. This corresponds to the real situation in which small values of concentrations are more precisely measured while larger values allow large discrepancy. It appears then that the log-normal model may be suitable statistical model for nuclide concentration in low-level radioactive waste if the available empirical data conform to it in some reasonable fashion.

In this section we consider the problem of choosing "the best" prediction of the crucial nuclide concentration C using measurements of the key nuclide concentrations K_i and their scaling factors f_j ($j = 1, \ldots, m$). We want to choose one or more key nuclides in order to estimate crucial nuclide concentration with smallest prediction interval.

2.1. Model with heteroscedastic error

Let $(K_1, \ldots, K_m)$ be the measurements of the concentration of the m key nuclides from any waste package. We assume that the measurement includes random errors. We suppose that crucial nuclide concentration is related to the key nuclide concentrations by

(2) $$C = f_j \cdot K_j \cdot e_j, \qquad (j = 1, \ldots, m),$$

where measurement error e_j of j-th nuclide follows Log-normal distribution law $LN(0, \sigma_j^2)$ with some known σ_j^2. The corresponding estimates for C are $\hat{C}_j = f_j K_j$ ($j = 1, \ldots, m$).

The crucial nuclide concentration satisfies (2) and consequently

$$\ln C = \ln f_j + \ln K_j + \varepsilon_j, \qquad j = 1, \ldots, m,$$

where the additive error $\varepsilon_j = \ln e_j$ has normal distribution with zero mean and σ_j^2 variance.

The last relationships are used to obtain confidence intervals for C. The $1 - \alpha$ confidence intervals for $\ln C$ are given by

$$\ln C \in \left[(\ln f_j + \ln K_j) \pm z_{\alpha/2} \sigma_j \right], \qquad j = 1, \ldots, m,$$

where $z_{\alpha/2}$ is the upper $\alpha/2$ critical value for the normal distribution function.

The corresponding $1 - \alpha$ confidence intervals for C are determined by

$$C \in \left[(f_j K_j) \exp(\pm z_{\alpha/2} \sigma_j) \right] \qquad j = 1, \ldots, m.$$

If we want to choose a single key nuclide to predict C then we should use the key nuclide with smallest prediction interval for C. That is the key nuclide with smallest σ_j^2.

Take now two key nuclides with smallest variances among all key nuclides. WOLOG we have K_1 and K_2 with variances σ_1^2 and σ_2^2 ($\sigma_1^2 < \sigma_2^2$). According model (2) measurements satisfy

$$\ln C = \ln f_j + \ln K_j + \ln e_j, \qquad (j = 1, 2).$$

Combining both equations we get

$$\ln C = \frac{1}{2}(\ln f_1 + \ln f_2) + \frac{1}{2}(\ln K_1 + \ln K_2) + \frac{1}{2}(\ln e_1 + \ln e_2),$$

where the additive error $\frac{1}{2}(\ln e_1 + \ln e_2)$ has normal distribution with zero mean and variance equal to $\frac{1}{4}(\sigma_1^2 + \sigma_2^2)$.

The corresponding prediction equation for C is

(3) $$\hat{C}_{1,2} = \sqrt{f_1 f_2}\sqrt{K_1 K_2}\sqrt{e_1 e_2},$$

where $\sqrt{f_1 f_2}$ is called *Generalized Scaling Factor* between crucial nuclide and the two key nuclides.

The last equation gives better prediction for C than any of the two key nuclide gives if the variance of the error term is less than the smallest variance σ_1^2. That is

$$\frac{1}{4}(\sigma_1^2 + \sigma_2^2) < \sigma_1^2.$$

Therefore, we use combined estimate (3) of C instead of $\hat{C}_1 = f_1 K_1$ if $\sigma_2^2 < 3\sigma_1^2$.

Further, let r key nuclides with smallest variances give better prediction interval than any smaller subset of key nuclides. The measurements satisfy

$$\ln C = \frac{1}{r}\sum_{j=1}^{r} \ln f_j + \frac{1}{r}\sum_{j=1}^{r} \ln K_j + \frac{1}{r}\sum_{j=1}^{r} \ln e_j.$$

The corresponding prediction equation for C is

(4) $$\hat{C}_{1,\ldots,r} = F\left(\prod_{j=1}^{r} K_j\right)^{\frac{1}{r}},$$

where $F = \left(\prod_{j=1}^{r} f_j \right)^{\frac{1}{r}}$ is the *Generalized Scaling Factor* of the r key nuclides.

The nuclide with the next smallest variance is included in the prediction equation (4) if the variance of $\frac{1}{r+1} \sum_{j=1}^{r} \ln e_j$ is less than the variance of $\frac{1}{r} \sum_{j=1}^{r} \ln e_{ji}$. That is

$$\sigma_{r+1}^2 < \frac{2r+1}{r^2} \sum_{j=1}^{r} \sigma_j^2.$$

3. Estimating the Generalized Scaling Factor

Suppose that scaling factors between a crucial nuclide and a set of key nuclides are not known. We want to estimate the generalized scaling factor through a sample of measurements of the key nuclides and the crucial nuclide.

The usual Scaling Factor Method is an empirical procedure for determining ratio between two nuclide concentrations in low-level waste (see [3]). If $(K_1, C_1) \ldots, (K_n, C_n)$ denote measurements of the concentrations of a key nuclide K and a crucial nuclide C from n randomly chosen waste packages then the following relations are assumed

(5) $$C_i = f.K_i \cdot e_i, \qquad (i = 1, \ldots, n),$$

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

The estimate $\hat{f}$ given by

(6) $$\widehat{f} = ^n \sqrt{\frac{C_1}{K_1} \frac{C_2}{K_2} \cdots \frac{C_n}{K_n}}$$

is an unbiased estimate of the scaling factor f between C and K. Moreover, for Log-normally distributed errors it is also the estimate with smallest variance among all estimates of the scaling factor.

The least squares estimate for the variance σ^2 is given by

$$s^2 = \frac{1}{n-1} \left[\sum_{i=1}^{n} \left(\ln \frac{C_i}{K_i} \right)^2 - n(\ln \widehat{f})^2 \right]$$

The estimated variance is used for estimating the confidence interval for the simple scaling factor and for the predicting interval of crucial nuclide concentration. A level $1 - \alpha$ confidence interval for the parameter $\ln f$ is determined by

$$\ln \hat{f} \pm z_{\alpha/2} \frac{s}{\sqrt{n}},$$

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

$$(7) \qquad \widehat{f}_{LL} = \widehat{f} \cdot \exp\left\{-z_{\alpha/2}\frac{s}{\sqrt{n}}\right\}, \qquad \widehat{f}_{UL} = \widehat{f} \cdot \exp\left\{z_{\alpha/2}\frac{s}{\sqrt{n}}\right\}.$$

Consider now m key nuclides and let $K_{1i}, \ldots, K_{mi}, C_i$ be measurements in the i-th waste package $(i = 1, \ldots, n)$. We suppose that measurements satisfy

$$(8) \qquad C_i = f_j K_{ij} \varepsilon_{ij}, \quad j = 1, \ldots, m; \; i = 1, \ldots, n,$$

where f_j is the simple scaling factor between crucial nuclide concentration and concentration of the j-th key nuclide.

The random error ε_{ji} is a product of the measurement error e_{0i} of the crucial nuclide and the measurement error e_{ji} of the j-th key nuclide. Assuming that e_{0i} and e_{ji} have Log-normal distribution $LN(0, \sigma_0^2)$ and $LN(0, \sigma_j^2)$, respectively, it follows that ε_{ji} is Log-normally distributed $LN(0, \sigma_0^2 \sigma_j^2)$.

The parameters f_j and σ_j^2 $(j = 1, \ldots, m)$ are assumed unknown.

Using (6) the least squares estimates for simple scaling factors are

$$(9) \qquad \hat{f}_j = \left(\prod_{i=1}^{n} \frac{C_i}{K_{ij}}\right) \quad j = 1, \ldots, m.$$

The measurements satisfy (8) and consequently

$$\ln C_i = \ln f_j + \ln K_{ij} + \ln \varepsilon_{ij}, \quad j = 1, \ldots, m; \; i = 1, \ldots, n,$$

where the error $\ln \varepsilon_{ij}$ is normally distributed $N(0, \sigma_0^2 + \sigma_j^2)$.

Summarizing over j and i we get

$$\frac{1}{n}\sum_{i=1}^{n} \ln C_i = \frac{1}{m}\sum_{j=1}^{m} \ln f_j + \frac{1}{mn}\sum_{i=1}^{n}\sum_{j=1}^{m} \ln K_{ij} + \frac{1}{mn}\sum_{i=1}^{n}\sum_{j=1}^{m} \ln \varepsilon_{ij}.$$

Since the measurement errors ε_{ij} are Log-normally distributed then $\eta = \frac{1}{mn}\sum_{i=1}^{n}\sum_{j=1}^{m} \ln \varepsilon_{ij}$ is normally distributed with zero mean and variance

$$\frac{m\sigma_0^2 + \sum_{j=1}^{m} \sigma_j^2}{m^2 n}.$$

Therefore the least squares estimate for $\ln F := \frac{1}{m}\sum_{j=1}^{m} \ln f_j$ is given by

$$\ln \widehat{F} = \frac{1}{n}\sum_{i=1}^{n} \ln C_i - \frac{1}{mn}\sum_{i=1}^{n}\sum_{j=1}^{m} \ln K_{ij},$$

and the corresponding estimate for the Generalized Scaling Factor $F = \left(\prod_{j=1}^{m} f_j\right)^{\frac{1}{m}}$ is

$$
(10) \qquad \widehat{F} = \left[\prod_{i=1}^{n} C_i \left(\prod_{j=1}^{m} K_{ij}\right)^{-\frac{1}{m}}\right]^{\frac{1}{n}}.
$$

The optimal estimate for the variance of η is

$$
(11) \qquad s^2 = \frac{\sum_{j=1}^{m} s_j^2}{m^2 n},
$$

where

$$
s_j^2 = \frac{1}{n-1}\left[\sum_{i=1}^{n}\left(\log \frac{C_i}{K_{ji}}\right)^2 - n(\log \widehat{f_j})^2\right]
$$

and

$$
\widehat{f_j} = \left(\prod_{i=1}^{n} C_i/K_{ji}\right)^{\frac{1}{n}}.
$$

3.1. Confidence limits for the Generalized Scaling Factor

Each measured value of the nuclide concentrations is subject to a random error e that enters into the computations of $\widehat{f_j}$ and $\widehat{F}$ 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 $K^* = (K_1^*, \ldots, K_m^*)$ of the key nuclides concentrations. The estimated value is

$$
(12) \qquad \hat{C} = \widehat{F}\left(\prod_{j=1}^{m} K_j^*\right)^{\frac{1}{m}},
$$

where $\widehat{F}$ is the estimated generalized scaling factor defined by (10).

If the equation (12) to estimate (predict) some value of C, the measurement errors will affect the estimation. Consequently, the variability of the random errors, measured by σ^2, reflects the estimation of C.

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

$$
\ln \hat{F} \pm z_{\alpha/2}\frac{s}{\sqrt{n}},
$$

where s is the standard deviation of η defined by (11) and $z_{\alpha/2,n}$ is the upper $\alpha/2$ critical value for the Standard Normal distribution.

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

$$(13) \qquad \widehat{F}_{LL} = \widehat{F} \exp\left\{-z_{\alpha/2}\frac{s}{\sqrt{n}}\right\}, \qquad \widehat{F}_{UL} = \widehat{F} \exp\left\{z_{\alpha/2}\frac{s}{\sqrt{n}}\right\}.$$

The interval $\left[\widehat{F}_{LL};\ \widehat{F}_{UL}\right]$ covers the true value of the scaling factor with probability $1-\alpha$. It is also the shortest confidence interval with this confidence level since estimate $\widehat{F}$ has smallest variance among all other estimates of the scaling factor. In section 4. 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 $K_1^*, \ldots, K_m^*$ of the key nuclide is

$$C \leq \widehat{f}\left(\prod_{j=1}^{m} K_j^*\right)^{\frac{1}{m}} \exp\left(z_{\alpha/2}\frac{s}{\sqrt{n}}\right).$$

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 (5) will reduce the size of the upper bound.

4.   Example results

^{36}Cl is soft beta emitter with a half-life of 3.01×10^5 years. In nuclear power plant ^{36}Cl is formed via neutron activation of ^{35}Cl in the cooling water system. The example uses real data coming from various waste streams of Paks NPP. 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 (9) we estimate the simple scaling factors of ^{36}Cl relative to ^{137}Cs and ^{60}Co, respectively. Then the 0.95% confidence limits for the true simple scaling factors are calculated using (7).

	low CL	$\widehat{\textbf{SF}}$	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
$^{36}Cl/^{60}Co^{137}Cs$	6.30E-06	**9.41E-06**	1.40E-05

The last row of the table give to the generalized scaling factor of ^{36}Cl relative to both ^{137}Cs and ^{60}Co (eq. 10), and confidence limits for this estimate (eq. 13).

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

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