

**R-05-76**

## **Assessment of uncertainty to correlation factors**

Peter Cronstrand, SwedPower

December 2005

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## Summary

Uncertainties for correlation factors have been assigned to nearly 40 radionuclides through a scheme, which uses both statistical methods as well, as qualitative expert judgements. When the data are sufficient, the approach is initiated by evaluating intervals of 90% confidence for the assumed log-normal distribution. The uncertainty is adjusted in order to compensate for various deficiencies in the data employed. The range of uncertainty factors is 5–50, where the highest uncertainty is associated with assessments made exclusively with data from a reference BWR spent fuel assembly or radionuclides with extremely contradictory data. Unless new data is provided the only possible route to higher accuracy would be to consider waste type-specific correlation factors.

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# 1 Introduction

## 1.1 Purpose

The purpose of this report is to determine the uncertainty associated with correlation factors employed in the safety assessment of SFL3–5 /1/ when estimating the activity and composition of nuclear waste repositories. Although the data for most radionuclides are scarce, the ambition is to develop a systematic scheme based on statistical methods, but supported with qualitative judgements.

## 1.2 Background

Since it is not feasible to perform a complete survey of all radionuclides present in all waste packages on a routine basis and because a substantial part of the waste has not been produced yet, estimates of the final content cannot entirely be based on measurements, but must be supported with calculation as well as assumption. Radionuclides that will dominate the activity may not constitute the main potential hazard since other radionuclides, less abundant and with a minor influence on the activity, may have higher mobility. Clearly, highest priority will be given to those of importance to the long-term safety of the repository and in order to critically review the long-term safety it is necessary to be able to estimate a wide selection of radionuclides. An essential technique in this context is correlation factors between a few easily detected key radionuclides and the remaining radionuclides, which are more difficult to detect. This correlation is for several reasons an intricate and non-trivial relation. The waste packages are not uniform and although the major portion of waste to be deposited in the nuclear waste repository originates from the operation and decommission of nuclear power plants, the repository will also receive waste from industry, hospitals and research. Moreover, the content of radionuclides in the waste packages will have different formation paths (see Table 1-1) and therefore are radionuclides mainly correlated with members within the same path of formation. Accordingly, activation products are mainly correlated to  $^{60}\text{Co}$ , fission products to  $^{137}\text{Cs}$  and transuranics to the sum of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ . It is emphasized that the classification is not completely exclusive and certain radionuclides may be created during alternative pathways. Despite the considerable variation of waste of different type and origin, only one distinction, referring to the type of activity, has been introduced. Alternative distinctions, referring to the reactor type or the waste type, could also be motivated on the basis of the observed variation. However, the presumed gain in accuracy would correspond to a loss of simplicity and generality.

It is evident from the brief discussion above that the correlation factors inherently contain large portions of uncertainty and the assessments of correlation factors benefit greatly from a detailed analysis and a scheme for quantifying the sources of uncertainty.

**Table 1-1. The radionuclides classified according to formation pathways. Key radionuclides that are easy to measure are marked with bold notation.**

Activation products	Fission products	Transuranics
$^3\text{H}$ , $^{10}\text{Be}$ , $^{14}\text{C}$ , $^{36}\text{Cl}$ , $^{55}\text{Fe}$ , <b><math>^{60}\text{Co}</math></b> , $^{59}\text{Ni}$ , $^{63}\text{Ni}$ , $^{93}\text{Zr}$ , $^{93}\text{Nb}$ , $^{93}\text{Mo}$ , $^{99}\text{Tc}$ , $^{107}\text{Pd}$ , $^{108}\text{Ag}$ , $^{113}\text{Cd}$ , $^{125}\text{Sb}$ , $^{133}\text{Ba}$ , $^{147}\text{Pm}$ , $^{151}\text{Sm}$ , $^{152}\text{Eu}$ , $^{154}\text{Eu}$ , $^{155}\text{Eu}$ , $^{166}\text{Ho}$	$^{79}\text{Se}$ , $^{90}\text{Sr}$ , $^{93}\text{Nb}$ , $^{99}\text{Tc}$ , $^{107}\text{Pd}$ , $^{108}\text{Ag}$ , $^{113}\text{Cd}$ , $^{126}\text{Sn}$ , $^{125}\text{Sb}$ , $^{129}\text{I}$ , $^{134}\text{Cs}$ , $^{135}\text{Cs}$ , <b><math>^{137}\text{Cs}</math></b> , $^{147}\text{Pm}$ , $^{151}\text{Sm}$ , $^{152}\text{Eu}$ , $^{154}\text{Eu}$ , $^{155}\text{Eu}$ , $^{166}\text{Ho}$	$^{238}\text{U}$ , $^{237}\text{Np}$ , $^{239}\text{Np}$ , $^{238}\text{Pu}$ , <b><math>^{239}\text{Pu}</math></b> , <b><math>^{240}\text{Pu}</math></b> , $^{241}\text{Pu}$ , $^{242}\text{Pu}$ , $^{244}\text{Pu}$ , $^{242}\text{Am}$ , $^{243}\text{Am}$ , $^{241}\text{Am}$ , $^{242}\text{Cm}$ , $^{243}\text{Cm}$ , $^{244}\text{Cm}$ , $^{245}\text{Cm}$ , $^{246}\text{Cm}$

## 2 Method of approach

### 2.1 The chemical basis for correlation factors

Correlation factors facilitate the estimates of the nuclide content of waste repositories by providing a linear relation between a few easily measured radionuclides as  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  and the other nuclides of interest. In a schematic model originally devised by Thierfeldt et al. /4/ the number of activated isotopes,  $N$ , can be represented by

$$\frac{dN}{dt} = \sigma M - \lambda N - \varepsilon N \quad (1)$$

where  $M$  is the number of inactivated source elements,  $\sigma$  the activation constant,  $\lambda$  the decay constant and  $\varepsilon$  the removal rate constant. Under the assumption that the number of inactivated source elements,  $M$ , is constant, the equation has the solution

$$N = \frac{\sigma M}{\lambda + \varepsilon} (1 - e^{-(\lambda + \varepsilon)t}) \quad (2)$$

For all radionuclides considered relevant for nuclear waste repository one can assume that the decay rate is much lower than the removal rate,  $\lambda \ll \varepsilon$ , and that the time of interest is long compared to the inverse of the removal rate and the decay rate, i.e. confined to asymptotic behaviour

$$N = \frac{\sigma}{\varepsilon} M \quad (3)$$

In a linear approximation the correlation between two elements,  $N_i$  and  $N_j$ , therefore is given by

$$b = \frac{N_i}{N_j} = \frac{\sigma_i M_i \varepsilon_j}{\sigma_j M_j \varepsilon_i} \quad (4)$$

Thus, within the assumptions above and with a constant ratio between source elements there is a linear correlation. However for many short-lived intermediate species acting as parent radionuclides the decay may be on the order of the removal rate,  $\lambda \approx \varepsilon$ , or even  $\lambda \gg \varepsilon$ . During this timescale the parent radionuclide may migrate to different parts of the system and the major production of the long-lived daughter nuclides may therefore occur at scattered locations depending on reactor type and other rather specific conditions. In conclusion, even though the asymptotic behaviour implies a linear correlation for the long-lived radionuclides, the transient behaviour for the intermediate species may cause a substantial spread in this correlation. This is the reason that for instance  $^{137}\text{Cs}$ , although easily detected, is non-trivial as a correlating radionuclide. Due to the rather specific formation path via the noble gas  $^{137}\text{Xe}$ ,  $^{137}\text{Cs}$  will exhibit different concentrations in PWR and BWR reactor waste.

This may be one of the reasons behind the observed non-linearity for the correlation factors. According to Thierfeldt et al. /4/ their data are more adequately described by introducing an exponent,  $a$ , in the following relation

$$N_i = bN_j^a \quad (5)$$

The same authors introduce mechanisms representing the removal of activity in their model, but this still does not explain the observed deviations. Instead they propose explanations related to varying ratio of source elements, unspecific non-linear effects or higher order

term in the original equation for instance introduced by a weak coupling between two activation constants. All hypotheses are reasonable, but impossible to verify with the available data. A less exotic, but considering the limited range of exponents, quite plausible interpretation is that the apparently non-linear correlation stems from the averaging of the data from many waste streams and many reactors. Thus, a different approach is to assume a linear correlation factor, but with such degree of fluctuation that it most adequately can be described by a log-normal distribution. This is the typical distribution occurring when the final parameter is a product of several independent variables. A linear correlation is also the most reasonable way to incorporate the wide selection of non-uniform sources of assessments into one single value. Admittedly, a rigorous  $\chi^2$ -test would be desirable, but not possible because the scarcity of data.

From the spread of the data, it is possible to identify at least three different types of variations dividing the waste in different classes with respect to the correlation, namely:

- Type of activity (surface contamination or induced activity).
- Type of reactor (BWR or PWR).
- Type of waste (core components, internal parts, resins, etc).

Seemingly, this would require at least  $2^3 = 8$  different sets of correlation factors, which would ruin the simple predictability. It is also questionable whether the existing data could support so many different types of correlation factors.

## 2.2 Uncertainty of correlation factors

The main purpose of this report is to evaluate the uncertainty associated with the correlation factors and introduce a scheme for quantifying these uncertainties. Taking into account the scattered character of the available data, the following critical issues can be identified:

- The overall sparse information for the majority of radionuclides.
- The variability of sources of data.
- The contradictions of data within and between different waste streams.
- The difficulty of estimating to which extent a certain data represents the general waste.

Based on these points various strategies can be constructed:

1. A set of pure qualitative judgements for each radionuclide.
2. Application of statistical methods where data is sufficient, compensating for the deficiencies with qualitative judgements.
3. Fully incorporate all data and qualitative aspects in a statistical model with conversion factors for each waste stream.

Due to the non-uniform character of the data as well as the goal to avoid judgements that may appear subjective, the strategy described in this report here follows the middle path and consists of.

1. A statistical module where the geometric mean and the standard deviation about the geometric mean are determined when data admits. The average and maximum deviation from the selected value is calculated for some of the waste streams.
2. A qualitative post-statistical process where the uncertainty is modified due to the inherent weaknesses of the data.

Whereas the first module tries to introduce an objective and systematic scheme, the second tries to compensate the heavily non-uniform character of the data and address questions such as: Is the amount of data sufficient for supporting general statements? Which of the data sets provides the most accurate correlation? Does the available data represent a general waste type or is it too specific?

### 2.2.1 The statistical module

It is not uncommon that data require transformation prior to analysis in order to normalize them. In particular, for skewed distributions the log-transformation is well suited. Log-normal distributions typically arise in situations where the final value is a product of many independent contributions. This can be compared with a situation where the final value is the sum of many independent contributions, which lead to the normal distribution.

As clearly depicted in the majority of diagrams presented in previous compilations, the sets of data appear to follow a log-normal distribution [1]. In fact, logarithmic scaled diagrams seem to be the only way to display the wide span of data points. Moreover, a logarithmic approach also ensures an equal statistical weight for all points. Otherwise single high values would influence the estimated correlation more strongly than many data points with low values. An alternative approach in order to assign a measure for strongly skewed distributions is to simply employ the median instead of the geometric mean. However, this does not support a more elaborate statistical approach where also other important measures, as an estimate for the variance, can be included in the analysis. The values presented will be averages from a wide selection of compilations obtained by different methods and hence the proposed values will have rather large uncertainty intervals, but thereby also to some extent reflecting the diversity among the waste packages.

A log-normal distribution is best characterized as a normal distribution of the logarithms of the data points. Let  $c$  denote the correlation factor and  $C$  the corresponding logarithm, i.e.

$$C = \log(c) \quad (6)$$

Where the available data admit an estimation of the logarithmic mean,  $\bar{C}$ , is simply calculated through

$$\bar{C} = \frac{1}{N} \sum_{i=1}^N C_i \quad (7)$$

which is equal to the geometric mean of the non-logarithmic values. A corresponding estimation,  $S$ , of the standard deviation in the log-realm is calculated through

$$S = \frac{1}{N} \sum_{i=1}^N (C_i - \bar{C})^2 \quad (8)$$

and an interval of confidence for the logarithmic values can be constructed as

$$I_m = (\bar{C} - t_{\alpha/2}(f)d, \bar{C} + t_{\alpha/2}(f)d) \quad (9)$$

where

$$d = \frac{S}{\sqrt{n}} \quad (10)$$

$n$  is the number of values and  $f$  the degree of freedom

$$f = n - 1 \quad (11)$$



and  $t_{\alpha/2}$  the percentile of  $\alpha$  for the Student's  $t$ -distribution. In principle this is a confidence interval about the geometric mean. However, there is a primary difficulty in the fact that the mean and the variance of the log-normally distributed variable are not independent. Instead there are several coexisting recommendations for the calculation of confidence intervals [14]. Since the statistical measures in this report are not important per se, but merely represent a quantitative guideline in a non-quantitative assessment, the confidence intervals for the non-logarithmic data simply are restored by a straightforward back transformation of Equation 6. In analogy, the following interval of confidence represents the original non-logarithmic values.

$$I_m = (e^{\bar{c}-t_{\alpha/2}(f)d}, e^{\bar{c}+t_{\alpha/2}(f)d}) \quad (12)$$

Clearly, the factor here represents the statistical uncertainty for the assessment of the correlation factor. Thus, the outermost limits of the confidence intervals can be calculated as

$$c_{\max} = \bar{c} \cdot e^{t_{\alpha/2}d} = \bar{c} \cdot \bar{\sigma} \quad (13)$$

$$c_{\min} = \bar{c} / e^{t_{\alpha/2}d} = \bar{c} / \bar{\sigma} \quad (14)$$

which in principle is a confidence interval about the geometric mean. The variable here is defined as the statistical uncertainty factor, not to be confused with a proper standard deviation.

For all series of data, the strategy has been to calculate a logarithmic average through the geometric mean and an accompanying uncertainty factor through an estimate of the standard deviation in order to evaluate a statistical uncertainty factor corresponding to a 90% interval of confidence. This factor is subsequently modified by various qualitative arguments. This final stage reflects the limitations of the data being too waste type-specific and is considered as a generalisation of the result derived by purely statistical means. The final uncertainty factor will be denoted  $\sigma$ .

## 2.2.2 The qualitative module

The uncertainty factor will, when data admits, be based on a statistical evaluation, but always modified by qualitative arguments. Because of the great variation of sources of data, issues will arise that need to be addressed outside the domain of statistics. These include typically

- Waste, plant or reactor type-specific considerations.
- Time aspects, especially for short-lived radionuclides.
- The composition of the material and the content of impurities.
- The chemistry underlying the surface contamination processes.

These issues reflect the fact that each data is specific, but the waste general. To some extent these aspects are included when the data stems from different sources and thereby expands the uncertainty intervals. However, when data is confined to a single value, additional uncertainty needs to be included. In available compilations one can identify following distinct categories of waste streams:

- Data based on measurements of Swedish operational waste and reactor coolant.
- Data based on non-Swedish waste.
- Data based on a reference BWR spent fuel assembly.
- Analogies with results obtained for induced activity.

For certain waste streams it is possible to evaluate a mean and maximum deviation from the selected value, see Table 2-1. The deviation,  $d$ , is here defined as the ratio of the selected value and the statistically derived value if the latter underestimates the former or vice versa to ensure that it is a value larger than 1. Technically this can be achieved by for instance the following definition:

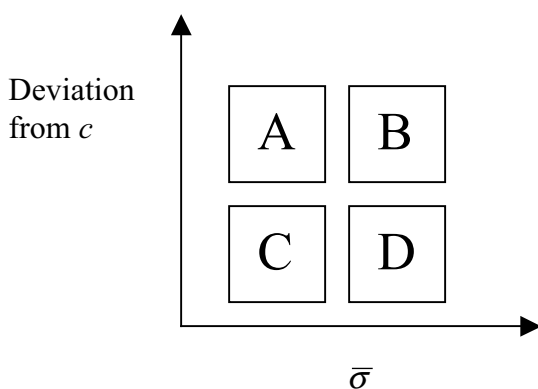
$$d = \exp(\text{abs}(\ln(c / \bar{c}))) \quad (15)$$

Although data are too scarce for providing conversion factors for the correlation for each waste type, the deviations do suggest limits for maximum uncertainty to radionuclides where information is sparse and/or the correlation factor is based on one single category. Thus, for radionuclides where the correlation factor entirely relies on a single data point from a reference BWR spent fuel assembly /9/, a maximum uncertainty of a factor 50 is selected.

Substantial differences between selected value and a geometric mean will call for special considerations. Is the difference an indication of large uncertainties or does it represent a systematic difference for a certain type of waste? In general, if the deviation is correlated to a small variance within the set of data, the corresponding geometric mean is considered to represent that particularly waste stream but apparently to a lesser extent the waste as such. The deviations from the selected value and the statistical uncertainty divide the region of uncertainty in four sub regions, as displayed in the scheme in Figure 2-1.

**Table 2-1. The maximum and the average deviation from the selected value for certain waste categories.**

Data source	Average deviation	Maximum deviation
Shippingport /6/	8	50
Swedish reactor coolant /4/	7	25
Reference BWR spent fuel assembly /9/	5	50
Swiss model inventory /8/	1.5	45



**Figure 2-1. Four different sub regions of uncertainty.**

Where

- A corresponds to radionuclides where the combination of low statistical uncertainty factor and large deviation from the selected value indicates a systematic difference.
- B defines a region with high uncertainty.
- C defines a region with low uncertainty.
- D is characterized by large variances within the set of data but also a – perhaps fortuitous – coincidence with the selected value. In this region it is valuable to consider the average deviation for the waste stream that actually was used for the assessment.

No general fixed limits guiding these decisions have been postulated, but a deviation is in general considered as large if it exceeds the average deviation for the particular waste stream.

In order to rationalize the assessment of uncertainty six different categories of uncertainty have been introduced according to Table 2-2.

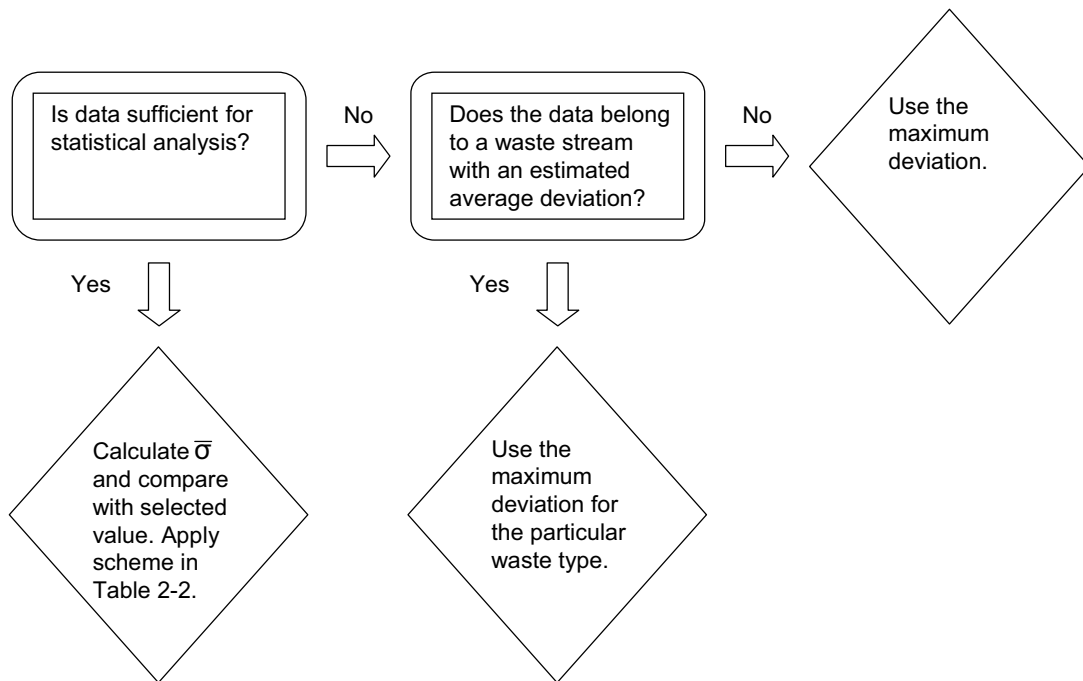
The overall methodology can be summarized in the scheme described in Figure 2-2. As revealed from this figure, the scheme tries to quantify the uncertainty, but also to address the deficiencies of the data employed. The major weakness arises due to the assumption that the measured spread for a particular set of data is adequate for assessments made upon other data (which are considered as more relevant, but even scarcer). Furthermore, the assumed maximum uncertainty is based on deviations from the selected data within a restricted set of data. A true maximum deviation may naturally be even larger.

An objective employed is to use  $^{137}\text{Cs}$  as the key nuclide for all fission products,  $^{60}\text{Co}$  for all nuclides with induced activity and  $^{239}\text{Pu}+^{240}\text{Pu}$  for all radionuclides in the natural decay chain (see Table 1-1). For species belonging both to activation and fission products the choice of key radionuclide is based on availability of data. This is, as seen from the discussion in Section 2.1, not entirely trivial due to the sometimes complicated formation paths via short lived intermediate species, for instance noble gases. However, subtle improvements which correlate radionuclides according to a more detailed formation path would require both additional correlating radionuclides as well as new sets of sophisticated – probably system specific – classes.

Despite these drawbacks, it is likely, due to the two-step process, that the scheme provides confidence intervals of at least 90%.

**Table 2-2. The criteria employed for assessing the uncertainty factors.**

Category	$\sigma$	Criteria
1	3	$\bar{\sigma} < 3$ <b>and</b> a low deviation from the selected value.
2	5	Either $3 < \bar{\sigma} < 5$ <b>or</b> $\sigma < 3$ in combination with large deviation from the selected value.
3	10	$5 < \bar{\sigma} < 10$ and low deviation from the selected value <b>or</b> $3 < \bar{\sigma} < 5$ and large variation from the selected value.
4	20	$10 < \bar{\sigma} < 20$ and low deviation from the selected value <b>or</b> $5 < \bar{\sigma} < 10$ and large variation from the selected value.
5	40	$20 < \bar{\sigma} < 40$ and low deviation from the selected value <b>or</b> $10 < \bar{\sigma} < 20$ and large variation from the selected value.
6	50	$\bar{\sigma} > 40$ <b>or</b> assessment based one single datapoint.



**Figure 2-2.** The scheme employed when assigning the uncertainty of correlation factors.

## 3 Surface contamination

### 3.1 Background

Surface contamination in BWR and PWR reactors comprises activated corrosion products, fission products and heavy nuclides from leaking fuel. The chemical processes involve two initial irreversible steps characterized as a corrosion of the base metal followed by an erosion of the corrosion film. The corrosion products are transported to the core and deposited on fuel surfaces and thus activated. The final products of the sequences of complicated equilibriums can then be released and transported to various sinks in the reactor coolant system. Because of the complexity of the ingoing processes it is not possible to evaluate any simple rules of thumb without the aid of extensive modelling. It is notable that Fe and Ni can precipitate independent on each other, whereas Co, according to /3/ cannot precipitate, neither as independent oxide nor in mixed oxide with Fe. The deposition occurs solely through absorption on and solid solution in the other metal oxides. Although, there appears to be a substantial difference in the chemistry behind the deposition of the key nuclide  $^{60}\text{Co}$  and the correlating radionuclides, such as  $^{63}\text{Ni}$ , one could still expect a linear correlation because the same type of system will lead to the same type of equilibrium processes. However, this also underscores that each radionuclide has a rather unique chemistry and analogues are of limited use.

### 3.2 Uncertainties for correlation factors to $^{60}\text{Co}$ and $^{137}\text{Cs}$

All radionuclides will be addressed through the scheme outlined in Sections 2.2.1–2.2.2 and the data and results summarized in tables of the following type;

<i>A</i>	<i>Key</i>	<i>c<sub>sfa</sub></i>	<i>c</i>	$\bar{c}$	<i>S</i>	<i>n</i>	$\bar{\sigma}$	$\sigma$
–	–	–	–	–	–	–	–	–

Where

- *A* denotes the radionuclide of study.
- *Key* denotes the key nuclide (i.e.  $^{60}\text{Co}$ ,  $^{137}\text{Cs}$ ,  $^{239,240}\text{Pu}$ ).
- *c<sub>sfa</sub>* is the assessment based on a reference BWR spent fuel assembly. The reason for including this value in a summarizing table is that it is the only type of assessment that applies for almost all radionuclides. If not otherwise stated this refers to the value after five years decay.
- *c* denotes the selected correlation factor by /1/.
- $\bar{c}$  denotes the correlation factor evaluated on statistical basis, i.e. the geometric mean of the employed data points.
- *S* denotes the standard deviation of the log-values.
- *n* denotes the number of data points employed in the statistical evaluation.
- $\bar{\sigma}$  denotes the statistical uncertainty factor, i.e.  $e^{t_{w/2}(f)d}$ .
- $\sigma$  denotes the final uncertainty factor assigned on top of the statistical uncertainty factor.

The maximum deviation between the values derived from a reference BWR spent fuel assembly /9/ and the selected value is occasionally as high as a factor of 50. This value consequently is assigned as the maximum uncertainty when data are confined to a single value stemming from a reference BWR spent fuel assembly. In cases when data consist of additional waste streams but information still is sparse (see Table 3-1), the uncertainty factor is chosen to reflect both the scarcity and the degree of mutual agreement between available data according to scheme in Figure 2-1.

**Table 3-1. A subset of radionuclides where the information is sparse.**

Assessed purely on data from a reference BWR spent fuel assembly	Assessed on data from a reference BWR spent fuel assembly + 1 other reference	Assessed on data from a reference BWR spent fuel assembly + 2 other references
<sup>10</sup> Be <sup>93</sup> Mo <sup>108m</sup> Ag <sup>166</sup> Ho	<sup>107</sup> Pd <sup>113m</sup> Cd, <sup>126</sup> Sn <sup>93</sup> Zr <sup>79</sup> Se <sup>133</sup> Ba <sup>152-155</sup> Eu	<sup>147</sup> Pm <sup>151</sup> Sm <sup>241</sup> Pu

### 3.2.1 Uncertainty factors

#### <sup>3</sup>H

The selected value is based on analogue with induced activity. If species such as FeO(OH) can be contained in the oxide layer this would lead to a higher correlation factor for surface contamination than for induced activity. In any case the uncertainty is increased and a maximum value for the uncertainty is selected.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>3</sup> H	<sup>60</sup> Co	–	1·10 <sup>-4</sup>	–	–	–	–	50

#### <sup>10</sup>Be

There are no data from measurements, except from a reference BWR spent fuel assembly /9/. The selected value corresponds to activity after a five-year decay period, which motivates the choice of the maximum uncertainty i.e. a factor 50.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>10</sup> Be	<sup>60</sup> Co	6·10 <sup>-10</sup>	6·10 <sup>-10</sup>	–	–	–	–	50

#### <sup>14</sup>C

Carbon shows a strong dependence on waste type and origin /2/. The BWR data from /4/ are scattered, but symmetrical on a log scale. The suggested correlation factor, 1·10<sup>-3</sup>, refers to Swedish operational waste /10/ and is based on the assumption that 1% of <sup>14</sup>C in the coolant water will be found in the operational waste. From the data from Shippingport station (7 points) compiled in /6/ merged with data from spent resins samples from Swedish reactors /20/ an estimate of the logarithmic standard deviation can be evaluated to 6.2 which gives a 90% interval of confidence with a statistical uncertainty factor of 7.7. The moderate deviation between the selected value and the geometric mean motivates a minor increase of the uncertainty factor to 10.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>14</sup> C	<sup>60</sup> Co	4·10 <sup>-4</sup>	1·10 <sup>-3</sup>	9·10 <sup>-4</sup>	6.2	19	7.7	10

### <sup>36</sup>Cl

The formation <sup>36</sup>Cl is strongly dependent on the specific water chemistry, which in conjunction with the scarcity of data leads to substantial uncertainties. The available data consist of Swiss measurements of resins ( $1.4 \cdot 10^{-6}$ ,  $2.2 \cdot 10^{-6}$  and  $1.7 \cdot 10^{-5}$ ) and reactor coolant water ( $5.8 \cdot 10^{-5}$ ). Data from a reference BWR spent fuel assembly /9/ yields  $1 \cdot 10^{-5}$  <sup>36</sup>Cl/<sup>60</sup>Co, which is higher though more uncertain since it refers to <sup>60</sup>Co instead of <sup>137</sup>Cs and is based on a reference BWR spent fuel assembly. Although the scheme suggests a value of 10, the low number of data points and their rather specific origin an uncertainty factor of 20 is selected.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>36</sup> Cl	<sup>137</sup> Cs	$1 \cdot 10^{-5}$	$1 \cdot 10^{-5}$	$4 \cdot 10^{-6}$	0.96	4	2.1	20

### <sup>55</sup>Fe

The Shippingport Station data for <sup>55</sup>Fe in /6/ are far less scattered than for <sup>14</sup>C and an estimate of the logarithmic standard deviation can be evaluated to 2.5, which gives a 90% interval of confidence with a statistical uncertainty factor of 4. Data from a reference BWR spent fuel assembly /9/,  $7 \cdot 10^{-1}$ , is in agreement with an average correlation based on Swedish operational waste,  $7 \cdot 10^{-1}$  /10/. An uncertainty factor of 5 is selected because the low statistical uncertainty and a deviation from the selected value in the order of the average deviation for the particular waste stream.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>55</sup> Fe	<sup>60</sup> Co	$7 \cdot 10^{-1}$	$7 \cdot 10^{-1}$	$1 \cdot 10^{-1}$	2.5	7	4	5

### <sup>59</sup>Ni

The data for <sup>59</sup>Ni are similar to <sup>55</sup>Fe quite uniform. Merging the Shippingport Station data from /6/ with data from measurements of Ni content in water from Swedish PWR reactors /4/ and recent measurements on ion-exchange resins from Swedish reactors /21/ give a low estimate of the standard deviation. Because of the – in this context – rather large set of data points a statistical uncertainty factor as low as 1.9 can be evaluated. No decisive difference was found between the two sets of data. Moreover data from other sources proposes values in close agreement ( $1 \cdot 10^{-3}$  by Thegerström et al. /10/ and  $7 \cdot 10^{-4}$  by Lundgren /5/). An uncertainty factor of 3 is therefore selected.

A	Key	c <sub>sfa</sub>	c	$\bar{c}$	S	n	$\bar{\sigma}$	$\sigma$
<sup>59</sup> Ni	<sup>60</sup> Co	$1 \cdot 10^{-3}$	$1 \cdot 10^{-3}$	$2 \cdot 10^{-3}$	1.8	15	1.9	3

### <sup>63</sup>Ni

The comprehensive data from reactor coolant water of Swedish BWR reactors /4/ in conjunction with recent measurements on ion-exchange resins from Swedish reactors show only minor mutual deviations and suggests a statistical uncertainty factor as low as 1.3. However, since the corresponding geometric mean differs from other assessments based on operational waste (0.1 by Thegerström /10/) and a reference BWR spent fuel assembly (0.2 by Kjellbert /9/) with an order of magnitude there seems to be reasons to increase the uncertainty factor at least to a factor of 5. Moreover, there is a considerable difference between the geometric mean based on ion-exchange resins from PWR and from BWR-reactors, 4.4 compared to 0.04, which may motivate the use of reactor-specific, or perhaps even waste type-specific correlation factors for this radionuclide.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>63</sup> Ni	<sup>60</sup> Co	0.2	0.2	0.03	2.0	97	1.3	5

### <sup>79</sup>Se

Data are scarce for Se. The available assessments are from a reference BWR spent fuel assembly /9/,  $4 \cdot 10^{-6}$ , and an evaluation based on Swiss reactor waste,  $8 \cdot 10^{-6}$  /8/. The uncertainty is therefore substantial and the maximum uncertainty factor for data from a reference BWR spent fuel assembly, 50, is chosen.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>79</sup> Se	<sup>137</sup> Cs	$4 \cdot 10^{-6}$	$4 \cdot 10^{-6}$	–	–	–	–	50

### <sup>90</sup>Sr

Cs and Sr should be produced at equal rate in fission reactions and a similar half-life (<sup>137</sup>Cs 30 years, <sup>90</sup>Sr 28 years) should lead to a correlation factor in the order of unity. The compilation of Thierfeldt et al. /4/ provides 37 data points from which such a low uncertainty factor as 1.5 can be derived. Nevertheless, the geometric mean for these values were almost a factor of 5 lower than from the assessments from calculations based on inventories in the pool water system (1 /5/) and data for a reference BWR spent fuel assembly (0.7 /9/). The uncertainty is therefore increased to 5.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>90</sup> Sr	<sup>137</sup> Cs	0.7	0.1	0.02	1.8	37	1.5	5

### <sup>93</sup>Zr

There are only two available sources for Zr; Information from contamination from US reference PWR /7/ which correlates to <sup>60</sup>Co with a value of  $1 \cdot 10^{-6}$  and an assessment based on a reference BWR spent fuel assembly /9/ which correlates to <sup>137</sup>Cs with a value of  $2 \cdot 10^{-5}$ . Although, the values are compatible, since the latter correlation roughly can be converted to  $1 \cdot 10^{-6}$  when correlating to <sup>60</sup>Co, the scarcity of data suggest a large uncertainty. The maximum uncertainty factor of 50 is therefore selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>93</sup> Zr	<sup>60</sup> Co	$2 \cdot 10^{-5}$	$1 \cdot 10^{-6}$	–	–	–	–	50

### <sup>93m</sup>Nb

Already the data from primary and secondary piping from the US nuclear piping /6/ yields a non-negligible statistical uncertainty factor of 5. Considering the deviation between the geometric mean of these values and the data from a reference BWR spent fuel assembly ( $2 \cdot 10^{-5}$  /9/) the uncertainty factor is increased even further to 20.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>93m</sup> Nb	<sup>60</sup> Co	$2 \cdot 10^{-5}$	$1 \cdot 10^{-3}$	$5 \cdot 10^{-3}$	2.9	7	5	20



**<sup>94</sup>Nb**

Data from a reference BWR spent fuel assembly /9/ give  $5 \cdot 10^{-5}$ , whereas measurements from US nuclear power station Shippingport /6/ suggest an interval of  $2 \cdot 10^{-5}$ – $2 \cdot 10^{-2}$  and a statistical uncertainty of 4. The close agreement between the geometric mean and the other assessments as well as those from induced activity suggests only a minor increase of the uncertainty to the values of 5.

<b>A</b>	<b>Key</b>	<b><i>c<sub>sfa</sub></i></b>	<b><i>c</i></b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b><i>n</i></b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>94</sup> Nb	<sup>60</sup> Co	$5 \cdot 10^{-5}$	$1 \cdot 10^{-5}$	$5 \cdot 10^{-5}$	2.6	7	4	5

**<sup>93</sup>Mo**

The only available data stems from a reference BWR spent fuel assembly /9/. The deviation from the value selected by an analogy with data for induced activity /8/ suggests a large uncertainty. The maximum uncertainty factor for a reference BWR spent fuel assembly data is therefore selected.

<b>A</b>	<b>Key</b>	<b><i>c<sub>sfa</sub></i></b>	<b><i>c</i></b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b><i>n</i></b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>93</sup> Mo	<sup>60</sup> Co	$8 \cdot 10^{-8}$	$5 \cdot 10^{-6}$	–	–	–	–	50

**<sup>99</sup>Tc**

Measurements from reactor coolant water from Swedish BWR /4/ are rather consistent, but predict a geometric mean lower than for instance a Swiss model inventory /8/. The low statistical uncertainty factor is therefore increased to 5.

<b>A</b>	<b>Key</b>	<b><i>c<sub>sfa</sub></i></b>	<b><i>c</i></b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b><i>n</i></b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>99</sup> Tc	<sup>137</sup> Cs	$1 \cdot 10^{-4}$	$5 \cdot 10^{-3}$	$2 \cdot 10^{-4}$	2.4	48	1.6	5

**<sup>107</sup>Pd**

Calculations based on a reference BWR spent fuel assembly give  $1 \cdot 10^{-6}$  /9/, which coincides with assessments from Swiss model inventory /8/. However since these are the only available values there is substantial uncertainty associated with these values. Nevertheless, the value from Swiss model inventory is an average and an uncertainty factor slightly below the maximum value of 50 is therefore selected.

<b>A</b>	<b>Key</b>	<b><i>c<sub>sfa</sub></i></b>	<b><i>c</i></b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b><i>n</i></b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>107</sup> Pd	<sup>137</sup> Cs	$1 \cdot 10^{-6}$	$1 \cdot 10^{-6}$	–	–	–	–	40

**<sup>108m</sup>Ag**

The only available sources are from a reference BWR spent fuel assembly /9/ and since the degree of correlation appears to differ depending on the choice of key nuclide, the uncertainties are large for this radionuclide. The maximum uncertainty factor of 50 is therefore selected.

<b>A</b>	<b>Key</b>	<b><i>c<sub>sfa</sub></i></b>	<b><i>c</i></b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b><i>n</i></b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>108m</sup> Ag	<sup>137</sup> Cs	$6 \cdot 10^{-5}$	$6 \cdot 10^{-5}$	–	–	–	–	50

**<sup>113m</sup>Cd**

Data are scarce, but there is an agreement between assessments based on Swiss model inventory /8/ and a reference BWR spent fuel assembly /9/. However, the Swiss data was based on a rather specific type and a maximum uncertainty factor of 50 is therefore selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>113m</sup> Cd	<sup>137</sup> Cs	5·10 <sup>-4</sup>	6·10 <sup>-4</sup>	–	–	–	–	50

**<sup>126</sup>Sn**

Data are again scarce. The agreement between an average correlation factor based on Swiss model inventory /8/ and the value suggested by data from a reference BWR spent fuel assembly /9/ slightly reduces the uncertainty below the maximum value.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>126</sup> Sn	<sup>137</sup> Cs	9·10 <sup>-6</sup>	5·10 <sup>-7</sup>	–	–	–	–	40

**<sup>125</sup>Sb**

Although some data for Sb appear to be contradictory and the assessments span over several orders of magnitude (0.01–4 /12/, /13/), the set of data employed for the statistical uncertainty factor, based on BWR spent fuel disassembly hardware from Shippingport /6/, show a variance, which suggests an uncertainty factor of 10.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>125</sup> Sb	<sup>60</sup> Co	–	0.1	0.5	4.1	10	6.4	10

**<sup>129</sup>I**

Already the variation within the values from the same source of data (Shippingport station, /6/) suggests large uncertainty with a statistical uncertainty factor of 15. Moreover, the recent study by Lundgren /19/ demonstrates differences in the order of magnitudes between the averages from BWR and PWR data (5·10<sup>-6</sup> and 3·10<sup>-5</sup>), partly explained by the activity transfer through the vapour in BWR. This is increased to 40 due to the moderate deviation from the selected value.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>129</sup> I	<sup>137</sup> Cs	3·10 <sup>-7</sup>	3·10 <sup>-7</sup>	6·10 <sup>-7</sup>	4.9	7	15	40

**<sup>134</sup>Cs**

The data from reactor coolant water /4/ spans the interval 0.06–14 with a standard deviation of the log-values around 1, which corresponds to a statistical uncertainty factor of 1.6. The selected value is higher than those proposed for US reference PWR /7/ and Swiss model inventory /8/ (0.4 and 0.02) as well as the value evaluated from a reference BWR spent fuel assembly /9/. However, the low variance within each of the data sets can be interpreted as systematic difference. The minor deviation from the selected value in conjunction with the low statistical uncertainty factor suggests a final uncertainty factor as low as 3.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>134</sup> Cs	<sup>137</sup> Cs	0.3	1	1.2	1	39	1.6	3

### <sup>135</sup>Cs

Unlike <sup>134</sup>Cs, there is no set of data sufficiently large to enable an evaluation of a statistical uncertainty factor. The available data are from various sources, but show a persuasive mutual agreement ( $5 \cdot 10^{-6}$  /10/,  $3 \cdot 10^{-6}$  /7/  $5 \cdot 10^{-6}$  /9/). Because of this mutual agreement an uncertainty factor of 40 is selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>135</sup> Cs	<sup>137</sup> Cs	$5 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	–	–	–	–	40

### <sup>133</sup>Ba

Relevant data for the contamination levels of <sup>133</sup>Ba are scarce and confined to data from a reference BWR spent fuel assembly /9/ and Swiss model inventory /8/, which suggests  $1 \cdot 10^{-6}$  and  $1 \cdot 10^{-5}$  respectively. Because of sparse information and their non-negligible deviation from each other the maximum uncertainty factor of 50 is selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>133</sup> Ba	<sup>60</sup> Co	$1 \cdot 10^{-5}$	$1 \cdot 10^{-5}$	–	–	–	–	50

### <sup>147</sup>Pm

Apart from data from a reference BWR spent fuel assembly only two additional sources were found; Swiss model inventory /8/ and US reference PWR /7/ which together span the substantial interval 0.02–1. Because of this deviation and the scarcity the maximum uncertainty factor is selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>147</sup> Pm	<sup>137</sup> Cs	0.9	0.9	–	–	–	–	50

### <sup>151</sup>Sm

As with <sup>147</sup>Pm, the available data are from a reference BWR spent fuel assembly /9/, Swiss model inventory /8/ and US reference PWR /7/ which results in a rather narrow interval  $2 \cdot 10^{-4}$ – $3 \cdot 10^{-3}$ . The scarcity of data still motivates a large uncertainty, but slightly lower than the maximum uncertainty. A factor of 40 is therefore selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>151</sup> Sm	<sup>137</sup> Cs	$3 \cdot 10^{-3}$	$3 \cdot 10^{-3}$	–	–	–	–	40

### <sup>152</sup>Eu

Except for data from a reference BWR spent fuel assembly /9/, the only available data for Eu consist of activity measurements on contamination solutions from cleaning of one of the Ågesta steam generators /1/. The maximum uncertainty factor is therefore selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>152</sup> Eu	<sup>137</sup> Cs	7·10 <sup>-5</sup>	7·10 <sup>-5</sup>	–	–	–	–	50

### <sup>154</sup>Eu

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>155</sup> Eu	<sup>137</sup> Cs	0.1	0.1	–	–	–	–	50

### <sup>155</sup>Eu

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>152</sup> Eu	<sup>137</sup> Cs	0.07	0.07	–	–	–	–	50

### <sup>166</sup>Ho

Assessment for <sup>166</sup>Ho is exclusively based on a reference BWR spent fuel assembly /9/ and as a consequence the maximum uncertainty factor is selected.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>166</sup> Ho	<sup>137</sup> Cs	4·10 <sup>-6</sup>	4·10 <sup>-6</sup>	–	–	–	–	50

## 3.3 Uncertainties for correlation factors to <sup>239,240</sup>Pu

Considering the availability of data, the transuranics do not differ in any aspect from other radionuclides regarding surface contamination. Hence, the same type of scheme and summarizing tables are employed when evaluating uncertainty factors.

### 3.3.1 Uncertainty factors

#### <sup>235</sup>U

The Swedish database in /4/ comprises 4 data points from which a statistical uncertainty factor of 2.5 can be evaluated. However, the geometric mean deviates with almost 2 orders of magnitude from the data evaluated from a reference BWR spent fuel assembly /9/, which suggests a rather dramatic increase of the uncertainty factor.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>235</sup> U	<sup>239,240</sup> Pu	2·10 <sup>-5</sup>	2·10 <sup>-5</sup>	2·10 <sup>-3</sup>	1.1	4	2.5	20

#### <sup>236</sup>U

Similar to <sup>235</sup>U, the Swedish database in /4/ only provides few data points and a statistical uncertainty factor in the order of 2.5, but unlike <sup>235</sup>U the geometric mean is in close agreement with the value based on a reference BWR spent fuel assembly.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>236</sup> U	<sup>239,240</sup> Pu	3·10 <sup>-4</sup>	3·10 <sup>-4</sup>	8·10 <sup>-4</sup>	0.8	3	2.6	10

**<sup>238</sup>U**

From the same database as for <sup>235</sup>U and <sup>236</sup>U a statistical uncertainty factor of 3.8 can be evaluated, which is increased further due to an order of magnitude of difference between the geometric mean and the value based on a reference BWR spent fuel assembly /9/.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>238</sup> U	<sup>239,240</sup> Pu	4·10 <sup>-4</sup>	4·10 <sup>-4</sup>	3·10 <sup>-3</sup>	1.9	5	3.8	10

**<sup>237</sup>Np**

The available data for <sup>237</sup>Np relates to 5 measured concentrations in reactor coolant from /4/ from which a statistical uncertainty factor of 3.2 can be evaluated. The difference between the geometric mean and the selected value based on a reference BWR spent fuel assembly /9/ is in an order of magnitude that, according to the scheme in Section 2.4, motivates an increase of the uncertainty to 10.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>237</sup> Np	<sup>239,240</sup> Pu	4·10 <sup>-4</sup>	4·10 <sup>-4</sup>	9·10 <sup>-3</sup>	1.7	5	3.2	10

**<sup>238</sup>Pu**

For this nuclide there exist at least 15 different series of measurements, each containing 1–80 values, which together span the huge interval of 0.03 to 3000. The data set from /4/ lies in the centre of this interval and qualifies as a good representative of the presumed waste in Swedish repositories. The statistical uncertainty factor ascertained from these data is 1.7. From the mere size of the total interval, as well as the scheme in Section 2.4, it appears reasonable to increase this value to 5, which then span at least 90% of all data employed in /1/.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>238</sup> Pu	<sup>239,240</sup> Pu	4	4	1.2	3.6	75	1.7	5

**<sup>241</sup>Pu**

There exist very few data on surface contamination of <sup>241</sup>Pu and assessments have to rely on data from a reference BWR spent fuel assembly /9/, reference PWR /7/ and the inventory of activated corrosion products /5/. The range of suggestions is rather limited, 100–167, but the number of data points too few to assign an uncertainty lower than the maximum i.e. 50.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>241</sup> Pu	<sup>239,240</sup> Pu	100	100	–	–	–	–	50

**<sup>241</sup>Am**

The compilation in /11/ employs 12 series of data, of which each contains 1–20 data points. The span of points is completely covered by the values given by /4/, which justifies the use of this limited subset in order to estimate the total uncertainty. A statistical uncertainty factor evaluated from these data is 3.9. A reasonable uncertainty factor based on this value and also reflecting the fact that the geometric mean of this subset differs somewhat from the selected value is therefore 10.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>241</sup> Am	<sup>239,240</sup> Pu	1	1	0.4	7	45	3.9	10

### <sup>243</sup>Am

The values in /4/ overlap all data given in the compilation /1/ and appear to qualify as a reasonable estimate for the whole range of data. A statistical uncertainty factor from this particular subset is 3.4, which considering the deviation of the geometric mean from the selected value should be increased to 10.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>243</sup> Am	<sup>239,240</sup> Pu	$3 \cdot 10^{-2}$	$3 \cdot 10^{-2}$	0.2	7	17	3.4	10

### <sup>243</sup>Cm

Except for the value based on a reference BWR spent fuel assembly /9/ all values employed in /1/ fall within the range of those employed in /4/. A statistical uncertainty factor evaluated from this subset is 1.2, but considering the deviation from the selected value, the proposed uncertainty factor is increased to 10. The remaining difference between the geometric mean and the selected value is interpreted as a systematic difference.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>243</sup> Cm	<sup>239,240</sup> Pu	$2 \cdot 10^{-2}$	$2 \cdot 10^{-2}$	1.5	0.6	15	1.2	10

### <sup>244</sup>Cm

A large number (16) of series of data are displayed in /1/, each of which contains 1–20 values. Except for an assessment based on scrape tests on fuel rods /11/ all data stays within the interval of the values from /4/ from which a statistical uncertainty factor of 1.9 can be evaluated. As with <sup>243</sup>Cm the difference between the geometric mean and the selected value is interpreted as being systematic. Thus, the uncertainty factor is assumed to be as low as 10.

<b>A</b>	<b>Key</b>	<b>c<sub>sfa</sub></b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>n</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<sup>244</sup> Cm	<sup>239,240</sup> Pu	3	3	0.05	1.2	7	1.9	10

## 4 Induced activity

Correlation factors for neutron activated material provide the same functionality as for surface contamination, i.e. shortcuts when estimating the present and predicting the future nuclear waste in repositories. A rough estimate of the relative activity compared to  $^{60}\text{Co}$  can be evaluated from the

- Decay rate compared to  $^{60}\text{Co}$ .
- Production cross section compared to  $^{60}\text{Co}$  (essentially the neutron capture cross section).
- The abundance compared to  $^{60}\text{Co}$ .

In principle this means that an adequate model needs to address following aspects;

- The composition of the steel i.e. the concentrations of the parent isotope.
- The quantities of major constituents.
- The quantities of impurities.
- The cross-section of the species.
- The duration of the irradiation.
- The neutron flux.

The majority of data stems from calculations and it is disputable if the codes can be applied to other materials and at other locations. The amount of impurities is unknown and the only available values are the limits guaranteed by the supplier. Unlike surface contamination the data is somewhat component-specific rather than reactor type-specific and several authors have assigned a particular uncertainty associated with calculations as such. Thus, when applying the same scheme as for surface contamination, an additional uncertainty is introduced inherent to the method. Moreover, because the data are less accessible than for surface contamination, only approximate confidence intervals evaluated from the figures presented in /1/ have been employed.

### 4.1 Uncertainties for correlation factors to $^{60}\text{Co}$

#### $^3\text{H}$

There is a wide span of assessments for correlation factors for  $^3\text{H}$ , ranging from  $10^{-8}$  /15/ to  $10^{-1}$  /10/, but the majority of values stay within the interval  $10^{-5} - 10^{-3}$ . Of the total 11 data points 8 values belong to Swiss model inventory and are based on the (unverified) assumption that the activity content is dominated by the contribution from induced activity. The statistical uncertainty is therefore increased to a factor of 20.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
$^3\text{H}$	$^{60}\text{Co}$	$1 \cdot 10^{-4}$	–	–	11	10	20

### **<sup>14</sup>C**

A calculation of induced activity in the core grid, the moderator tank and the core spray support in Swedish BWR suggests a value in the order of  $10^{-3}$  /17/. The centre of mass of the data points describing the correlation between <sup>14</sup>C and <sup>60</sup>Co is  $5 \cdot 10^{-4}$ . The corresponding interval that approximately captures 90% of the values is achieved by assigning a statistical uncertainty factor of 5. Due to the inherent uncertainty of calculated values, the uncertainty factor is increased to 10.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<b><sup>14</sup>C</b>	<sup>60</sup> Co	$5 \cdot 10^{-4}$	–	–	23	5	10

### **<sup>36</sup>Cl**

The majority of values belong to Swiss model inventory /8/ and are centred on  $5 \cdot 10^{-6}$  with a mean deviation about a factor 5. However, the most plausible value according to /1/ is  $7 \cdot 10^{-7}$  /16/ and the considerable deviation motivates a substantial increase of the uncertainty to a factor of 20.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<b><sup>36</sup>Cl</b>	<sup>60</sup> Co	$7 \cdot 10^{-7}$	–	–	13	5	20

### **<sup>55</sup>Fe**

The majority of assessment for correlation factors between <sup>55</sup>Fe and <sup>60</sup>Co are distributed in the interval 1–10 and hence is the correlation factor selected to 5 and the corresponding statistical uncertainty factor also to 5. The inherent uncertainty of calculations suggests a minor increase to 10.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<b><sup>55</sup>Fe</b>	<sup>60</sup> Co	5	–	–	27	5	10

### **<sup>59</sup>Ni**

The assessments for <sup>59</sup>Ni show almost identical order of spread as <sup>55</sup>Fe and therefore are the same values selected.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<b><sup>59</sup>Ni</b>	<sup>60</sup> Co	5	–	–	27	5	10

### **<sup>63</sup>Ni**

<sup>63</sup>Ni have been examined to the same degree as <sup>59</sup>Ni with similar result. An uncertainty factor of 10 is selected.

<b>A</b>	<b>Key</b>	<b>c</b>	<b><math>\bar{c}</math></b>	<b>S</b>	<b>N</b>	<b><math>\bar{\sigma}</math></b>	<b><math>\sigma</math></b>
<b><sup>63</sup>Ni</b>	<sup>60</sup> Co	1	–	–	27	5	10



### <sup>93m</sup>Nb

<sup>93m</sup>Nb is perhaps the least investigated radionuclide in this section. The available values ( $5 \cdot 10^{-7} / 15$ ,  $4 \cdot 10^{-4} / 8$ ,  $1 \cdot 10^{-3} / 8$  and  $1 \cdot 10^{-2} / 18$ ) span a large interval with a geometric mean of  $2 \cdot 10^{-4}$  and a statistical uncertainty factor of 27. Because of the scarcity of data, the huge statistical uncertainty factor and the uncertainty associated with calculations in general, an increase to 50 is suggested.

A	Key	c	$\bar{c}$	S	N	$\bar{\sigma}$	$\sigma$
<sup>93m</sup> Nb	<sup>60</sup> Co	$1 \cdot 10^{-3}$	–	–	4	27	50

### <sup>94</sup>Nb

The majority of correlation factors for <sup>94</sup>Nb are centred on  $1 \cdot 10^{-5}$ , but with a few values (3) exceeding  $1 \cdot 10^{-4}$ . An approximate interval embracing 90% of the data points can be ascertained from an uncertainty factor of 10. Due to the presumed uncertainty of calculations *per se*, this value is modified to 20.

A	Key	c	$\bar{c}$	S	N	$\bar{\sigma}$	$\sigma$
<sup>94</sup> Nb	<sup>60</sup> Co	$1 \cdot 10^{-5}$	–	–	16	10	20

### <sup>93</sup>Mo

The distribution of suggested correlation factors for <sup>93</sup>Mo forms an approximate interval of 90% confidence around  $5 \cdot 10^{-6}$  with statistical uncertainty factors around 6. In order to address the general uncertainty of calculations the uncertainty is modified to 10.

A	Key	c	$\bar{c}$	S	N	$\bar{\sigma}$	$\sigma$
<sup>93</sup> Mo	<sup>60</sup> Co	$5 \cdot 10^{-6}$	–	–	14	6	10

### <sup>99</sup>Tc

Although less abundant, <sup>99</sup>Tc share many characteristics of Fe and Ni and the uncertainty factor is selected in accordance with these radionuclides to 10.

A	Key	c	$\bar{c}$	S	N	$\bar{\sigma}$	$\sigma$
<sup>99</sup> Tc	<sup>60</sup> Co	$5 \cdot 10^{-7}$	–	–	18	5	10

## 5 Result

### 5.1 Surface contamination

A summary of the results derived in previous sections is presented in Tables 5-1 to 5-3.

**Table 5-1. Correlation factors to  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  for surface contamination.**

	<b>c</b>	<b><math>\sigma</math></b>	<b>Min</b>	<b>Max</b>
$^3\text{H}$	1.00E-04	50	2.00E-06	5.00E-03
$^{10}\text{Be}$	6.00E-10	50	1.20E-11	3.00E-08
$^{14}\text{C}$	1.00E-03	10	1.00E-04	1.00E-02
$^{36}\text{Cl}$	1.00E-05	20	5.00E-07	2.00E-04
$^{55}\text{Fe}$	7.00E-01	5	1.40E-01	3.50E+00
$^{59}\text{Ni}$	1.00E-03	3	3.33E-04	3.00E-03
$^{63}\text{Ni}$	0.2	5	4.00E-02	1.00E+00
$^{79}\text{Se}$	4.00E-06	50	8.00E-08	2.00E-04
$^{90}\text{Sr}$	0.1	5	2.00E-02	5.00E-01
$^{93}\text{Zr}$	1.00E-06	50	2.00E-08	5.00E-05
$^{93\text{m}}\text{Nb}$	1.00E-03	20	5.00E-07	2.00E-04
$^{94}\text{Nb}$	1.00E-05	5	2.00E-06	5.00E-05
$^{93}\text{Mo}$	5.00E-06	50	1.00E-07	2.50E-04
$^{99}\text{Tc}$	5.00E-03	5	1.00E-03	2.50E-02
$^{107}\text{Pd}$	1.00E-06	40	2.50E-08	4.00E-05
$^{108\text{m}}\text{Ag}$	6.00E-05	50	1.20E-06	3.00E-03
$^{113\text{m}}\text{Cd}$	6.00E-04	50	1.20E-05	3.00E-02
$^{126}\text{Sn}$	5.00E-07	40	1.25E-08	2.00E-05
$^{125}\text{Sb}$	0.1	10	1.00E-02	1.00E+00
$^{129}\text{I}$	3.00E-07	40	7.50E-09	1.20E-05
$^{134}\text{Cs}$	1	3	3.33E-01	3.00E+00
$^{135}\text{Cs}$	5.00E-06	40	1.25E-07	2.00E-04
$^{133}\text{Ba}$	1.00E-05	50	2.00E-07	5.00E-04
$^{147}\text{Pm}$	0.9	50	1.80E-02	4.50E+01
$^{151}\text{Sm}$	3.00E-03	40	7.50E-05	1.20E-01
$^{152}\text{Eu}$	7.00E-05	50	1.40E-06	3.50E-03
$^{155}\text{Eu}$	0.1	50	2.00E-03	5.00E+00
$^{152}\text{Eu}$	0.07	50	1.40E-03	3.50E+00
$^{166}\text{Ho}$	4.00E-06	50	8.00E-08	2.00E-04

## 5.2 Surface contamination; Transuranics

Table 5-2. Correlation factors to transuranics for surface contamination.

A	c	$\sigma$	Min	Max
<sup>235</sup> U	2.00E-05	20	1.00E-06	4.00E-04
<sup>236</sup> U	3.00E-04	10	3.00E-05	3.00E-03
<sup>238</sup> U	4.00E-04	10	4.00E-05	4.00E-03
<sup>237</sup> Np	4.00E-04	10	4.00E-05	4.00E-03
<sup>238</sup> Pu	4	5	8.00E-01	2.00E+01
<sup>241</sup> Pu	100	50	2.00E+00	5.00E+03
<sup>241</sup> Am	1	10	1.00E-01	1.00E+01
<sup>243</sup> Am	3.00E-02	10	3.00E-03	3.00E-01
<sup>243</sup> Cm	2.00E-02	10	4.00E-03	1.00E-01
<sup>244</sup> Cm	3	10	3.00E-01	3.00E+01

## 5.3 Induced activity

Table 5-3. Correlation factors to <sup>60</sup>Co for induced activity.

A	c	$\sigma$	Min	Max
<sup>3</sup> H	1.00E-04	20	5.00E-06	2.00E-03
<sup>14</sup> C	5.00E-04	10	5.00E-05	5.00E-03
<sup>36</sup> Cl	7.00E-07	20	3.50E-08	1.40E-05
<sup>55</sup> Fe	5	10	5.00E-01	5.00E+01
<sup>59</sup> Ni	5.00E-03	10	5.00E-04	5.00E-02
<sup>63</sup> Ni	1	10	1.00E-01	1.00E+01
<sup>93m</sup> Nb	1.00E-03	50	2.00E-05	5.00E-02
<sup>94</sup> Nb	1.00E-05	20	5.00E-07	2.00E-04
<sup>93</sup> Mo	5.00E-06	10	5.00E-07	5.00E-05
<sup>99</sup> Tc	5.00E-07	10	5.00E-08	5.00E-06

## 6 Comments and conclusions

The uncertainty associated with the correlation factors of nearly 40 radionuclides have been assessed by a combined statistical and qualitative method. The range of uncertainty factors is 5–50, where the highest uncertainty is related to assessments based exclusively on a single data point, in most cases on a reference BWR spent fuel assembly. Particularly low uncertainty is noted for the correlation factors for the radionuclides  $^{55}\text{Fe}$ ,  $^{59}\text{Ni}$ ,  $^{63}\text{Ni}$ ,  $^{90}\text{Sr}$ ,  $^{94}\text{Nb}$ ,  $^{99}\text{Tc}$ ,  $^{134}\text{Cs}$ ,  $^{237}\text{Np}$ ,  $^{238}\text{Pu}$ ,  $^{241}\text{Am}$ ,  $^{243}\text{Am}$  and  $^{243}\text{Cm}$ . An essential step towards higher accuracy would require new measurements and/or a reorientation from general correlation factors embracing all types of waste to more waste type-specific factors. Although the separation between wastes generated by surface contamination and induced activity represent an initial step in this direction, the substantial variations within the data seem to call for new categories. For instance, the differences in concentrations of  $^{63}\text{Ni}$  in ion-exchange resins between PWR and BWR-reactors are of the same order or larger than the differences between data from surface contamination and induced activity.

Despite the major difficulties as the scarcity and the non-uniform character of the data, the application of statistical methods when data is sufficient and the use of average and maximum deviations from the selected value for the particular waste stream whenever data are too scarce, enables to achieve intervals of at least 90% confidence. It is highly questionable whether these values can be improved much further without additional data.

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