R-07-05

Correlation factors for C-14, Cl-36, Ni-59, Ni-63, Mo-93, Tc-99, I-129 and Cs-135

In operational waste for SFR 1

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Keywords: 1089144, Correlation factor, SFR 1 SAR-08.

Preface

This document compiles information on correlation factors used to estimate the radionuclide inventory for SFR 1.

Maria Lindgren, Michael Pettersson and Marie Wiborgh, Kemakta AB have compiled the report.

This report has been reviewed and all comments have been documented.

Stockholm, November 2007

Agneta Innergård

Project manager, SFR 1 SAR-08

Summary

The use of correlation factors is one possibility to estimate a reference radionuclide inventory for the purpose of safety assessment studies, when waste-type specific information is not at hand. The correlation factor approach requires that there is a correlation between the activity content of the difficult-to-measure nuclide and the key nuclide.

A safety assessment of a future deep repository for low and intermediate level waste (SFL 3-5) was conducted during the second half of the 1990's. Within that project, correlation factors for estimating the inventory of radionuclides which are difficult to measure experimentally were developed. These factors have also partly been used to estimate a reference inventory for SFR 1.

Based on a literature survey and recent reports published by SKB, it is concluded that new information is available making an update of some correlation factors relevant. For these radionuclides, reported data from estimations and measurements of radionuclide content and correlation factors in different types of radioactive wastes are summarised. The data is evaluated and updated correlation factors representative for fresh waste for eight radionuclides (C-14, Cl-36, Ni-59, Ni-63, Mo-93, Tc-99, I-129 and Cs-135) are suggested. New information from measurements in Swedish NPP has made it possible to define factors that are BWR and PWR specific for C-14, Ni-59 and Ni-63.

The uncertainties in suggested data and the applicability of the correlation factors for estimates of the radionuclide content in operational waste are commented upon.

Sammanfattning

En uppskattning av aktivitetsinnehållet i avfallet krävs för att genomföra en säkerhetsanalys. När uppmätta data saknas är användandet av korrelationsfaktorer ett sätt att uppskatta ett referensinventar för radionuklidinnehållet. En förutsättning är att det går att hitta ett samband mellan den svårmätbara nukliden och nyckelnukliden.

En säkerhetsanalys av ett framtida djupförvar för låg- och medelaktivt avfall (SFL 3-5) genomfördes under den andra halvan av 1990-talet. Inom denna utredning togs korrelationsfaktorer fram i avsikt att uppskatta ett radionuklidinventar för svårmätbara radionuklider i det avfall som är tänkt att deponeras i detta förvar. Dessa faktorer har delvis även använts för att uppskatta ett referensinventar för SFR 1.

SKB har tagit fram rapporter med syfte att uppskatta inventariet av nuklider som tidigare har varit dåligt kartlagda men som säkerhetsanalyser har visat var av betydelse. Baserat på dessa rapporter och en litteraturundersökning genomförd inom föreliggande utredning, konstateras att det är motiverat att göra en förnyad bedömning av korrelationsfaktorn för vissa radionuklider. Rapporterade data, från mätningar eller modelleringar, avseende radionuklidinnehåll eller korrelationsfaktorer i olika avfallstyper har sammanställts. Dataunderlaget har utvärderats och för åtta nuklider (C-14, Cl-36, Ni-59, Ni-63, Mo-93, Tc-99, I-129 och Cs-135) ges förslag på uppdaterade korrelationsfaktorer som är representativa för nyproducerat avfall. Nya mätningar från svenska reaktorer har även gjort det möjligt att definiera faktorer som är specifika för BWR och PWR för C-14, Ni-59 och Ni-63.

Contents

1 Introduction

The use of correlation factors is one possibility to estimate a reference radionuclide inventory for the purpose of safety assessment studies, when waste-type specific information is not at hand. During the second half of the 1990's a preliminary safety assessment of a future deep repository for low and intermediate level waste (SFL 3-5) was conducted. Within that project, correlation factors for radionuclides which are difficult to measure experimentally were compiled /Lindgren et al. 1998/ and used for estimating a reference inventory for SFL3-5 waste.

The aim of the present project is to update the correlation factors for the waste to be deposited in SFR 1. SFL 3-5 is foreseen to contain waste with induced activity and waste that have been in contact with primary coolant water and thus contain surface contamination (Crud). The waste in SFR 1 on the other hand consists mainly of operational waste in the form of ion-exchange resins, and thus contain no or limited amount of induced activity. Surface contamination is more relevant than induced activity, but available data on ion-exchange resins is given priority when evaluating correlation factors. Transuranic elements and Sr-90 are not within the scope of this work. The content of these elements in operational waste for SFR 1 is determined by regular analyses on e.g. reactor coolant water and water in fuel pools and estimations of water flow through the ion-exchange resins /Johansson 1999/.

The update is based on a literature survey covering relevant literature published since the correlation factors where presented in 1998 /Lindgren et al. 1998/ and on the results of recent investigations performed by SKB for estimating the inventory of nuclides considered to be of specific importance for the safety assessment of SFR 1.

The correlation factors estimated in this report are not based on any measurements on waste from Clab or from Studsvik. The correlation factors should be used carefully when applied to this type of waste, especially for waste from Studsvik that origins from many different sources.

2 Information sources

A literature review has been made. The review has focused on data published since 1998. Earlier data is covered in /Lindgren et al. 1998/. Key words used in the literature search are compiled in Appendix 1. In addition, data in reports published recently by SKB regarding estimates of nuclide content in waste /Håkansson 2005, Lundgren 2005, Persson 2005, Lundgren 2006 and Magnusson and Stenström 2006/ as well as SSI /Ingemansson 2001/ have been utilized. Older data, presented in /Lindgren et al. 1998/, have been considered as well. A reprint of /Lindgren et al. 1998/ regarding the compilation of correlation factors used for SFL 3-5 reference inventory is given in its complete form in Appendix 2.

3 Correlation factors

With the results of the literature review and reports published later than 1998 at hand, it is concluded that new information is available for eight nuclides, C-14, Cl-36, Ni-59, Ni-63, Mo-93, Tc-99, I-129 and Cs-135. A revision of the correlation factor for these nuclides is therefore meaningful.

Reported estimations, measurements or modelled data of radionuclide content or correlation factors in different types of radioactive wastes are summarised for each nuclide below. Correlation factors from different studies are given and the data are compiled in diagrams. The data is evaluated and for each nuclide a correlation factor representative for one year old waste is suggested for use in updated estimates of the radionuclide inventory in SFR 1. Since the data represent different types of waste, data from different countries, calculated data, estimated data and measured data expert judgement have been used to value the most relevant data for the choice of correlation factor.

The main source for the activity in SFR 1 is ion-exchange resins from cleaning of the reactor coolant water. The update of the correlation factors is therefore based as much as possible on measurements/calculations for these ion-exchange resins.

For some nuclides a “correlation factor” is calculated based on information from two different references, a modelled total activity of the nuclide of interest and a measured total activity of the key nuclide. It is important to note that such a “correlation factor” is not a correlation factor in its real definition since it is not calculated from measurements on waste streams or from fuel composition estimates. In this report this type of “correlation factor” is denoted activity ratio.

The correlation factor is updated for eight nuclides in this report. However, a complete list of correlation factors for SFR 1 (except for transuranic elements and Sr-90) is given in Chapter 4. Nuclides for which the correlation factor has not been updated in this work, the reported correlation factor is that given by /Lindgren et al. 1998/ for low and intermediate level waste in SFL 3-5.

3.1 Inventory of key nuclides in SFR 1

The basis for a correlation factor is the content of the key nuclides Co-60 and Cs-137.
The gamma emitters Co-60 and Cs-137 are often readily measured with ordinary plant equipment and therefore used as key nuclides to which other, more difficult to measure, radionuclides can be correlated. Information about these two nuclides is summarised below.

3.1.1 Co-60

Co-60 is an activation product formed from Co-59 and Ni-60 /Thierfeldt and Deckert 1995/, and the source for Co-60 is cobalt as an impurity in nickel used in steel alloys. The half-life of Co-60 is 5.3 years.

A prognosis of the total amount of Co-60 in operational waste from the nuclear power plants that will be placed in SFR 1 was estimated through PROSIT which extracts data from the TRIUMF database. TRIUMF is a database containing measured activity on the waste packages delivered to SFR 1 as well as estimated activity for prognosticated waste packages. The amount of Co-60 in operational waste produced in the nuclear power plants until year 2005 is $1.6 \cdot 10^{14}$ Bq, which corresponds to $8.58 \cdot 10^{14}$ Bq if decay is not accounted for /Gordon 2006/.

A rough estimate of the amount of Co-60 belonging to waste from BWR and PWR respectively can be made from data on activity of Co-60 in all waste for SFR 1 presented in /Ingemansson 2001/. Based on these data it is assumed that 96% of the total Co-60 activity belongs to waste from BWR and 4% from PWR. Together with Gordon's data, this gives that the activity in waste produced until 2005 from BWR is $8.24 \cdot 10^{14}$ Bq and from PWR $3.43 \cdot 10^{13}$ Bq if decay is not accounted for.

The total amount of Co-60 in operational waste from the nuclear power plants that will be placed in SFR 1 until 2030 is $9.0 \cdot 10^{13}$ Bq, which corresponds to $1.52 \cdot 10^{15}$ Bq if decay is not accounted for /Gordon 2006/. Together with the assumption that 96% of the total Co-60 activity belongs to waste from BWR and 4% from PWR this gives the activity in waste year 2030 from BWR being $1.46 \cdot 10^{15}$ Bq and from PWR $6.07 \cdot 10^{13}$ Bq if decay is not accounted for.

During the last years some measures have been done to reduce the cobalt content in the reactor parts. For example removal of Stellite in the service valve in system 313 in Ringhals 1, exchange of valves covered with Stellite in the turbine system of Oskarshamn 3 and Forsmark 3. The effect of these measures might give reduced production of Co-60 /Ingemansson 2001/.

3.1.2 Cs-137

Cs-137 is a fission product from U-235. The production of Cs-137 goes through the noble gas isotope Xe-137, which has a significant influence on how Cs-137 is transported in the system /Lundgren 2005/. The half-life of Cs-137 is 30 years.

A prognosis of the total amount of Cs-137 in operational waste from the nuclear power plants that will be placed in SFR 1 was estimated through PROSIT which extracts data from the TRIUMF database. The amount of Cs-137 in operational waste produced in the nuclear power plants until year 2005 is $1.1 \cdot 10^{14}$ Bq, which corresponds to $1.73 \cdot 10^{14}$ Bq if decay is not accounted for /Gordon 2006/.

3.2 C-14

C-14	Selected correlation factor C-14/Co-60	
	$3 \cdot 10^{-3}$	BWR
Half-life = $5.7 \cdot 10^{4}$ years	$8 \cdot 10^{-2}$	PWR

C-14 is produced through fast neutron reactions with N-14 and O-17 in the coolant water and reactor components and by activation of oxygen in UO_2. It can also be produced by activation of C-13 (thermal neutrons) /Thierfeldt and Deckert 1995/. C-14 is also a fission product from the fuel. The main production of C-14 in reactor coolant in BWRs occurs through neutron activation of O-17, while nitrogen is negligible as source for C-14 /Lundgren and Ingemansson 2002/.

Most of the activity is released in gaseous form, but some of it will be present in the waste in insoluble form /Thierfeldt and Deckert 1995/. How much C-14 that get caught in the ion exchange resins and how much is released directly to the atmosphere varies between different references. /Ingemansson 2001/ reports that 80–99.9% of the C-14 production will be released to the atmosphere. Measurements of C-15 activity indicate that typically 40% of the production is transferred with the steam. That means that about 1% of the C-14 production in the reactor water will be captured in the cleaning system if the same partitioning between steam and water is assumed for C-14 at every core passage /Lundgren and Ingemansson 2002/.

The treatment of the ion exchange resins influence the content of C-14. If the resin is heated in connection with the solidification process CO_2 will be released to the atmosphere /Salonen and Snellman 1984/. Forsmark dries the resins in a temperature of 150°C before bitumenization of the resins, while all other Swedish power plant uses cement that do not need dry resins /Magnusson and Stenström 2006/. Hence, if the solidification is performed without heating, more carbon will remain in the resins.

It is clear that the chemical form of C-14 varies between BWR and PWR. Operational factors can also affect chemical form, especially for the PWR. The chemical speciation of C-14 compounds found in the ion exchange resins is directly correlated to the prevailing chemical conditions in the coolant, i.e. oxidizing or reducing. In BWRs, where oxidizing conditions prevail (i.e. NWC, normal water chemistry) oxidized forms such as carbon dioxide and bicarbonate (i.e. inorganic carbon compounds) are expected in the coolant /Lundgren and Ingemansson 2002/. PWRs are operated under reducing condition, resulting in the formation of mainly organic carbon species and carbon oxide /Magnusson and Stenström 2006/. The chemical conditions in BWRs operated with hydrogen injection (i.e. HWC, hydrogen water chemistry) will generally be closer to NWC operated BWRs rather than the PWRs. Therefore, inorganic carbon species are expected to be predominant also for HWC operated BWRs /Lundgren and Ingemansson 2002/.

In /Riggare and Johansson 2001/ it is made clear that the major C-14 sources are operational wastes from reactors, although Clab and Studsvik also produce non-trivial inventories of C-14.

Data from Sweden

Measurements of C-14 on spent ion exchange resins in various process water systems and ejector off-gas in both BWR-HWC, BWR-NWC and PWR in Sweden have been reported in /Magnusson and Stenström 2006/. Table 3-1 is a summary of calculated correlation factors as well as the organic fraction in the measured samples of ion exchange resins. The geometric mean of the measured correlation factors for BWR is $3 \cdot 10^{-3}$ and for PWR $8 \cdot 10^{-2}$. The heating used in Forsmark in connection with the solidification process implies that the C-14 will be released to the atmosphere and the correlation factor will be less. This is not included in the reported values from /Magnusson and Stenström 2006/ since the measurements are made on resins before solidification.

Table 3-1. Measured correlation factors and organic fraction in ion-exchange resins from Swedish reactors /from Magnusson and Stenström 2006/.

Reactor type	Reactor	System	Organic fraction (%)	C-14/ Co-60
BWR-HWC	B2	condensate cleaning	8	0.022
BWR-HWC	B2	condensate cleaning	16	0.039
BWR-HWC	O1+O2	spent fuel pool	–	< 2.6e–6
BWR-HWC	B2	reactor cleaning	6	7.3e–4
BWR-NWC	F1+F2+F3	reactor cleaning	9	5.4e–4
BWR-NWC	F1+F2+F3	condensate cleaning	4	0.1
BWR-NWC	F1+F2+F3	spent fuel pool	6	0.074
BWR-NWC	F1+F3	waste processing	5	6.5e–4
PWR	R3	several systems	29	0.92
PWR	R2	several systems	28	0.084
PWR	R4	several systems	35	0.023
PWR	R3	several systems	30	0.029

Based on measured production rates of C-14 and production rates of energy for BWR and PWR from Swedish and Finish power plants the total content of C-14 to be stored in SFR 1 has been calculated to be 277 GBq from BWR and 41 GBq from PWR /Ingemansson 2001/. The uncertainties are large and the result is compared with data from USA that indicate 30 times more C-14 in BWR and 125 times in PWR /Ingemansson 2001/. Activity ratios calculated based on the data from Swedish and Finish power plants and information about the Co-60 inventory in SFR 1 year 2030 without decay (see 3.1.1) are $2 \cdot 10^{-4}$ for BWR and $7 \cdot 10^{-4}$ for PWR.

The correlation between C-14 and Co-60 is reported in /Thegerström and Hård 1981/ to be 10^{-3} in Swedish operational waste. This value is based on the assumption that 1% of C-14 in the coolant water will be found in the operational waste.

The correlation factor between C-14 and Co-60 calculated from the activities in a reference BWR spent fuel assembly /Håkansson 2000/ is calculated to be $3 \cdot 10^{-4}$ one year after discharge.

Other information in /Magnusson and Stenström 2006/ makes it possible to calculate an activity ratio for all resins in SFR 1. Based on the measured correlation factors and calculated C-14 production, a prognosis of the total C-14 activity in spent resins from all reactors during 40 years was calculated to be 4.8 TBq of which 19% will be in organic form /Magnusson and Stenström 2006/. An activity ratio calculated based on this information and the prognosis of the Co-60 inventory in SFR 1 year 2030 without decay (see 3.1.1) is $3 \cdot 10^{-3}$.

Data from other countries

Correlation factors evaluated from the large database by /Thierfeldt and Deckert 1995/ that cover information from Germany, Japan and USA give a geometric mean of the correlation factor of $3 \cdot 10^{-3}$ for BWR and $1 \cdot 10^{-2}$ for PWR. The data show a wide distribution from 10^{-6} to 150 for BWR and $8 \cdot 10^{-5}$ to 5 for PWR.

/Robertson et al. 1993/ reported radionuclide concentrations, analysed from Crud from primary and secondary coolant piping from the US nuclear power station Shippingport (light-water breeder reactor core, typical of a commercial pressurised water reactor core). These data (5 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between C-14 and Co-60. For this report the correlation factors have been recalculated to be valid for one year old waste. This results in an interval of the correlation factor between $2 \cdot 10^{-8}$ to $3 \cdot 10^{-5}$, with a geometric mean of $2 \cdot 10^{-6}$.

Selection of correlation factors

The correlation of C-14 with Co-60 is doubted, for example in /Ingemansson 2001/, due to different behaviour of the nuclides in the nuclear power plant. Other authors, for example /Mitamura et al. 1992/, defend the correlation in PWR plants. Even though the use of Co-60 as key-nuclide for C-14 is doubted no other nuclide has been proved to be better and hence it is used in this report.

The correlation factor for C-14/Co-60 is 10^{-3} in /Lindgren et al. 1998/ based on data from Swedish operational waste reported in /Thegerström and Hård 1981/.

The correlation factor recommended in this report for C-14/Co-60 is $3 \cdot 10^{-3}$ for BWR. This value is based on the measurements on spent resins from Swedish power plants /Magnusson and Stenström 2006/. The correlation factor recommended for C-14/Co-60 is $8 \cdot 10^{-2}$ for PWR. This value is based on the measurements on spent resins from Swedish power plants /Magnusson and Stenström 2006/ and is supported by other data.

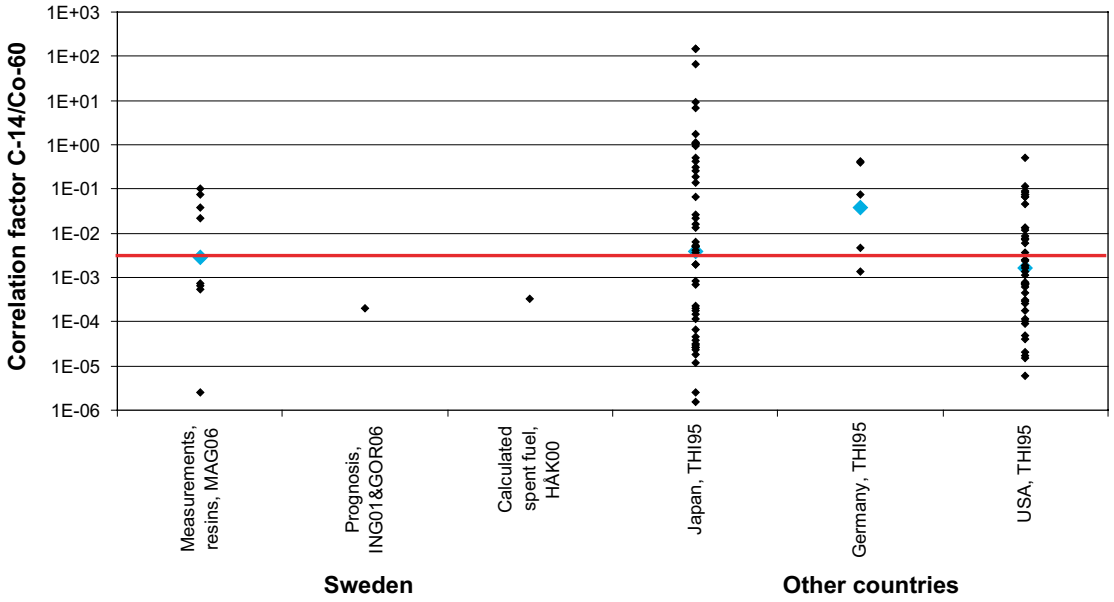

Figure 3-1. *Correlation factors for C-14/Co-60 for BWR. Black dots indicate measured or calculated data. Blue dots indicate geometric mean values, red line represents the selected correlation factor.*

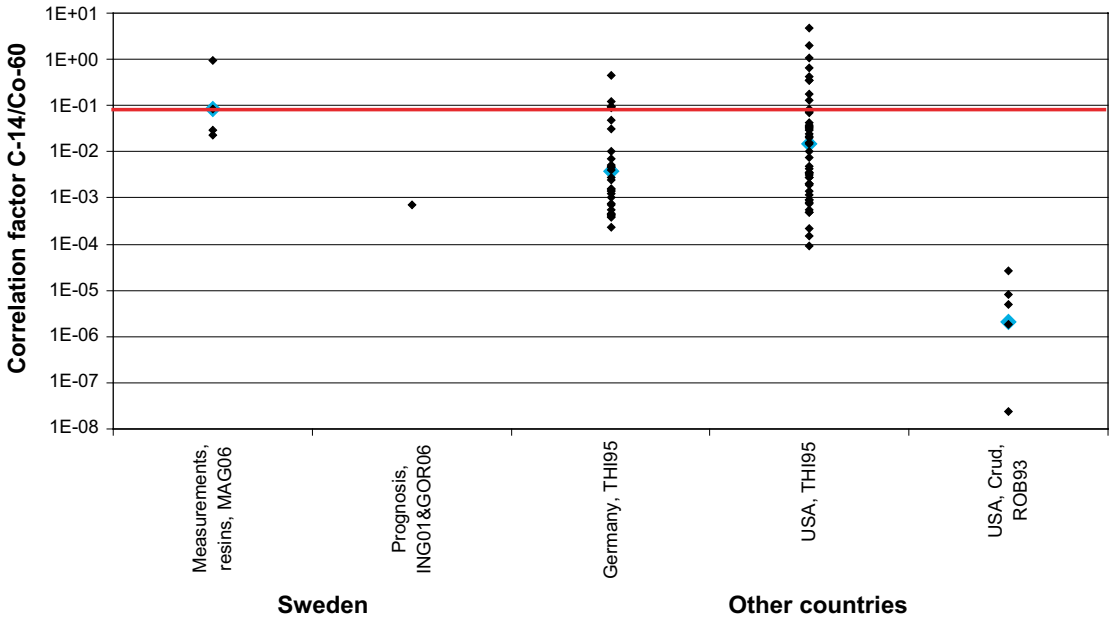

Figure 3-2. *Correlation factors for C-14/Co-60 for PWR.*

3.3 Cl-36

Cl-36	Selected correlation factor Cl-36/Co-60	
Half-life = $3.0 \cdot 10^5$ years	$6 \cdot 10^{-7}$	BWR and PWR

Cl-36 is an activation product almost exclusively produced from chloride in cooling water but also from chloride impurities in reactor construction materials /Robertson et al. 2000/, e.g. in Inconel /Thierfeldt and Deckert 1995/, stainless steel /NKA 1990/ and Zircaloy. Leakages from condensers and seawater salt concentration govern the amount of Cl-36 formed in a BWR

/Håkansson 2005/. Accordingly, there is a variation in chloride content between different reactors and also a variation over time. A source for Cl-36 in PWR is chloride present as an impurity in chemical additives /Håkansson 2005/.

In /Lindgren et al. 1998/ Cl-36 is correlated to Cs-137 which is a fission product. It has been commented by SKI and SSI /SKI/SSI 2001/ that it would be more appropriate to correlate to Co-60 which is an activation product just as Cl-36. In /Robertson et al. 2000/ correlation factors are presented with regard to both Co-60 and Cs-137. /Xinqi et al. 1991/ focuses on the correlation to Cs-137, but they present measurement data for Co-60 also. In the present report, correlations between Cl-36 and Co-60 as well as Cs-137 are discussed.

Data from Sweden

A model for estimating the activity of Cl-36 in operational waste from Swedish nuclear power plants has been developed /Håkansson 2005/. The model is based on reactor specific data on chloride content in reactor water. Using this model, the total amount of Cl-36 in operational waste from the nuclear power plants until year 2005 has been calculated to be $5.3 \cdot 10^{8}$ Bq /Håkansson 2005/. Waste produced at the nine BWRs in Sweden contribute with $4.3 \cdot 10^{8}$ Bq and the three PWRs with $9.7 \cdot 10^{7}$ Bq. The corresponding activity of Co-60, recalculated to fresh waste, has been estimated to $8.2 \cdot 10^{14}$ Bq (BWR) and $3.4 \cdot 10^{13}$ Bq (PWR), see section 3.1.1. Consequently, the activity ratio Cl-36/Co-60 for a BWR can be estimated to $5.2 \cdot 10^{-7}$ and for a PWR to $2.8 \cdot 10^{-6}$. A general activity ratio to be used for both BWR and PWR waste would be $6.2 \cdot 10^{-7}$.

The correlation between Cl-36 and Co-60, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Håkansson 2000/ is $3.6 \cdot 10^{-6}$ one year after fuel discharge. This is about the same as in older calculations performed by /Kjellbert 1990/ that was referred to in /Lindgren et al. 1998/.

The activity of Cs-137 in operational waste from Swedish nuclear power plants until 2005 (sum of BWR and PWR), recalculated to fresh waste, has been estimated to $1.7 \cdot 10^{14}$ Bq, see section 3.1.2. Together with total activity of Cl-36 ($5.3 \cdot 10^{8}$ Bq) from /Håkansson 2005/, this gives an activity ratio between Cl-36 and Cs-137 of $3.1 \cdot 10^{-6}$.

The correlation between Cl-36 and Cs-137, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Håkansson 2000/ is $1.2 \cdot 10^{-7}$ one year after fuel discharge.

Data from other countries

Measurements on resins used for reactor water clean-up in two different PWRs in USA /Robertson et al. 2000/ suggest a correlation factor between Cl-36 and Co-60 in five year old waste of $1.1 \cdot 10^{-5}$ and $5.3 \cdot 10^{-4}$, respectively. Recalculating this to one year old waste gives a correlation of $6.3 \cdot 10^{-6}$ and $3.1 \cdot 10^{-4}$, respectively. In the same study Cl-36 was also analysed in a resin from a BWR but could not be detected. An upper limit for the correlation factor can be estimated by assuming that the concentration of Cl-36 is the same as the detection limit. This gives a correlation factor for one year old waste of $2 \cdot 10^{-5}$ for the BWR resin. It is possible to calculate a correlation factor between Cl-36 and Cs-137 for one year old waste for one of the PWR resins. This gives a correlation factor of 10^{-5}. Again using the upper limit of Cl-36 in the BWR resin a factor between Cl-36 and Cs-137 for one year old waste is $2 \cdot 10^{-2}$.

The correlation factor between Cl-36 and Cs-137 and between Cl-36 and Co-60 has been evaluated based on measurements in reactor coolant water and resins from the reactor coolant cleanup system in Swiss nuclear power plants /Xinqi et al. 1991/. The correlation factor reported for a reactor coolant water sample from PWR and from resins in PWR and in BWR are summarised in Table 3-2 below.

Table 3-2. Correlation factors for water and resin from Swiss PWR and BWR /Xinqi et al. 1991/.

Waste	Cl-36/Co-60	Cl-36/Cs-137
PWR resin (A)	$5.7 \cdot 10^{-7}$	$1.4 \cdot 10^{-6}$
PWR resin (B)	$8.0 \cdot 10^{-6}$	$2.2 \cdot 10^{-6}$
BWR resin	$1.2 \cdot 10^{-4}$	$1.7 \cdot 10^{-5}$
Cooling water PWR (A)	$6.4 \cdot 10^{-4}$	$5.8 \cdot 10^{-5}$

German data on both BWR and PWR suggest much lower correlation factors; $2.8 \cdot 10^{-16}$ to $7.3 \cdot 10^{-9}$ when correlated to Co-60 and $1.9 \cdot 10^{-14}$ to $8.8 \cdot 10^{-8}$ when correlated to Cs-137 /Thierfeldt and Deckert 1995/.

Selection of correlation factors

In /Lindgren et al. 1998/ a correlation factor between Cl-36 and Cs-137 is selected to be $1 \cdot 10^{-5}$. This is based on Swiss data /Xinqi et al. 1991/.

The correlation factor between Cl-36 and Cs-137 is selected to be $3 \cdot 10^{-6}$, and between Cl-36 and Co-60 to be $6 \cdot 10^{-7}$. The selected correlation factors should be used for waste from both BWRs and PWRs. The selected correlation factor Cl-36/Cs-137 is three times lower than that given by /Lindgren et al. 1998/. The selected correlation factors are exclusively based on recently published data by /Håkansson 2005/ and /Gordon 2006/. This is motivated by the fact that these data are estimations of the activity of Co-60, Cs-137 and Cl-36 made especially for the waste produced in Swedish nuclear power plants, and are based on plant specific data. It is important to note the activity ratios given for Cl-36 are not correlation factors in its real definition since they are not calculated from activity measurements on waste streams or from fuel composition estimates. This is discussed in Chapter 5.

Since the source for Cl-36 is the chloride content in reactor water, the source for Co-60 is impurities in metal parts and the source for Cs-137 is the fuel there is no theoretical possibility to correlate Cl-36 to neither of Co-60 nor Cs-137. This is also supported by the large variation in reported correlation factors, see Figure 3-3 and Figure 3-4. However, to be able to treat data for all radionuclides in a similar way one correlation factor with Co-60 as key nuclide is selected ($6 \cdot 10^{-7}$). The choice of key nuclide is further discussed in section 5.2.

3.4 Ni-59

Ni-59	Selected correlation factor Ni-59/Co-60	
Half-life = $7.5 \cdot 10^{4}$ years	$1 \cdot 10^{-3}$	BWR
	$3 \cdot 10^{-2}$	PWR

Ni-59 is an activation product formed from Ni-58 by irradiation with thermal neutrons and from Ni-60 by irradiation with fast neutrons. Sources for Ni-59 are austenitic steel in the reactor and activation of nickel dissolved in the coolant and in corrosion particles deposited on the core /Thierfeldt and Deckert 1995/. The content of nickel in stainless steel is approximately 10% and in Inconel 50–75%. In addition, nickel is found as an impurity in Zircaloy (≈ 40 ppm) and in reactor fuel (≈ 20 ppm).

The steam generators, about 1,500 m^2, in a PWR consists of Inconel, which means that the PWR reactors are supplied with more nickel and that more Ni-59 will be formed than in a BWR /Ingemansson 2001/.

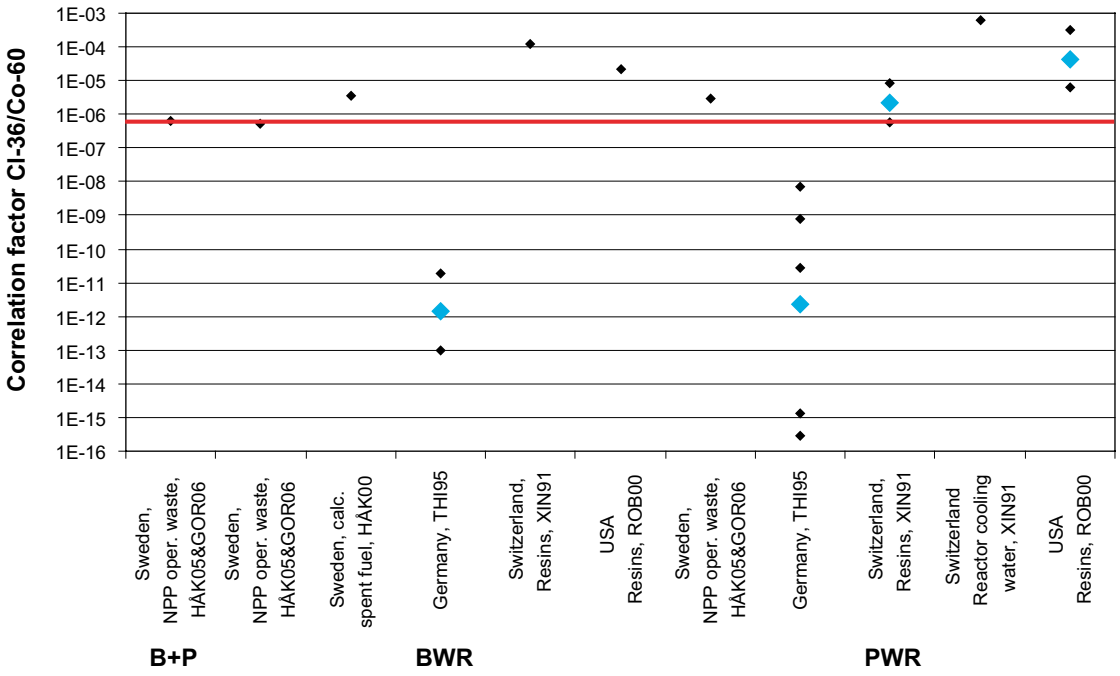

Figure 3-3. *Correlation factors for Cl-36/Co-60.*

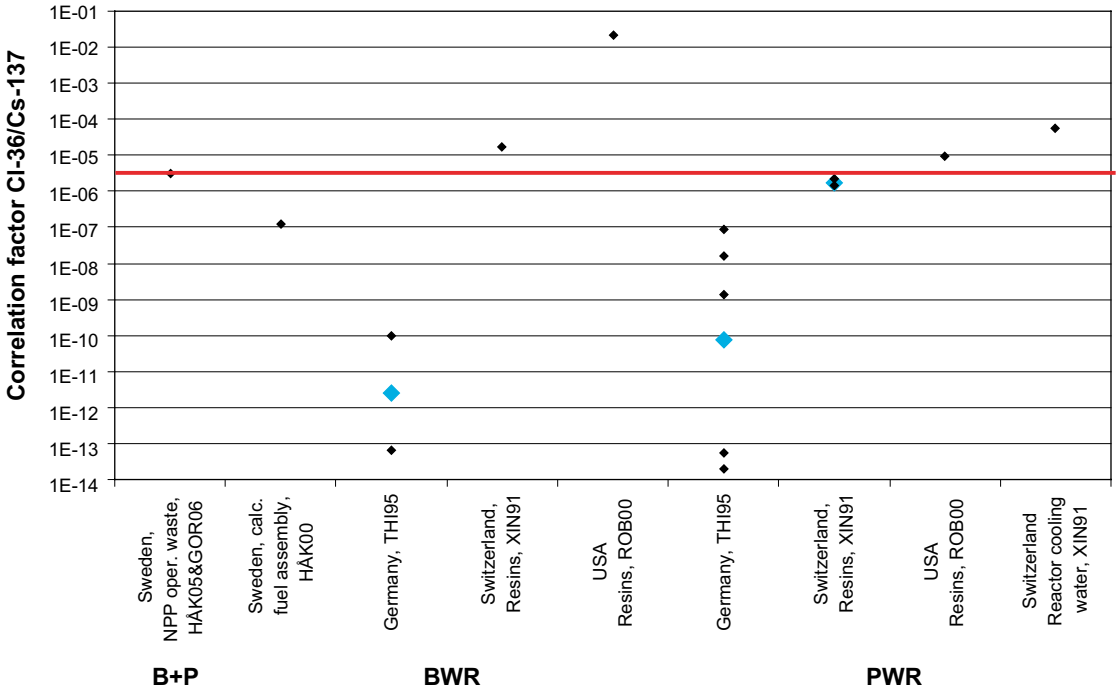

Figure 3-4. *Correlation factors for Cl-36/Cs-137.*

Data from Sweden

Measurements of Ni-59 as well as Co-60 in spent ion-exchange resins from Swedish BWRs and PWRs have been performed by /Persson 2005/. The results are given in Table 3-3. The geometric mean of the correlation factor for BWR is $1 \cdot 10^{-3}$ and 0.03 for PWR.

The content of Ni-59 in water from Swedish PWR reactors has been measured and reported /Roos 1994/ (the same data are included in the Swedish database of /Thierfeldt and Deckert 1995/).

Table 3-3. Measured correlation factors in spent ion-exchange resins from Swedish reactors /Persson 2005/.

Reactor type	Reactor	System	Ni-59/Co-60
BWR-HWC	B2	condensate cleaning	1.51e–3
BWR-HWC	B2	condensate cleaning	2.39e–3
BWR-HWC	O1+O2	spent fuel pool	7.33e–5
BWR-HWC	B2	reactor cleaning	1.54e–4
BWR-NWC	F1+F2+F3	reactor cleaning	1.42e–3
BWR-NWC	F1+F2+F3	condensate cleaning	6.29e–3
BWR-NWC	F1+F2+F3	spent fuel pool	3.21e–3
BWR-NWC	F1+F3	waste processing	7.84e–4
PWR	R3	several systems	0.018
PWR	R2	several systems	0.029
PWR	R4	several systems	0.063

These data were used to calculate correlation factors between Ni-59 and Co-60 (6 data sets). The correlation factors fall within the interval $4 \cdot 10^{-4}$ to $4 \cdot 10^{-3}$ with a geometric mean of $1 \cdot 10^{-3}$.

/Lundgren 1991/ has calculated the inventories of activated corrosion products in the pool water system and in the fuel pools in a BWR. The reported data were used to calculate a correlation factor between Ni-59 and Co-60 of $7 \cdot 10^{-4}$ one year after shutdown.

A correlation factor of 10^{-3} was evaluated based on information from Swedish operational waste /Thegerström and Hård 1981/.

The correlation between Ni-59 and Co-60, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Håkansson 2000/ is $5.8 \cdot 10^{-4}$ one year after fuel discharge.

An estimation of the total amount of Ni-59 produced in the Swedish BWR reactors during 40 years have been calculated based on a prognosis given in /Ingemansson 2001/ to be about $8 \cdot 10^{11}$ Bq and for the PWR reactors to be $7.2 \cdot 10^{12}$ Bq. The amount Ni-59 from BWR has been modeled with plant specific data, while the background for the estimated amount from PWR is not reported. Activity ratios calculated based on this information and prognosis of the Co-60 inventory in SFR 1 year 2030 without decay (see 3.1.1) are $5 \cdot 10^{-4}$ for BWR and 0.12 for PWR.

Data from other countries

Measurements on French operational waste from PWR reactors resulted in a correlation factor for Ni-59/Co-60 of $1 \cdot 10^{-4}$ to $1 \cdot 10^{-3}$ for filter cartridges based on four measurements /Raymond et al. 1996/.

The contamination level in an American reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. By using these data a correlation factor of 10^{-4} after five years of decay could be calculated (or $6 \cdot 10^{-5}$ in one year old waste).

Radionuclide concentrations in surface contamination analysed from primary and secondary coolant piping from the American nuclear power station Shippingport was reported by /Robertson et al. 1993/. These data (5 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Ni-59 and Co-60, and the factors vary within the interval $3 \cdot 10^{-3}$ to $7 \cdot 10^{-3}$, with a geometric mean of $5 \cdot 10^{-3}$. The correlation factors recalculated to one year old waste vary with in the interval $2 \cdot 10^{-3}$ to $4 \cdot 10^{-3}$, with a geometric mean of $3 \cdot 10^{-3}$.

The concentration of radionuclide on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR and PWR) in USA /Robertson et al. 1984/. The correlation factor calculated from the average activities reported from these measurements is $1 \cdot 10^{-3}$.

Selection of correlation factors

The correlation factor for Ni-59 for surface contamination in /Lindgren et al. 1998/ was calculated from data reported for Swedish waste and power units /see Roos 1994, Lundgren 1991, Thegerström and Hård 1981 above). The correlation factor was $1 \cdot 10^{-3}$ for both BWR and PWR.

Since available data indicate a difference in correlation factors for BWR and PWR reactors separate correlation factors are given.

Ion-exchange resin is the main source for the activity in SFR 1 waste. Therefore the latest investigation with measurements on ion-exchange resin /Persson 2005/ is seen as the main source for the choice of correlation factors.

The correlation factor recommended for BWR is $1 \cdot 10^{-3}$. This is the geometric mean of the measurements on ion-exchange resins from Swedish BWR /Persson 2005/. It is somewhat higher than the calculated values $5 \cdot 10^{-4}$ and $7 \cdot 10^{-4}$ calculated from data in /Ingemansson 2001, Lundgren 1991/, see Figure 3-5.

The correlation factor recommended for PWR is $3 \cdot 10^{-2}$. This is the geometric mean of the measurements on ion-exchange resins from Swedish PWR /Persson 2005/. It is higher than all measurements on reactor coolant in Swedish PWR reported in /Roos 1994/ and it is also higher than all data from other countries, see Figure 3-6. The recommended value is however less than the calculated correlation factor with estimated data from /Ingemansson 2001/. The quality of the estimate in /Ingemansson 2001/ is not possible to value from the information in the report.

3.5 Ni-63

Ni-63	Selected correlation factor Ni-63/Co-60	
	0.08	BWR
Half-life = 96 years	4	PWR

Ni-63 is an activation product formed from Ni-62 by irradiation with thermal neutrons and from Zn-66 and Cu-63 by fast neutrons. Sources for Ni-63 are austenitic steels in the core, activation of dissolved Ni-62 in the coolant and in corrosion particles in the core /Thierfeldt and Deckert 1995/.

The steam generators, about 1 500 m^2, in a PWR consists of Inconel, which means that the PWR reactors are supplied with more nickel and that more Ni-63 will be formed than in a BWR /Ingemansson 2001/.

/Ghysels 1993/ concludes that the correlation factor for Ni-63/Co-60 show a high level of uniformity between plants and waste streams except when design variability of the plants is too great. Examples of design variability include cobalt and nickel amounts in activated materials and the use of Stellite in wear bearings or valve facings.

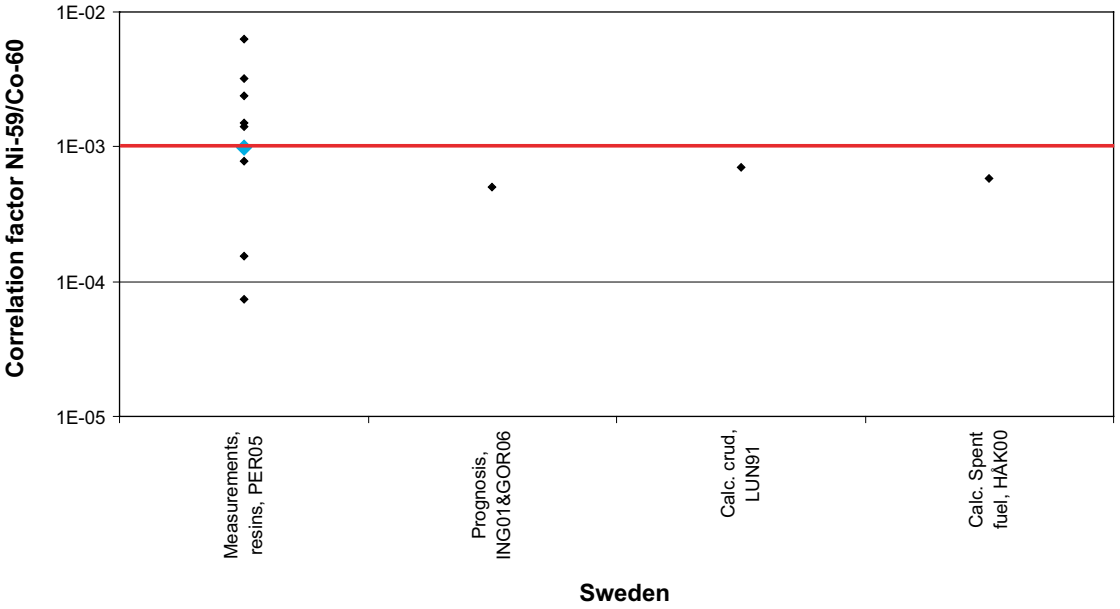

Figure 3-5. *Correlation factors for Ni-59/Co-60 for BWR.*

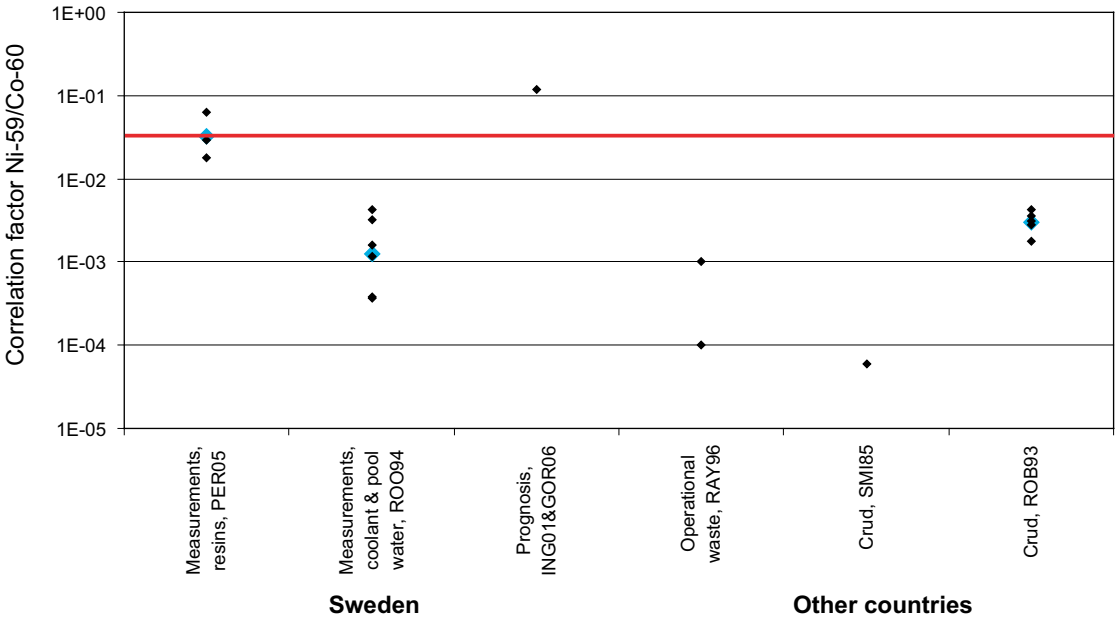

Figure 3-6. *Correlation factors for Ni-59/Co-60 for PWR.*

Data from Sweden

Measurements of Ni-63 as well as Co-60 in spent ion-exchange resins from Swedish BWRs and PWRs have been performed by /Persson 2005/. The results are given in Table 3-4. A difference in scaling factors can be seen between BWR and PWR, but also between BWR reactors that are hydrogen water chemistry operated with hydrogen injection (HWC) and BWR reactors with normal water chemistry (NWC). This is in contradiction to /James 2004/ who states that changing the plant condition by hydrogen injection have little impact on scaling factors. This is also shown in a figure in /James 2004/ where the correlation factors for Ni-63/Co-60 are constant during a twelve year period that include changed water chemistry from normal to hydrogen. In the study by /Persson 2005/ the geometric mean of the correlation factor for BWR is 0.08 and 4 for PWR.

Table 3-4. Measured correlation factors in spent ion-exchange resins from Swedish reactors /Persson 2005/.

Reactor type	Reactor	System	Ni-63/Co-60
BWR-HWC	B2	condensate cleaning	0.073
BWR-HWC	B2	condensate cleaning	0.075
BWR-HWC	O1+O2	spent fuel pool	0.0092
BWR-HWC	B2	reactor cleaning	0.015
BWR-NWC	F1+F2+F3	reactor cleaning	0.13
BWR-NWC	F1+F2+F3	condensate cleaning	0.22
BWR-NWC	F1+F2+F3	spent fuel pool	0.47
BWR-NWC	F1+F3	waste processing	0.18
PWR	R3	several systems	2.55
PWR	R2	several systems	5.66
PWR	R4	several systems	6.04

The content of Ni-63 and Co-60 was measured in reactor coolant water of Swedish BWRs /Roos 1994/ and from these data a correlation factor has been calculated for each of the 46 data sets, 34 for BWR and 12 for PWR. The calculated correlation factor for BWR falls within 0.01–0.5 with a geometric mean of 0.07 and for PWR within 0.3–5 with a geometric mean of 0.8. The Swedish database of /Thierfeldt and Deckert 1995/ comprises the same data as given in /Roos 1994/.

The inventory of activated corrosion products in pool water system and fuel pool systems in a BWR was calculated by /Lundgren 1991/. From these data a correlation factor between Ni-63 and Co-60 of 0.1 one year after shutdown could be calculated.

Data for Swedish operational waste were used to calculate a correlation factor of 0.1 /Thegerström and Hård 1981/.

The correlation factor between Ni-63 and Co-60 calculated from the activities in a reference BWR spent fuel assembly /Håkansson 2000/ is 0.08 one year after discharge. This is about the same as in older calculations performed by /Kjellbert 1990/ that was referred to in /Lindgren et al. 1998/.

A prognosis of the total amount of Ni-63 produced at the Swedish BWR reactors during 40 years have been calculated based on information given in /Ingemansson 2001/ to be about $1 \cdot 10^{14}$ Bq and for the PWR reactors to be $9.6 \cdot 10^{14}$ Bq. The amount of Ni-63 from BWR has been modeled with plant specific data, while the background for the estimated amount from PWR is not reported. Activity ratios calculated based on this information and the prognosis of the Co-60 inventory in SFR 1 year 2030 without decay (see 3.1.1) are 0.07 for BWR and 16 for PWR.

Data from other countries

Correlation factors evaluated from the large database by /Thierfeldt and Deckert 1995/ cover information from Japan, Germany, and USA. The geometric mean of the correlation factor for BWR is $3 \cdot 10^{-2}$, $1 \cdot 10^{-1}$ and $2 \cdot 10^{-2}$ for the data from the three different countries Japan, Germany, and USA. The corresponding geometric mean values are $3 \cdot 10^{-1}$, $2 \cdot 10^{-1}$ and $3 \cdot 10^{-1}$ for PWR.

/Kashiwagi et al. 2000/ show figures with measured data from Germany, Japan, France and USA on scaling factors for Ni-63/Co-60 that show that there are an appreciable difference between different reactor types (BWR and PWR), but no remarkable difference between different countries. The probability distribution is shown with a geometric mean value of the correlation factor for BWR of 0.04 and 0.5 for PWR.

In Belgium, a code (LLWAA –Low level waste activity assessment) is used to predict the inventories and/or scaling factors in waste generated by the operation of the nuclear power

plants. A calculated typical correlation factor for Ni-63/Co-60 for a mix of spent ion-exchange resin from the Tihange PWR reactors is 0.4 /Vandersperre et al. 2002/.

Measurements on French operational wastes from PWR reactors resulted in a correlation factor for Ni-63/Co-60 of 1.3 for ion-exchange resins /Raymond et al. 1994/.

The contamination level in an American reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. By using these data a correlation factor of 0.02 after five years of decay can be calculated. Recalculated to represent one year old waste the correlation factor is 0.01.

Radionuclide concentrations in surface contamination (Crud) analysed from primary and secondary coolant piping, from the US nuclear power station Shippingport was reported by /Robertson et al. 1993/. These data (5 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Ni-63 and Co-60, and the correlation factors vary within the interval $1 \cdot 10^{-3}$ to 0.9 with a geometric mean of 0.5. The correlation factors recalculated to one year old waste vary with in the interval $6 \cdot 10^{-4}$ to 0.5, with a geometric mean of 0.2.

The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR and PWR) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activities reported from these measurements was 0.1.

A correlation between the Ni-59 and Ni-63 is often used instead of a correlation to Co-60. Earlier measurements from USA reported by /Cline et al. 1980, 1985/ had significantly higher Ni-59/Ni-63 activity ratios (perhaps by as much as a factor of 10) compared to more recent values /Robertson et al. 2000/. More recent analyses and assessments indicate that this ratio should range between 0.006 and 0.012. The old data have large uncertainties due to both systematic and random analytical errors.

Selection of correlation factors

The correlation factor for Ni-63 for surface contamination selected in /Lindgren et al. 1998/ is 0.2 for both BWR and PWR reactors. The selected correlation factor was mainly based on measurement in reactor coolant, and was well supported by other data from Sweden and other countries.

Since available data indicate a difference in correlation factors for BWR and PWR reactors separate correlation factors are given.

Ion-exchange resin is the main source for the activity in SFR 1 waste. Therefore the latest investigation with measurements on ion-exchange resin /Persson 2005/ is seen as the main source for the choice of correlation factors.

The correlation factor recommended for BWR is 0.08. This is the geometric mean of the measurements on ion-exchange resins from Swedish BWR /Persson 2005/. It is somewhat higher than the geometric mean (0.07) from measurements on reactor coolant in Swedish BWR /Roos 1994/ and from the correlation factor based on estimates of the total inventory of Ni-63 and Co-60 in SFR 1 (0.07), see Figure 3-7.

The correlation factor recommended for PWR is 4. This is the geometric mean of the measurements on ion-exchange resins from Swedish PWR /Persson 2005/. It is higher than the geometric mean (0.8) from measurements on reactor coolant in Swedish PWR /Roos 1994/ and it is also high in comparison with most of the data from other countries, see Figure 3-8. The recommended value is however less than the calculated activity ratio with estimated data from /Ingemansson 2001/ and measured data from /Gordon 2006/. The quality of the estimate in /Ingemansson 2001/ is not possible to value from the information in the report.

The correlation between Ni-59 and Ni-63 calculated from the recommended correlation factors results in 0.012 for BWR and 0.007 for PWR. This is within the interval reported in /Robertson et al. 2000/.

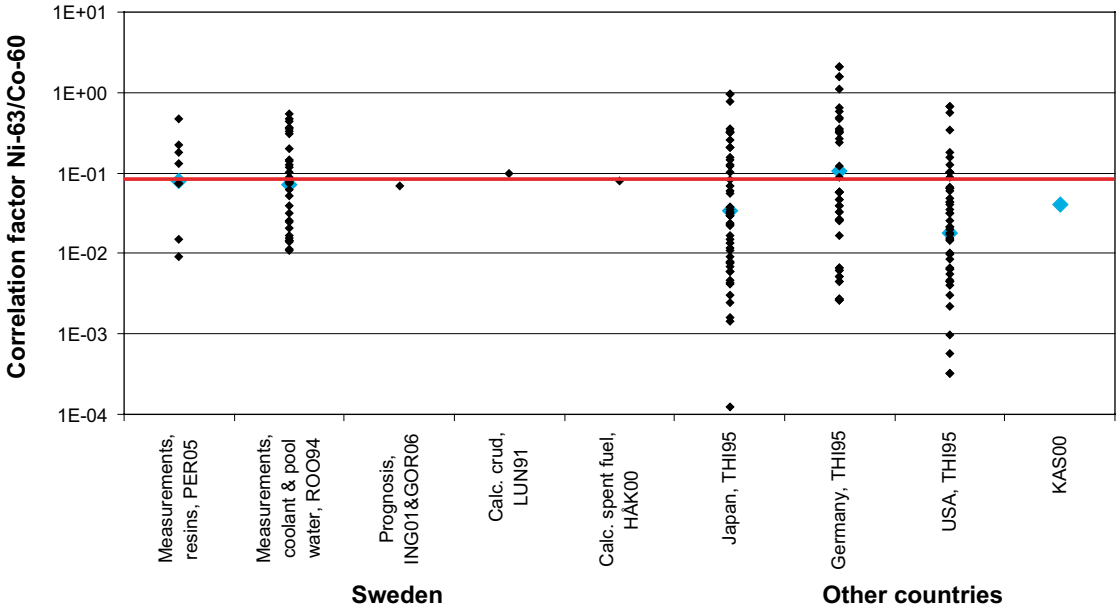

Figure 3-7. *Correlation factors for Ni-63/Co-60 for BWR.*

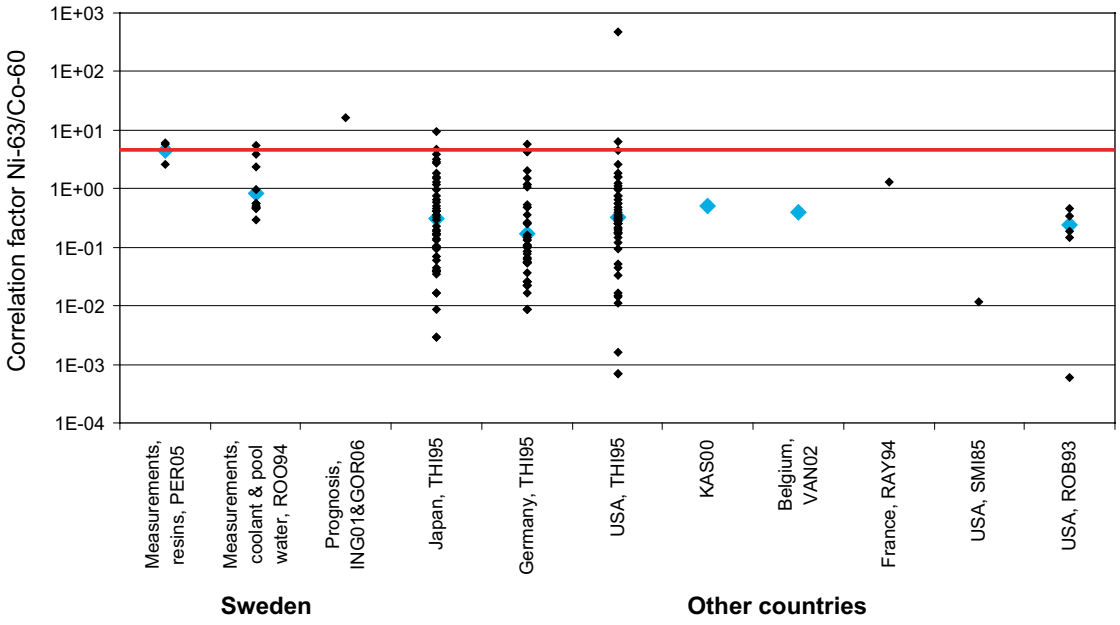

Figure 3-8. *Correlation factors for Ni-63/Co-60 for PWR.*

3.6 Mo-93

Mo-93	Selected correlation factor Mo-93/Co-60
Half-life = $3.5 \cdot 10^3$ years	$1 \cdot 10^{-6}$ BWR and PWR

Mo-93 is formed from Mo-92 by irradiation with thermal neutrons. In addition, formation from Ru-96 and Mo-94 by fast neutrons is possible, but to a lesser extent. Ruthenium and molybdenum are present as impurities in the fuel. Other sources for Mo-93 are metallic materials (steels and Inconel in the core region), and activation of molybdenum in dissolved form or as

corrosion particles in the coolant /Thierfeldt and Deckert 1995, Lundgren 2006/ states that due to the low solubility of Mo-isotopes in the coolant the main part of the Mo-93 formed in the Crud will remain in the crud and only to a less extent be present in the cleaning systems.

Data from Sweden

The activity of Mo-93 in operational waste from Swedish nuclear power plants has been estimated /Lundgren 2006/. The estimation is based on models for the activity inventory in BWR and PWR fuel, fuel leakage models and a model for estimating the amount of Mo-93 in resins in reactor water cleaning and in condensate cleaning. Based on these models the accumulated activity of Mo-93 in the resins from each of the twelve Swedish nuclear power plants is calculated. The total activity until 2005 of Mo-93 has been calculated to be $1.1 \cdot 10^{9}$ Bq. The corresponding measured activity of Co-60, recalculated to fresh waste, has been estimated to $8.6 \cdot 10^{14}$ Bq, see section 3.1.1. These two figures give an activity ratio of $1.3 \cdot 10^{-6}$.

The correlation between Mo-93 and Co-60 was calculated to be $4 \cdot 10^{-8}$ at fuel discharge and $8 \cdot 10^{-8}$ after a 5 years decay period from reported activities in a reference BWR spent fuel assembly /Kjellbert 1990/. New calculations of the activity in a BWR spent fuel assembly /Håkansson 2000/ was used to calculate a correlation factor of $2.9 \cdot 10^{-7}$ one year after fuel discharge.

Data from other countries

Calculated activities including both induced activity and contamination of Mo-93 are reported in the Swiss model inventory /Alder and McGinnes 1994/. The correlation factor between Mo-93 and Co-60 has here been evaluated for a selected number of Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) and the correlation factor vary within the interval 10^{-6} to 10^{-5}.

Measurements on BWR resin with a mixture of resins from reactor water clean up and liquid radwaste clean up resulted in a correlation factor of $1.25 \cdot 10^{-5}$ after 5 years decay /Robertson et al. 2000/, corresponding to $7.4 \cdot 10^{-6}$ after one year decay.

Measurements on a resin from reactor water clean up in a PWR gave a correlation factor of $3.7 \cdot 10^{-6}$ after 5 years decay /Robertson et al. 2000/, corresponding to $2.2 \cdot 10^{-6}$ after one year decay.

Selection of correlation factor

The correlation factor between Mo-93 and Co-60 selected in /Lindgren et al. 1998/ is $5 \cdot 10^{-6}$. It is stated that the selected correlation factor is not supported by any relevant measurements and therefore is very uncertain.

The correlation factor between Mo-93 and Co-60 selected in this report is $1 \cdot 10^{-6}$. This correlation factor should be used for waste from both BWRs and PWRs. The correlation factor is based exclusively on estimations of the Mo-93 activity /Lundgren 2006/ and of the Co-60 /Gordon 2006/ in operational waste from the power plants until 2005. This is motivated by the fact that these data are estimations of the activity of Mo-93 and Co-60 made especially for the waste produced in Swedish nuclear power plants, and are based on plant specific data. It is important to note that the activity ratio calculated for Mo-93 is not correlation factors in its real definition since it is not calculated from activity measurements on waste streams or from fuel composition estimates. This is discussed in Chapter 5.

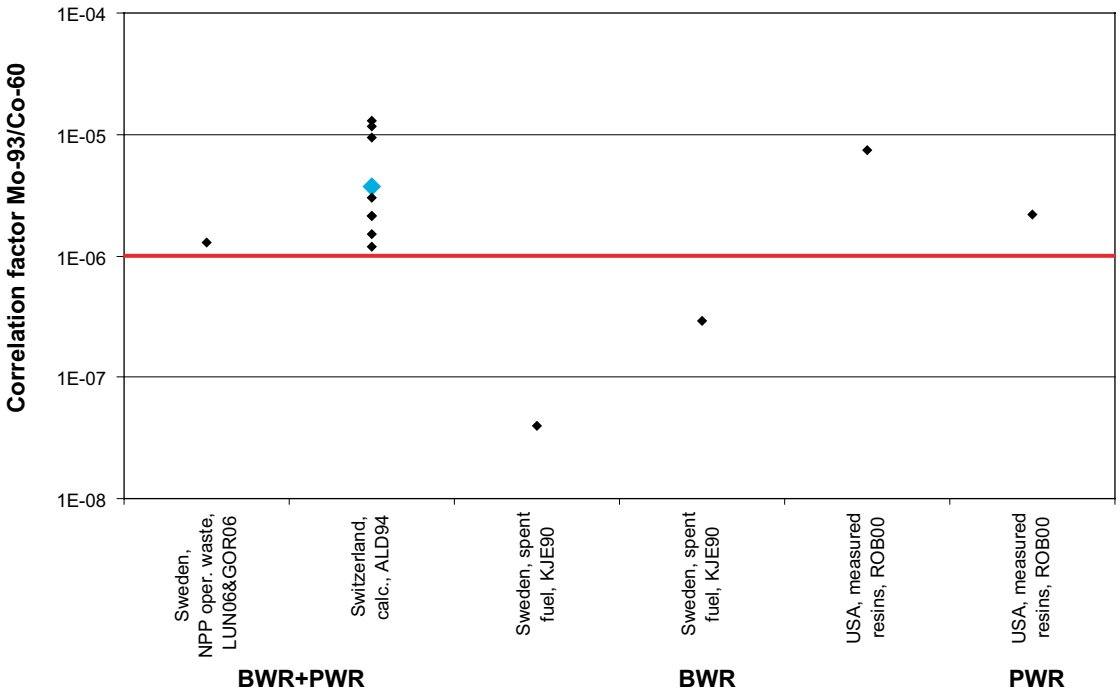

Figure 3-9. *Correlation factors for Mo-93/Co-60.*

3.7 Tc-99

Tc-99	Selected correlation factor Tc-99/Cs-137
Half-life = $2.1 \cdot 10^5$ years	$9 \cdot 10^{-4}$ BWR and PWR

Tc-99 is formed by irradiation of Mo-98 with thermal neutrons and as a fission product. Sources for Tc-99 are uranium fission in the fuel and irradiation of molybdenum as an alloy element in steels and Zircaloy /Thierfeldt and Deckert 1995/.

Data from Sweden

The content of Tc-99 was measured in reactor coolant water and pool water from Swedish BWRs and PWRs /Roos 1994/. Calculated correlation factors based on these measurements for BWRs (31 points of measuring) fall within an interval of $1 \cdot 10^{-4}$ and 1, with a geometric mean value of $4 \cdot 10^{-3}$. For PWRs (12 points of measuring) the correlation factors are in the interval $5 \cdot 10^{-5}$ to $1 \cdot 10^{-1}$, with a geometric mean value of $1 \cdot 10^{-3}$.

By using the Swedish database in /Thierfeldt and Deckert 1995/ to calculate correlation factors, an interval of $8 \cdot 10^{-7}$ and $5 \cdot 10^{-3}$ was obtained for BWR waste with a geometric mean value of $2 \cdot 10^{-4}$. Based on the same type of waste from PWR the correlation factor varies between $1 \cdot 10^{-4}$ and $1 \cdot 10^{-2}$, with a geometric mean value of $5 \cdot 10^{-4}$.

The activity of Tc-99 in operational waste from Swedish nuclear power plants has been estimated /Lundgren 2006/. The estimation is based on models for the activity inventory in BWR and PWR fuel, fuel leakage models and a model for estimating the amount of Tc-99 in resins in reactor water cleaning and in condensate cleaning. Based on these models the accumulated activity of Tc-99 in the resins from each of the twelve Swedish nuclear power plants is calculated and the total activity until 2005 of Tc-99 is calculated to be $1.3 \cdot 10^{10}$ Bq. The corresponding measured activity of Cs-137 recalculated to fresh waste, has been estimated to $1.7 \cdot 10^{14}$ Bq, see section 3.1.2. These two figures give an activity ratio of $7.4 \cdot 10^{-5}$.

The ratio between Tc-99 and Cs-137 in ion-exchange resins from BWR is $5 \cdot 10^{-5}$ /Thegerström and Hård 1981/.

The correlation factor between Tc-99 and Cs-137 calculated from the activities in a reference BWR spent fuel assembly /Håkansson 2000/ is $1.3 \cdot 10^{-4}$one year after discharge. This is about the same as in older calculations referred to in /Lindgren et al. 1998/.

Data from other countries

A variety of low-level radioactive waste samples from commercial nuclear power plants in USA (primary and secondary resins, primary coolant water, dry active waste, primary coolant purification filters etc) have been analysed for Tc-99 and Cs-137, and the results are summarized in /Robertson et al. 2000/. The geometric mean correlation factor between Tc-99 and Cs-137 based on measurements on primary resins in BWR and PWR is $3.2 \cdot 10^{-4}$ and $1.0 \cdot 10^{-7}$, respectively.

The contamination level in an American reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. These data were used to calculate a correlation factor of 10^{-4} after five years of decay, corresponding to $9 \cdot 10^{-5}$ for one year old waste.

The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR and PWR) in USA /Robertson et al. 1984/. The correlation factor between Tc-99 and Cs-137 calculated from the average activities reported from these measurements was $3 \cdot 10^{-3}$.

The average activity of Tc-99 in all reactor internals, reported in the Swiss model inventory /Alder and McGinnes 1994/, has been used to calculate an average correlation factor between Tc-99 and Cs-137 of $5 \cdot 10^{-3}$.

Selection of correlation factor

In /Lindgren et al. 1998/ the selected value of the correlation factor between Tc-99 and Cs-137 is $5 \cdot 10^{-3}$ based on Swedish measurements, but also well supported by both American data and Swiss data.

In this report, a correlation factor of $9 \cdot 10^{-4}$ is recommended. This is five times lower than the value used in /Lindgren et al. 1998/. The reduction is mainly based on Lundgrens new calculations performed for Tc-99 in operational waste from Swedish nuclear power plants. These calculations indicate an even lower correlation factor, but since most of the other investigations result in higher correlation factors (except for one study of PWR resins from USA) a more conservative value is chosen. The chosen value is the geometric mean value of all Swedish data reported.

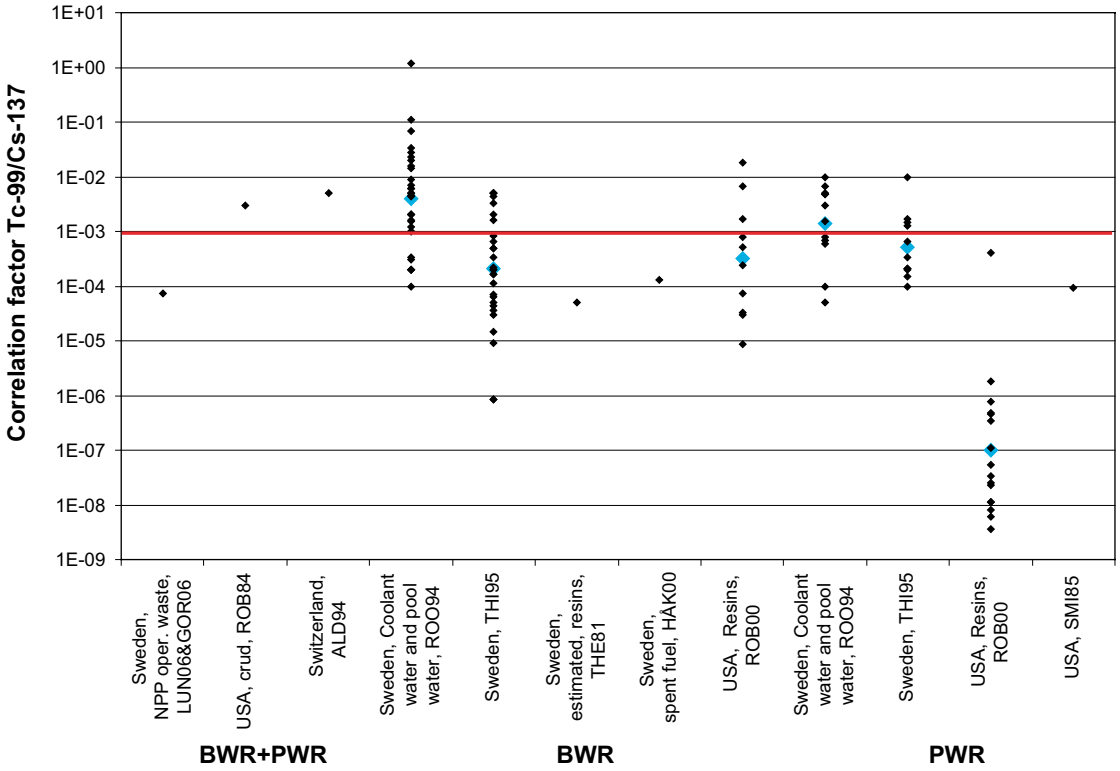

Figure 3-10. *Correlation factors for Tc-99/Cs-137.*

Data given in /Robertson et al. 2000/ indicate a large difference in correlation factor between BWR and PWR. /Roos 1994/ support this to some extent, but the difference is much smaller than suggested by Robertson. Swedish data compiled in /Thierfeldt and Deckert 1995/ on the other hand does not support this. It is not possible to evaluate separate correlation factors for BWR and PWR from the data presented in /Lundgren 2006/. The data set is judged to be too uncertain to recommend different correlation factors for BWR and PWR.

3.8 I-129

I-129	Selected correlation factor I-129/Cs-137	
Half-life = $1.6 \cdot 10^{7}$ years	$3 \cdot 10^{-6}$	BWR and PWR

I-129 is mainly a fission product from uranium. The source terms for I-129 are fuel elements and uranium material impurities /Thierfeldt and Deckert 1995/. The decay chain to I-129 contains no nuclide with high volatility /Lundgren 2005/, but the production of Cs-137, which I-129 is correlated to, goes via Xe-137 which is noble gas nuclide. Iodine and caesium have similar transport properties in reactor systems and release mechanisms from reactor fuel, but they have different retention properties in the filter systems. In addition, different correlation behaviour may exist for the ranges of low and high specific activities /Thierfeldt and Deckert 1995/.

Data from Sweden

The activity of I-129 in operational waste from Swedish nuclear power plants has been estimated /Lundgren 2005/. The estimation is based on models for the activity inventory in BWR and PWR fuel, fuel leakage models and a model for estimating the amount of I-129 in resins in reactor water cleaning and in condensate cleaning. Based on these models the accumulated activity of I-129 in the resins from each of the twelve Swedish nuclear power plants is

calculated and the total activity until 2005 of I-129 has been calculated to be $4.8 \cdot 10^{8}$ Bq. The corresponding activity of Cs-137, recalculated to fresh waste, has been measured to $1.7 \cdot 10^{14}$ Bq, see section 3.1.2. These two figures give an activity ratio of $2.8 \cdot 10^{-6}$.

The correlation factor in ion-exchange resins from BWR reported by /Thegerström and Hård 1981/ is $3 \cdot 10^{-7}$. The activities of I-129 and Cs-137 were reported for core components and internal parts in /Thegerström and Hård 1981/. From these data a correlation factor of $4 \cdot 10^{-7}$ could be calculated.

The correlation factor between I-129 and Cs-137 calculated from the activities in a reference BWR spent fuel assembly /Håkansson 2000/ is $3 \cdot 10^{-7}$one year after discharge. This is about the same as in older calculations by /Kjellbert 1990/ referred to in /Lindgren et al. 1998/.

Data from other countries

Based on a compilation of data on different types of waste from BWR in Germany and PWR and BWR in Japan and USA /Thierfeldt and Deckert 1995/ it can be concluded that there is a very large variation in correlation factor between different wastes. The estimated correlation factor I-129/Cs-137 varies between 10^{-8} and 10. /Thierfeldt and Deckert 1995/ conclude that a correlation factor between I-129 and Cs-137 is plant specific.

A variety of samples from commercial nuclear power plants in USA (primary and secondary resins, primary coolant water, dry active waste, primary coolant purification filters etc) have been analysed for I-129 and Cs-137, and the results are summarized in /Robertson et al. 2000/. It is concluded that the correlation between I-129 and Cs-137 is quite good, even though diverse waste types with radionuclide concentrations ranging over eight orders of magnitude were studied. The correlation was much better for PWR waste than for BWR waste. This is contributed to the fact that much of the iodine can be transported by steam to the condensate resin in BWR reactors while the caesium is expected to be removed by the primary demineralization system. The geometric mean correlation factor between I-129 and Cs-137 based on measurements on primary resins in BWR and PWR is $4.6 \cdot 10^{-7}$ and $5.2 \cdot 10^{-8}$, respectively. The corresponding values based on different types of waste, other than primary resins, are $3.7 \cdot 10^{-7}$ (BWR) and $8.7 \cdot 10^{-8}$ (PWR), respectively.

The average activity of I-129 in all reactor internals that was reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate an average correlation factor between I-129 and Cs-137 of $2 \cdot 10^{-4}$.

The concentration of radionuclides on piping exposed to primary reactor coolant has been measured at seven nuclear power stations (BWR and PWR) in USA /Robertson et al. 1984/. The correlation factor calculated from the average activity reported from these measurements was $3 \cdot 10^{-5}$.

Selection of correlation factor

The correlation factor between I-129 and Cs-137 selected in /Lindgren et al. 1998/ is $3 \cdot 10^{-7}$. This is mainly based on data from the spent fuel assembly, but also supported by old data used for Swedish radioactive waste /Thegerström and Hård 1981/.

The correlation factor between I-129 and Cs-137 selected in this report is $3 \cdot 10^{-6}$. This correlation factor should be used for waste from both BWRs and PWRs. The correlation factor is based exclusively on the calculated activity ratio from the calculated activity of I-129 /Lundgren 2005/ and of the measured Cs-137 activity /Gordon 2006/ in operational waste from nuclear power plants. This is motivated by the fact that these data are estimations of the activity of Cs-137 and I-129 made especially for the waste produced in Swedish nuclear power plants, and are based on plant specific data. It is important to note that the activity ratio given for I-129 is not correlation factors in its real definition since it is not calculated from activity measurements of iodine on waste streams or from fuel composition estimates. This is discussed in Chapter 5.

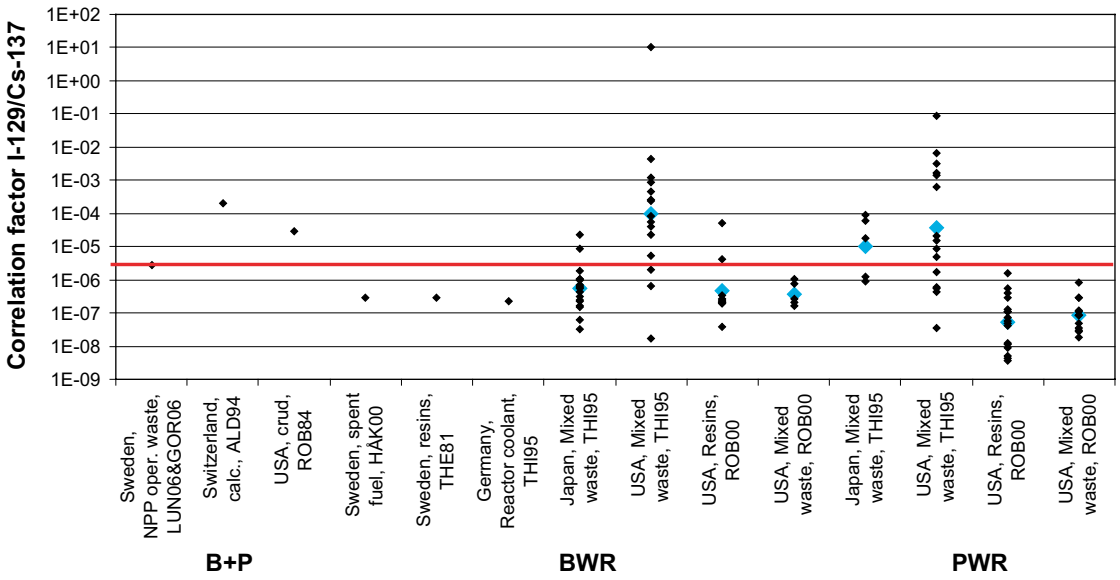

Figure 3-11. *Correlation factors for I-129/Cs-137.*

Even though the calculated correlation factors given in the review above indicate that there is a difference in correlation factor between BWR and PWR of about one order of magnitude, it should be noted that there is a large variation between different samples, even though only analyses on primary resins are considered. The large variation in correlation factors is especially shown by the data in /Thierfeldt and Deckert 1995/. The data does not support that there should be a difference in correlation factor between a BWR and a PWR.

3.9 Cs-135

Cs-135	Selected correlation factor Cs-135/Cs-137	
Half-life = $2.3 \cdot 10^6$ years	$1 \cdot 10^{-5}$	BWR and PWR

Cs-135 is a fission product from uranium. Cs-135 is found in spent fuel elements and in materials containing uranium as impurities /Thierfeldt and Deckert 1995/. Cs-135 and Cs-137 are both single neutron fission products and a simple linear correlation between them should be expected /Thierfeldt and Deckert 1995/.

Data from Sweden

The activity of Cs-135 in operational waste from Swedish nuclear power plants has been estimated /Lundgren 2006/. The estimation is based on models for the activity inventory in BWR and PWR fuel, fuel leakage models and a model for estimating the amount of Cs-135 in resins in reactor water cleaning and in condensate cleaning. Based on these models the accumulated activity of Cs-135 in the resins, from each of the twelve Swedish nuclear power plants is calculated and the activity until 2005 of Cs-135 has been calculated to be $2.2 \cdot 10^9$ Bq. The corresponding measured activity of Cs-137, recalculated to fresh waste, has been estimated to $1.7 \cdot 10^{14}$ Bq, see section 3.1.2. These two figures give an activity ratio of $1.3 \cdot 10^{-5}$.

/Thegerström and Hård 1981/ report a correlation factor of $3 \cdot 10^{-6}$ in ion-exchange resins from BWR.

The correlation between Cs-135 and Cs-137 was calculated to be $4.6 \cdot 10^{-6}$ one year after discharge from reported activities in a reference BWR spent fuel assembly /Håkansson 2000/. This is about the same as in older calculations by /Kjellbert 1990/ referred to in /Lindgren et al. 1998/.

A theoretical value for the correlation between Cs-135 and Cs-137 has been reported to be $1 \cdot 10^{-5}$ by /Roos 1994/.

Data from other countries

/Thierfeldt and Deckert 1995/ reported some measurements (number of 5) from which a geometric mean value of $6 \cdot 10^{-6}$ of the correlation factors could be calculated.

Evaluation of contamination data from an American reference PWR /Smith et al. 1985/ gave a correlation factor of $3 \cdot 10^{-6}$ after five years of decay, corresponding to $2.7 \cdot 10^{-6}$ in one year old waste.

The average activity of Cs-135 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate the average correlation factor between Cs-135 and Cs-137 of $7 \cdot 10^{-6}$.

Selection of correlation factor

The correlation factor between Cs-135 and Cs-137 selected in /Lindgren et al. 1998/ is $5 \cdot 10^{-6}$. The correlation factor between Cs-135 and Cs-137 selected in this report is $1 \cdot 10^{-5}$. This correlation factor should be used for waste from both BWRs and PWRs. The correlation factor is based exclusively on estimations of the Cs-135 activity /Lundgren 2006/ and the Cs-137 activity /Gordon 2006/ in operational waste from the power plants until 2005. This is motivated by the fact that these data are estimations of the activity of Cs-135 and Cs-137 made especially for the waste produced in Swedish nuclear power plants, and are based on plant specific data. It is important to note that the activity ratios given for Cs-135 is not correlation factors in its real definition since it is not calculated from activity measurements on waste streams or from fuel composition estimates. This is discussed in Chapter 5.

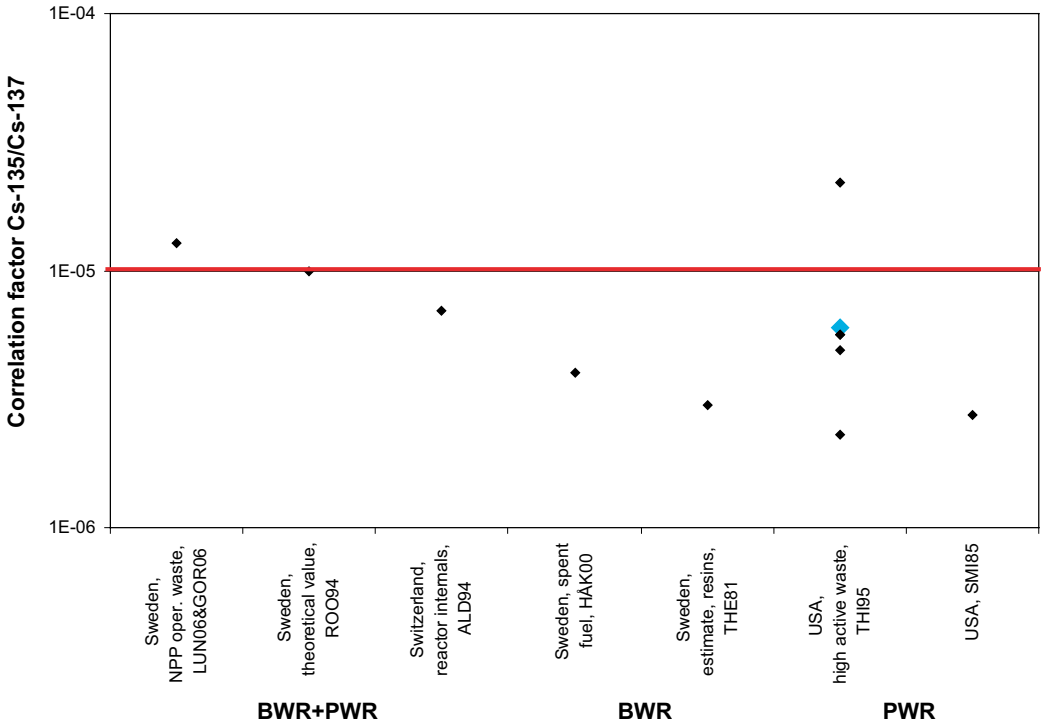

Figure 3-12. *Correlation factors for Cs-135/Cs-137.*

4 Compilation of correlation factors

As shown in this report, newer data making an update of the correlation factors previously suggested for a repository for low and intermediate level waste (SFL 3-5) possible are available for a few nuclides only. Due to lack of data the correlation factors used for estimating the radionuclide inventory for SFR 1 is therefore to a large extent the same as those used for SFL 3-5. The updated correlation factors for C-14, Cl-36, Ni-59, Ni-63, Tc-99, Mo-93, I-129 and Cs-135 are compiled in Table 4-1 together with the correlation factors for surface contamination developed for SFL 3-5 for the nuclides not updated in the present study.

Table 4-1. Correlation factors for SFR 1. The correlation factors are aimed to be representative for waste one year after discharge from the nuclear facility. The bold correlation factors are updated based on new information whereas the other factors are from a previous study for similar waste.

Radionuclide	Half-life (year)	Correlation factor	Comment	Source for selected correlation factor
H-3 *	12	$1 \cdot 10^{-4}$	LIN 98	Data on induced activity in metals
Be-10 *	$1.6 \cdot 10^{6}$	$6 \cdot 10^{-10}$	LIN 98	Data on BWR spent fuel assembly
C-14 * (BWR)	$5.7 \cdot 10^{3}$	$3 \cdot 10^{-3}$	Updated	Measurements of C-14 and Co-60 n Swedish ion-exchange resins
C-14 * (PWR)	$5.7 \cdot 10^{3}$	$8 \cdot 10^{-2}$	Updated	Measurements of C-14 and Co-60 in Swedish ion-exchange resins
Cl-36 *	$3.0 \cdot 10^{5}$	$6 \cdot 10^{-7}$	Updated	Activity ratio – Modelled activity of Cl-36 in resins from Swedish NPP and measured activity of key nuclide in operational waste
Fe-55 *	2.7	1	LIN 98	Data on Swedish operational waste
Ni-59 * (BWR)	$7.5 \cdot 10^{4}$	$1 \cdot 10^{-3}$	Updated	Measurements of Ni-59 and Co-60 in Swedish ion-exchange resins
Ni-59 * (PWR)	$7.5 \cdot 10^{4}$	$3 \cdot 10^{-2}$	Updated	Measurements of Ni-59 and Co-60 in Swedish ion-exchange resins
Ni-63 * (BWR)	96	$8 \cdot 10^{-2}$	Updated	Measurements of Ni-63 and Co-60 in Swedish ion-exchange resins
Ni-63 * (PWR)	96	4	Updated	Measurements of Ni-63 and Co-60 in Swedish ion-exchange resins
Se-79 **	$6.5 \cdot 10^{4}$	$4 \cdot 10^{-6}$	LIN 98	Data on Swedish BWR spent fuel assembly
Mo-93 *	$3.5 \cdot 10^{3}$	$1 \cdot 10^{-6}$	Updated	Activity ratio – Modelled activity of Mo-93 in resins from Swedish NPP and measured activity Co-60 in operational waste
Zr-93 *	$1.5 \cdot 10^{6}$	$1 \cdot 10^{-6}$	LIN 98	Data on American PWR spent fuel assembly
Nb-93m *	14	$1 \cdot 10^{-3}$	LIN 98	Correlation to Nb-94
Nb-94 *	$2.0 \cdot 10^{4}$	$1 \cdot 10^{-5}$	LIN 98	Data on spent fuel assembly
Tc-99 **	$2.1 \cdot 10^{5}$	$9 \cdot 10^{-4}$	Updated	All Swedish data
Pd-107 **	$6.5 \cdot 10^{6}$	$1 \cdot 10^{-6}$	LIN 98	Data on spent fuel assembly
Ag-108m *	$1.3 \cdot 10^{2}$	$6 \cdot 10^{-5}$	LIN 98	Data on Swedish BWR spent fuel assembly
Cd-113m **	14	$6 \cdot 10^{-4}$	LIN 98	Data on Swedish BWR spent fuel assembly
Sb-125 *	2.8	$1 \cdot 10^{-1}$	LIN 98	Mainly on measured Swedish data
Sn-126 **	$1.0 \cdot 10^{5}$	$5 \cdot 10^{-7}$	LIN 98	Data on Swiss model inventory, reactor internals
I-129**	$1.6 \cdot 10^{7}$	$3 \cdot 10^{-6}$	Updated	Activity ratio - Modelled activity of I-129 in resins from Swedish NPP and measured activity of Cs-137 in operational waste
Ba-133 *	11	$1 \cdot 10^{-5}$	LIN 98	Data on Swiss model inventory, reactor internals

Radionuclide	Half-life (year)	Correlation factor	Comment	Source for selected correlation factor
Cs-134 **	2.1	1	LIN 98	Mainly on measured Swedish data
Cs-135 **	$2.3 \cdot 10^{6}$	$1 \cdot 10^{-5}$	Updated	Activity ratio – Modelled activity of Cs-135 in resins from Swedish NPP and measured activity of Cs-137 in operational waste
Pm-147 **	2.6	$9 \cdot 10^{-1}$	LIN 98	Data on Swedish BWR spent fuel assembly
Sm-151 **	90	$3 \cdot 10^{-3}$	LIN 98	Data on Swedish BWR spent fuel assembly
Eu-152 **	13	$7 \cdot 10^{-5}$	LIN 98	Data on Swedish BWR spent fuel assembly
Eu-154 **	8.8	$1 \cdot 10^{-1}$	LIN 98	Data on Swedish BWR spent fuel assembly
Eu-155 **	5.0	$7 \cdot 10^{-2}$	LIN 98	Data on Swedish BWR spent fuel assembly
Ho-166m *	$1.2 \cdot 10^{3}$	$4 \cdot 10^{-6}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-232 ***	72	$3 \cdot 10^{-5}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-233 ***	$1.6 \cdot 10^{5}$	$2 \cdot 10^{-8}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-234 ***	$2.4 \cdot 10^{5}$	$1 \cdot 10^{-3}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-235 ***	$7.0 \cdot 10^{8}$	$2 \cdot 10^{-5}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-236 ***	$2.3 \cdot 10^{7}$	$3 \cdot 10^{-4}$	LIN 98	Data on Swedish BWR spent fuel assembly
U-238 ***	$4.5 \cdot 10^{9}$	$4 \cdot 10^{-4}$	LIN 98	Data on Swedish BWR spent fuel assembly

* Correlates to Co-60 ** Correlates to Cs-137 *** Correlates to Pu-239 + Pu-240

5 Discussion

5.1 Uncertainties

The use of correlation factors is one possibility to estimate a reference radionuclide inventory for the purpose of safety assessment studies, when waste-type specific information is not at hand. The correlation factor approach requires that there is a correlation between the activity content of the difficult-to-measure (DTM) nuclide and the key nuclide. Data presented in the literature /e.g. Thierfeldt and Deckert 1995/ shows that this is not always the case. This can be explained by the fact that the correlation factor is based on measurements of different types of materials (fuel, core components, coolant water, resins etc) and on waste from different reactor types (BWR and PWR). There is also a difference between reactors in terms of reactor operation and the chemistry of the coolant water, which also have an effect on the possibility to estimate a general correlation factor. Consequently, large uncertainty is related to general correlation factors. The correlation factors evaluated for specific radionuclides vary within orders of magnitude, which gives an indication of the uncertainty interval.

It can be shown statistically that the quality of the correlation increases with the amount of data used for estimating it, up to a certain point. There are statistical parameters that can be used to judge the quality of a correlation between the key nuclide and the DTM nuclide (for example *the correlation coefficient*). Another method to cope with the uncertainty of a correlation factor that has been proposed /Kashiwagi et al. 2000/ is to calculate a confidence interval for the correlation factor, and to use the upper confidence limit as the correlation factor.

An assessment of the uncertainty to correlation factors employed in /Lindgren et al. 1998/ have been performed by /Cronstrand 2005/. The uncertainty was evaluated as the intervals of 90% confidence for the assumed log-normal distribution of the correlation factors.

No statistical evaluations have been made in this study to determine the quality of the correlation factors selected. The data is not uniform, since it represents different types of waste and therefore no detailed statistical evaluation is judged to be suitable. The uncertainty is illustrated by looking at the reported maximum and minimum value. In Table 5-1 the total uncertainty is given as the ratio between reported maximum and minimum value for each of the correlation factors updated in this study. In Table 5-2 the uncertainty given by the minimum and maximum value from the reference judged to be most relevant for the waste in SFR 1 is given, i.e. the reference which the choice of correlation factor is made upon. The number of data used to calculate the geometric mean, min and max is also given in the table. As can be seen in the table the number of data is limited.

The activity ratios given for Cl-36, Mo-93, Tc-99, I-129 and Cs-135 are not correlation factors in its real definition since they are not calculated from activity measurements on waste streams or from fuel composition estimates. Instead, they are based on data evaluated in two different ways: a calculated activity content of Cl-36, Mo-93, Tc-99, I-129 and Cs-135 /Håkansson 2005, Lundgren 2005, 2006/ and a measured activity of Co-60 and Cs-137 in operational waste produced until 2005 /Gordon 2006/. The calculated activity content is valid for all produced waste until 2005, while the measured activity of the key nuclides is the activity for waste deposited in SFR 1. This means that already produced waste stored at the plants is not accounted for in the activity of the key nuclides. For example, /Ingemansson 2001/ states that all resins from system 331 in O3 are still stored at the plant and that these resins are the main source for Co-60 (60-65% of the Co-60 activity in operational waste). This implies that the activity ratios for Cl-36, Mo-93, Tc-99, I-129 and Cs-135 will overestimate the inventory of these nuclides, if used together with activities of Co-60 and Cs-137 in all produced waste.

It should be stressed that the correlation factors for Cl-36, Mo-93, Tc-99, I-129 and Cs-135 are based on activity ratios and are calculated only with the purpose to be used by SKB in their program PROSIT for estimating a nuclide inventory in SFR 1.

Table 5-1. Uncertainty in selected correlation factors.

Nuclide	Uncertainty (ratio between reported maximum and minimum value)	Number of references
C-14 BWR	10^8	6
C-14 PWR	10^8	5
Cl-36/Co-60	10^{12}	10
Cl-36/Cs-137	10^{12}	9
Ni-59 BWR	10^3	3
Ni-59 PWR	10^4	6
Ni-63 BWR	10^4	8
Ni-63 PWR	10^6	11
Mo-93	10^3	6
Tc-99	10^9	12
I-129	10^9	14
Cs-135	10	7

Table 5-2. Uncertainty in the correlation factors that are judged to be most relevant for the selection of correlation factors.

Nuclide	Correlation factor	Uncertainty		Number of data*
		Minimum	Maximum	
C-14 BWR	3.10^{-3}	$3{\cdot}10^{-6}$	$1{\cdot}10^{-1}$	8
C-14 PWR	8.10^{-2}	$2{\cdot}10^{-2}$	$9{\cdot}10^{-1}$	4
Cl-36/Co-60	$6{\cdot}10^{-7}$	–	–	–
Cl-36/Cs-137	$3{\cdot}10^{-6}$	–	–	–
Ni-59 BWR	$1{\cdot}10^{-3}$	7.10^{-5}	$6{\cdot}10^{-3}$	8
Ni-59 PWR	$3{\cdot}10^{-2}$	$2{\cdot}10^{-2}$	$6{\cdot}10^{-2}$	3
Ni-63 BWR	$8{\cdot}10^{-2}$	$9{\cdot}10^{-3}$	$5{\cdot}10^{-1}$	8
Ni-63 PWR	4	3	6	3
Mo-93	$1{\cdot}10^{-6}$	–	–	–
Tc-99	$9{\cdot}10^{-4}$	$8{\cdot}10^{-7}$	1	83
I-129	$3{\cdot}10^{-6}$	–	–	–
Cs-135	$1{\cdot}10^{-5}$	–	–	–

* Each data is based on several measurements.

The correlation factors recommended in this report and also those given in /Lindgren et al. 1998/ are meant to be applied together with activity estimations of Co-60 and Cs-137 for one year old waste. This means that the correlation factor must be based on activity estimates relevant for one year old waste. The referenced literature do not always give information about the age of the waste for which presented activity measurements or correlation factors are based on. It is judged that Co-60 (half-life 5.3 years) is the only nuclide discussed in this report, that this could have an effect on. The correlation factors given for C-14, Ni-59, Ni-63 and Mo-93 are based on Co-60 activities estimated in waste that could be up to six years of age. Consequently, the estimated correlation factors could be overestimated by a factor of two. However, if the age of the waste is known the data have been recalculated to one year old waste.

Correlation factors reported in /Lindgren et al. 1998/ are median values of a set of experimental data. In this report on the other hand, correlation factors have been estimated as geometric mean values. The latter approach has also been adopted in other countries, for example USA and Spain /Kashiwagi et al. 2000/. How the correlation factor is estimated have a significant influence on the estimated value, see Table 5-3.

Sometimes it is concluded that a correlation factor is plant specific /e.g. Thierfeldt and Decker 1995 for I-129/. This is reasonable since the operating conditions vary between different countries or even between different plants within a country. Examples of such conditions affecting correlation factors are composition of fuel and reactor internals, water chemistry and clean-up system layout.

In /Magnusson et al. 2006/ and /Persson 2005/ reactor specific correlation factors are given for C-14, Ni-59 and Ni-63. However, the amount of data for these correlation factors is very limited why it is judged that it is not relevant to specify reactor specific correlation factors. The calculations made for the production of Cl-36 /Håkansson 2005/, Mo-93, Tc-99, Cs-135 /Lundgren 2006/ and I-129 /Lundgren 2005/ is made for each reactor separately. However, the data reported by the power plants on the activity of Co-60 and Cs-137 in the produced waste, is given as a total for all reactors at the plant. Plant specific activity ratios could be calculated for Cl-36, Mo-93, Tc-99, I-129 and Cs-135 but not for each reactor. In addition, the program PROSIT does not handle reactor or plant specific correlation factors.

For C-14, Ni-59 and Ni-63 different activity ratios are calculated for BWR and PWR. The activity ratios are based on prognoses of both the activity of the difficult to measure nuclide and the key nuclide Co-60, but also on estimates of the distribution of the nuclides between BWR and PWR. All these circumstances indicate that there is a large uncertainty in the estimated activity ratios. Despite this, they support the chosen correlation factors for C-14, Ni-59 and Ni-63 based on measurements performed on ion-exchange resins.

The correlation factors estimated in this report are not based on any measurements on waste from Clab or from Studsvik. The correlation factors should be used carefully when applied to this type of waste. This is especially true for the Studsvik waste which is inhomogeneous since it to a large extent consists of materials other than the waste from the power plants, for example RMI (research, medicine and industry) waste. Estimates of the total activity from Clab and Studsvik are made for Mo-93, Tc-99, Cs-135 in /Lundgren 2006/ and for I-129 in /Lundgren 2005/.

Table 5-3. Comparison of correlation factors calculated as geometric mean values and median values, respectively. Only data from the references that are judged to be most relevant for the selection of correlation factors are included.

Nuclide	Correlation factor [1)]	
	Geometric mean	Median value
C-14 BWR	$3 \cdot 10^{-3}$	$1 \cdot 10^{-2}$
C-14 PWR	$8 \cdot 10^{-2}$	$6 \cdot 10^{-2}$
Ni-59 BWR	$1 \cdot 10^{-3}$	$1 \cdot 10^{-3}$
Ni-59 PWR	$3 \cdot 10^{-2}$	$3 \cdot 10^{-2}$
Ni-63 BWR	$8 \cdot 10^{-2}$	$1 \cdot 10^{-1}$
Ni-63 PWR	4	6
Tc-99	$9 \cdot 10^{-4}$	$1 \cdot 10^{-3}$

[1)] Correlation factors for Cl-36, Mo-93, I-129 and Cs-135 are not mean values but based on a single estimate of the activity ratio and are therefore not included in this table.

5.2 Application of correlation factors for resins

Ion-exchange resins contain the main part of the activity in SFR 1 waste. Limited amount of measured data are available to evaluate correlation factors specific for ion-exchange resins. Instead, the correlation factors are based on for example measurements on reactor coolant or modelling of activity content in spent fuel. Applying such correlation factors to waste consisting of ion-exchange resins mainly as in SFR 1 can be questioned since the ratio in activity between two nuclides estimated in for example a spent fuel assembly or in coolant water possibly is not the same as in the waste package deposited in the repository.

Correlation factors for C-14, Cl-36, Ni-59, Ni-63 and I-129 evaluated in this report are all based on measurements on resins from Swedish power plants or on results from modelling of the activity in such resins using plant specific input data. These correlation factors are therefore considered to be reliable for estimating the activity in resins in SFR 1. For many other nuclides in Table 4-1, the correlation factors are based on estimated activities in a Swedish BWR spent fuel assembly. Even though they are based on data specific for a Swedish reactor, there is an uncertainty whether the activity ratio between the DTM nuclide and the key nuclide is the same in the resins as in the fuel. For other nuclides the correlation factor is based on foreign data, in some cases even foreign data about induced activity. These correlation factors must be considered even more uncertain.

It is impossible to quantify the error introduced when applying the correlation factors given in Table 4-1 to the waste in SFR 1. An indication that measurements on coolant water may not reflect the relative relationship between different radionuclides in resins is given in /Thierfeldt and Deckert 1995/. They state that although I-129 and Cs-137 are released from reactor fuel with the same mechanisms and are transported similarly in reactor systems, filter systems have different retention properties for them. Their comment is also supported by data on decontamination factors observed for clean-up system in a German nuclear power plant, see Table 5-4. Even though no information is given how the decontamination factor is defined, the data shows that the effectiveness of the clean-up system used in that plant varies for different radionuclides.

The correlation factors between Cl-36 and Cs-137 and between Cl-36 and Co-60 in both reactor coolant water and in resins have been investigated by /Xinqi et al. 1991/. The correlation factor Cl-36/Cs-137 for PWR resins is about 40 times lower than that for PWR coolant water. For Cl-36/Co-60 the difference is even larger, the correlation factor is three orders of magnitude lower for the resins than for the coolant water. These results indicate that the activity of Cl-36 in resins will be overestimated if a correlation factor obtained from measurements on coolant water is applied, and that a correlation to Co-60 will overestimate the activity of Cl-36 more than a correlation to Cs-137.

Table 5-4. Nuclide specific decontamination factors in PWRs (from /Thierfeldt and Decker 1995/).

Nuclide	Decontamination factor
I-133	1,000–10,000
I-135	500–3,000
Cs-134/Cs-137	1–1.2
Cr-51	10–80
Mn-54	20–70
Co-58	10–100
Co-60	2–30

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Appendix 1

Literature search

Literature search were conducted in IAEA INIS (International Nuclear Information Service) database, INSPEC database (technical literature in physics, electrical engineering, electronics, communications, control engineering, computers, computing, information technology, manufacturing, production and mechanical engineering) and Bibas (SKB's own database) as well as on internet at www.google.com and www.scholar.google.se.

The results from the search in INIS are given in Table 1.

Search number	Search criteria	Hits
18	((difficult to measure)and(radio*)) and (PY:IN = 1998–2006)	52
17	((((characteri*)and(waste) and (low) and (intermediate) and (activ*) and (PY:IN = 1998–2006))	108
16	(((characteri*)and(waste) and (low) and (intermediate) and (PY:IN = 1998–2006))	272
15	((characteri*)and(waste) and (low) and (PY:IN = 1998–2006))	1300
14	(characteri*)and(waste) and (PY:IN = 1998–2006)	6036
13	((scal*) and (surface contamina*) and (PY:IN = 1998–2006))	62
12	((scal* factor) and (surface contamina*) and (PY:IN = 1998–2006))	0
11	((surface contamina*) and (PY:IN = 1998–2006))	1291
10	((crud)and(scaling factor)) and (PY:IN = 1998–2006)	0
9	(((crud) and (resin*) and(PY:IN = 1998–2006))	13
8	((crud) and (PY:IN = 1998–2006))	156
7	((difficult to measure)and(crud) and (PY:IN = 1998–2006))	0
6	(difficult to measure)and(surface contamina*) and (PY:IN = 1998–2006)	1
5	(correlation factor) and (PY:IN = 1998–2006)	45
4	(correlation factor)and(dtm) and (PY:IN = 1998–2006)	0
3	(correlation factor)and(difficult to measure) and (PY:IN = 1998–2006)	0
2	(difficult to measure)and(scaling factor) and (PY:IN = 1998–2006)	4
1	(difficult to measure)and(scaling factor)	5

The results from the search in INSPEC are given in Table 2.

Search number	Search criteria	Hits*
11	”long-lived” IN TI	6
10	tru IN TI, Advanced Search	47
9	”co-60” IN TI, Advanced Search	4
8	“cs-137” IN TI, Advanced Search	10
7	“corrosion products”	2
6	crud	16
5	“surface contamination”	0
4	“key nuclide”	9
3	“scaling factor”	0
2	“difficult to measure”	5
1	dtm	0

* Hits after a first manual selection

The results from the search in Bibas are given in Table 3.

Search number	Search criteria	Hits
12	”long-lived” IN TI, Advanced Search	42
11	tru IN TI, Advanced Search	8
10	”co-60” IN TI, Advanced Search	1
9	”cs-137” IN TI, Advanced Search	11
8	”corrosion products” IN TI, Advanced Search	7
7	(”corrosion products”), Advanced Search	145
6	(crud), Advanced Search	31
5	(”surface contamination”), Advanced Search	27
4	(”key nuclide”), Advanced Search	4
3	(”scaling factor”), Advanced Search	19
2	(”difficult to measure”), Advanced Search	47
1	(dtm), Advanced Search	10

The search at www.google.com and www.scholar.google.se were performed using the same key words as the searches in the databases together with several additional key words for example ion exchange resin, operational waste, hard to measure and inventory.

Reprint of

”Low and intermediate level waste in SFL 3-5: Reference inventory”

Appendix A: Basis for the reference radionuclide inventory

Lindgren M, Pers K, Skagius K, Wiborgh M, Brodén K,
Carlsson J, Riggare P, Skogsberg M, 1998

Contents

1 General

The purpose of this appendix is to describe the basis for the estimated reference radionuclide inventory in SFL 3-5 at repository closure.

It is not feasible to measure all radionuclides in all individual waste packages. Many radionuclides are difficult to measure and in addition some wastes are not yet produced. Therefor estimates of the radionuclide content in the waste will have to be based on a combination of measurements, calculations and assumptions. In this appendix correlation factors between easy to measure key radionuclides and more difficult to measure radionuclides are selected for the waste foreseen in SFL 3-5. Correlation factors are selected both for surface contamination and neutron-induced activities.

The major tasks performed to develop the reference radionuclide inventory for the SFL 3-5 waste is summarised here. Firstly, the radionuclides that are of importance for the performance of the repository have to be identified. This is done in Chapter 2 by comparing the radionuclide inventory from the pre-study /Lindgren et al. 1994/ with radionuclide inventories reported for similar wastes, in safety studies performed in Sweden and in other countries. In addition, possible formation pathways and sources for individual radionuclides are discussed.

Chapter 3 gives an overview of the studied literature on performed activity measurements and calculations. The information sources comprise mainly of measurements of radionuclides in different waste streams at the Swedish nuclear power plants and in the Central Interim Storage for Spent Fuel (Clab), calculations of activities in different wastes, and general investigations of activity levels and radionuclide composition.

The evaluation of the information sources is discussed in Chapter 4. From this evaluation, three easy to measure radionuclides are selected and correlation factors between these easy to measure key radionuclides and the difficult to measure radionuclides have been selected. In addition contamination levels for surfaces in contact with primary coolant water and other contaminated water have been defined.

The uncertainties in the selected correlation factors are discussed in Chapter 5. There are large uncertainties involved in the selection of general correlation factors. Measured data were for most radionuclides relatively limited and the study does not include an investigation to prove if there are correlations between the key radionuclides and selected radionuclides. In addition, extrapolations were made both to other nuclear facilities and other types of waste.

The selected correlation factors are used to estimate the radionuclide contents in the SFL 3-5 wastes where no waste specific information is at hand, see Appendices B to E. The total radionuclide inventories, estimated at repository closure, for the different repository parts are given in Table 5-4 in the main report.

2 Radionuclide inventory

2.1 Selection of important radionuclides

The radionuclides to be considered in a reference radionuclide inventory for SFL 3-5 is selected in this section. The waste to be disposed of in SFL 3-5 is waste from decommissioning and operation of nuclear facilities, and waste from research, medicine and industries.

Table 2-1 is a compilation of the radionuclides considered in the waste inventory for the SFL 3-5 pre-study /Lindgren et al. 1994/ and some other waste inventories from safety studies performed in Sweden and in other countries. Radionuclides that have been considered in earlier Swedish safety studies are given in three columns. The safety studies SKB 91 /SKB 1992/ and SR 95 /SKB 1995/ concern the disposal of spent fuel and SFR 1 /SKB 1991/ the disposal of operational waste. Radionuclides in the SFL 3-5 waste inventory and in the safety studies for the Finnish VLJ repository /Nykeri 1991/ and in the Swiss repository Wellenberg /NAGRA 1993/, with similar wastes as the SFL 3-5 waste are given in the following columns. In the last two columns radionuclides reported from calculations of the activity content in a reference BWR spent fuel assembly /Kjellbert 1990/ and reported in MIRA, the model inventory for Swiss radioactive waste /Alder and McGinnes 1994/, are given. However, it must be noted that the two last columns do not contain all radionuclides reported in the references.

The aim has been to prioritise radionuclides that are important in the evaluation of the long-term safety of the SFL 3-5 repository. Therefore radionuclides that can play a major role in an operational safety point of view can be missing. In Table 2-2 the radionuclides to be considered in the revised waste inventory are marked with a “In” and the radionuclides to be excluded are marked with “Ex”. The radionuclides already considered in the waste inventory for the pre-study are marked in bold. Radionuclides with half-lives shorter than approximately 2 years and radionuclides with very long half-lives, that are more or less stable, and with expected small content have been excluded from the radionuclide list. In addition, radionuclides occurring in wastes that are not foreseen to be deposited in SFL 3-5 have been excluded. The reasons for excluding radionuclides reported in other waste inventories are given in Table 2-2. However, the list may be further limited for individual wastes by excluding radionuclides with very low activity content or low radiotoxicity. The revised list contains more than 20 additional radionuclides compared to the inventory defined in the pre-study: Na-22, Cl-36, K-40, Ca-41, Se-79, Pd-107, Ag-108m, Cd-113m, Sn-126, Ba-113, Pm-147, Sm-151, Ho-166m, Tl-204, Th-229, U-232, U-233, Pu-242, Pu-244, Am-242m, Cm-245 and Cm-246.

The radionuclide content in the SFL 3-5 waste at repository closure will be determined both by the origin of the waste and the time for waste production. The main sources for the radionuclides in the waste from the nuclear power plants, the central interim storage facility for spent nuclear fuel (Clab) and the encapsulation plant (EP) are surface contamination and neutron induced activity. Therefore, a similar radionuclide composition in the different wastes can be expected. However, the radionuclide composition in waste from Research, Medicine and Industry (RMI) treated at Studsvik will be more waste type specific.

Only a few of the radionuclides in Table 2-2 are easy to measure. Therefore estimates of the activity content of many of the radionuclides will be made with correlation factors between radionuclides that are difficult to measure and key radionuclides that are easy to measure. To achieve as good correlations as possible information about major formation pathways and sources for different radionuclides are essential.

Table 2-1. Compilation of radionuclides considered in different studies.

Atomic Number	Radio-nuclide	Half-life (year)	Pre-study SFL 3-5 (LIN94)	SKB 91 (SKB92)	SR 95 (SKB95)	SFR 1 (THE81)	VLJ (NYK91)	WELLEN-BERG (NAGRA93)	Spent fuel assembly (KJE90)	NAGRA MIRA (ALD94)
1	H-3	1.2E+01	X			X			X	X
4	Be-10	1.6E+06	X					X	X	X
6	C-14	5.7E+03	X	X	X	X	X	X	X	X
11	Na-22	2.6E+00								X
17	Cl-36	3.0E+05		X	X			X	X	X
18	Ar-39	2.7E+02							X	X
19	K-40	1.3E+09						X	X	X
20	Ca-41	1.4E+05						X	X	X
26	Fe-55	2.7E+00	X			X			X	X
26	Fe-60	3.0E+05						X		X
27	Co-60	5.3E+00	X		X	X	X		X	X
28	Ni-59	7.5E+04	X	X	X	X	X	X	X	X
28	Ni-63	9.6E+01	X	X	X	X	X	X	X	X
34	Se-79	6.5E+04		X	X			X	X	X
36	Kr-85	1.1E+01			X				X	X
37	Rb-87	4.8E+10							X	X
38	Sr-90	2.9E+01	X	X	X	X	X	X	X	X
40	Zr-93	1.5E+06	X	X	X			X	X	X
41	Nb-93m	1.4E+01	X						X	X
41	Nb-94	2.0E+04	X	X	X	X		X	X	X
42	Mo-93	3.5E+03	X					X	X	X
43	Tc-99	2.1E+05	X	X	X	X	X	X	X	X
44	Ru-106	1.0E+00	X			X			X	X
46	Pd-107	6.5E+06		X	X				X	X
47	Ag-108m	1.3E+02			X			X	X	X
48	Cd-109	1.2E+00							X	X
48	Cd-113m	1.4E+01			X				X	X
50	Sn-126	1.0E+05		X	X			X	X	X
51	Sb-125	2.8E+00	X						X	X
53	I-129	1.6E+07	X	X	X	X	X	X	X	X
55	Cs-134	2.1E+00	X			X			X	X
55	Cs-135	2.3E+06	X	X	X	X	X	X	X	X
55	Cs-137	3.0E+01	X	X	X	X	X	X	X	X
56	Ba-133	1.1E+01							X	X
61	Pm-147	2.6E+00							X	X
62	Sm-151	9.0E+01		X	X			X	X	X
63	Eu-152	1.3E+01	X						X	X
63	Eu-154	8.8E+00	X		X				X	X
63	Eu-155	5.0E+00	X						X	X
67	Ho-166m	1.2E+03			X			X	X	X
81	Tl-204	3.8E+00							X	X
82	Pb-210	2.2E+01	X	X					X	X
84	Po-210	3.8E-01							X	X
88	Ra-226	1.6E+03	X	X	X			X	X	X
89	Ac-227	2.2E+01	X						X	X
90	Th-228	1.9E+00						X	X	X
90	Th-229	7.3E+03		X	X			X	X	X

Atomic Number	Radio-nuclide	Half-life (year)	Pre-study SFL 3-5 (LIN94)	SKB 91 (SKB92)	SR 95 (SKB95)	SFR 1 (THE81)	VLJ (NYK91)	WELLEN-BERG (NAGRA93)	Spent fuel assembly (KJE90)	NAGRA MIRA (ALD94)
90	Th-230	7.7E+04	X	X	X			X	X	X
90	Th-232	1.4E+10	X	X	X				X	X
91	Pa-231	3.3E+04	X	X	X			X	X	X
92	U-232	7.2E+01						X	X	X
92	U-233	1.6E+05		X	X			X	X	X
92	U-234	2.4E+05	X	X	X			X	X	X
92	U-235	7.0E+08	X	X	X			X	X	X
92	U-236	2.3E+07	X	X	X			X	X	X
92	U-238	4.5E+09	X	X	X			X	X	X
93	Np-237	2.1E+06	X	X	X			X	X	X
93	Np-239	6.4E-03						X	X	X
94	Pu-238	8.8E+01	X	X	X	X	X	X	X	X
94	Pu-239	2.4E+04	X	X	X	X	X	X	X	X
94	Pu-240	6.5E+03	X	X	X	X	X	X	X	X
94	Pu-241	1.4E+01	X			X	X	X	X	X
94	Pu-242	3.8E+05		X	X			X	X	X
94	Pu-244	8.3E+07						X	X	X
95	Am-241	4.3E+02	X	X	X	X	X	X	X	X
95	Am-242m	1.5E+02						X	X	X
95	Am-243	7.4E+03	X	X	X			X	X	X
96	Cm-242	4.5E-01			X			X	X	X
96	Cm-243	2.9E+01	X		X			X	X	X
96	Cm-244	1.8E+01	X		X	X	X	X	X	X
96	Cm-245	8.5E+03			X			X	X	X
96	Cm-246	4.7E+03			X			X	X	X

Table 2-2. Reasons for excluding radionuclides in SFL 3-5 waste. Radionuclides marked with bold are considered in the reference radionuclide inventory.

	Nuclide	Half-life(yr)	Comments	Base for exclusion
In	H-3	1.2E+01		
In	Be-10	1.6E+06		
In	C-14	5.7E+03		
In	Na-22	2.6E-00	Originates from RMI-waste stored in Studsvik (THE81) - Reported in MIRA (ALD94) - Not in Wellenberg inventory (NAGRA93)	Included only when waste type specific information is at hand
In	Cl-36	3.0E+05		
Ex	Ar-39	2.7E+02	Noble gas - Originates from neutron activation of Ar-38 in the biological shield (concrete), which is not foreseen to be deposited in SFL (THE81) - Reported for ”RA”, ”SA” and ”MIF” waste in MIRA [NTB 93-21] - Not in Wellenberg inventory (NAGRA93)	Noble gas
In	K-40	1.3E+09	Mainly from neutron activation of K in the biological shield, not foreseen to be deposited in SFL - About 2.5E+8 Bq found in 2 moulds with non-burnable trash Studsvik - Reported only for ”SA” waste in MIRA (ALD94).	Occur mainly in activated concrete, included when waste type specific information is at hand
In	Ca-41	1.4E+05	Originates from neutron activation of Ca in the biological shield (concrete), which is not foreseen to be deposited in SFL - Other potential sources are targets from Studsvik - Reported for ”RA”, ”SA” and ”MIF” waste in MIRA (ALD94)	Occur mainly in activated concrete, included when waste type specific information is at hand
In	Fe-55	2.7E-00		
Ex	Fe-60	3.0E+05	Reported for the SMA-1 waste (NAGRA93). The nuclide is only found in the decommissioning waste from the PSI accelerator, copper, waste types (ALD94)	No such waste is foreseen
In	Co-60	5.3E-00		
In	Ni-59	7.5E+04		
In	Ni-63	9.6E+01		
In	Se-79	6.5E+04		
Ex	Kr-85	1.1E+01	Noble gas - Originates from RMI-waste in Studsvik (THE81) - Reported for ”RA”, ”SA” and ”MIF” waste in MIRA (ALD94) - Not in Wellenberg inventory (NAGRA93).	Noble gas
Ex	Rb-87	4.8E+10	Fission product -The activity content in spent fuel is about 1E-9 of the Cs-137 content (THE81)	Excluded due to small amounts and long half-life
In	Sr-90	2.9E+01		
In	Zr-93	1.5E+06		
In	Nb-93m	1.4E+01		
In	Nb-94	2.0E+04		

	Nuclide	Half-life(yr)	Comments	Base for exclusion
In	Mo-93	3.5E+03		
In	Tc-99	2.1E+05		
Ex	Ru-106	1.0E-00	Fission product - Reported for ”BA”, ”RA” and ”MIF” waste in MIRA (ALD94) - Not in Wellenberg inventory (NAGRA93)	Excluded due to half-life shorter than 2 years
In	Pd-107	6.5E+06		
In	Ag-108m	1.3E+02		
Ex	Cd-109	1.2E-00	Formed by activation of Cd-108, natural concentration 1% and relatively small cross section	Excluded due to expected small amounts and half-life shorter than 2 years
In	Cd-113m	1.4E+01		
In	Sn-126	1.0E+05		
In	Sb-125	2.8E-00		
In	I-129	1.6E+07		
In	Cs-134	2.1E-00		
In	Cs-135	2.3E+06		
In	Cs-137	3.0E+01		
In	Ba-133	1.1E+01		
In	Pm-147	2.6E-00	Fission product - The initial activity content in spent fuel is about 0.9 of the Cs-137 content (THE81) - Reported for all waste types in MIRA (ALD94) - Not in Wellenberg inventory (NAGRA93)	
In	Sm-151	9.0E+01		
In	Eu-152	1.3E+01		
In	Eu-154	8.8E-00		
In	Eu-155	5.0E-00		
In	Ho-166m	1.2E+03		
In	Tl-204	3.8E-00	Originates from RMI-waste in Studsvik (THE81) - Reported only for ”MIF” waste in MIRA (ALD94) - Not in Wellenberg inventory (NAGRA93)- Used as target source for ”thickness measuring”.	Included only when waste type specific information is at hand
In	Pb-210	2.2E+01		
Ex	Po-210	3.8E-01	Daughter to Pb-210 (Bi-110) ”MIF” waste predominant source in MIRA (ALD94).	Excluded due to half-life shorter than 2 years
In	Ra-226	1.6E+03		
In	Ac-227	2.2E+01		
Ex	Th-228	1.9E-00	Daughter to Th-232 (Ra-228, Ac-228) and U-232	Excluded in the inventory due to half-life shorter than 2 years

	Nuclide	Half-life(yr)	Comments	Base for exclusion
In	Th-229	7.3E+03		
In	Th-230	7.7E+04		
In	Th-232	1.4E+10		
In	Pa-231	3.3E+04		
In	U-232	7.2E+01	Daughter to Pu-236 - Reported for ”BA” waste in the NAGRA inventory (ALD94) and in Wellenberg inventory (NAGRA93)	
In	U-233	1.6E+05		
In	U-234	2.4E+05		
In	U-235	7.0E+08		
In	U-236	2.3E+07		
In	U-238	4.5E+09		
In	Np-237	2.1E+06		
Ex	Np-239	6.4E-03	Daughter to Am-243 - Reported in MIRA (ALD94) and in Wellenberg inventory (NAGRA93)	Excluded due to half-life shorter than 2 years
In	Pu-238	8.8E+01		
In	Pu-239	2.4E+04		
In	Pu-240	6.5E+03		
In	Pu-241	1.4E+01		
In	Pu-242	3.8E+05		
In	Pu-244	8.3E+07	Daughter to Cm-248 - Reported only for ”BA” and ”WA” waste in MIRA (ALD94) and in Wellenberg inventory (NAGRA93)	
In	Am-241	4.3E+02		
In	Am-242m	1.5E+02		
In	Am-243	7.4E+03		
Ex	Cm-242	4.5E-01		Excluded due to half-life shorter than 2 years
In	Cm-243	2.9E+01		
In	Cm-244	1.8E+01		
In	Cm-245	8.5E+03		
In	Cm-246	4.7E+03		

MIRA Swiss model radioactive waste inventory, **PSI** Paul Scherrer Institute in Switzerland), **BA** Operational waste (Swiss), **SA** Decommissioning waste (Swiss), **RA** Changeable non fuel reactor core components (Swiss), **MIF** Medicine, industry and research (Swiss)

2.2 Properties and formation of radionuclides in the SFL 3-5 waste

In this section an overview is given of possible formation pathways and sources for radionuclides included in the SFL 3-5 waste inventory. This information together with know how about material composition and chemical and physical properties of formed radionuclides are the base for the selection of correlation factors.

The radionuclides have been grouped based on the major formation pathways or sources. The radionuclide groups are:

- ***Activation products***, which are formed by neutron induced activation of main constituents and impurities in irradiated materials (mainly from irradiation of core components, internal parts and fuel).
- ***Fission products,*** which are formed by neutron bombardment of uranium isotopes, mainly U-235, followed by fission and formation of new radionuclides. Fission products enter the system due to fuel leakage.
- ***Transuranics,*** which are formed by absorption of neutrons, which yields new radionuclides with higher atomic numbers, e.g. U-238 to Pu-239. Transuranics can enter the system due to fuel leakage.
- ***Other radionuclides*** that are used in the industry, medicine or research work.

A brief overview of formation pathways and sources for potential radionuclides in SFL 3-5 waste are given below /Thierfeldt and Deckert 1995/. In addition, the half-life of the radionuclide is given within brackets.

Irradiated concrete materials will not be disposed in the SFL 3-5 repository. Therefore, radionuclides mainly formed by irradiation of elements found in concrete materials, e.g. K-40, Ca-41 and Na-22, are not included in the overview below. In addition, Tl-204 originates mainly from RMI where it is e.g. used to gauge material thickness. These four radionuclides will be included in the reference waste inventory only when waste type specific information is at hand.

H-3 (Half-life = 12 years)

H-3 is an activation product mainly from Li-6 and Li-7 (negligible). Sources for H-3 are irradiation of B and Li in the absorber materials in control elements, B and Li in the fuel elements and B and Li in the reactor coolant water /Thierfeldt and Deckert 1995/. H-3 is also a fission product from UO_2.

Be-10 (Half-life = 1.6·106 years)

Be-10 is formed by activation of B-10 /Thegerström and Hård 1981/ and beryllium /SKI 1990/. B-10 occurs in control rods. It is also formed by activation of impurities in the fuel and as a fission product from uranium /Kjellbert 1990/.

C-14 *(Half-life = $5.7 \cdot 10^3$ years)*

C-14 is produced through fast neutron reactions with N-14 and O-17 in the coolant water and reactor components and by activation of oxygen in UO_2. It can also be produced by activation of C-13 (thermal neutrons) /Thierfeldt and Deckert 1995/. C-14 is also a fission product from the fuel. Most of the activity is released in gaseous form, but some of it will be present in the waste in insoluble form /Thierfeldt and Deckert 1995/.

Cl-36 ***(Half-life = $3.0 \cdot 10^5$ years)***

Cl-36 is formed mainly by activation of Cl-35 by thermal neutrons in the coolant water and possibly by activation of Cl-37 by fast neutrons. Chlorine can be present as an impurity in the fuel and also in alloys e.g. in Inconel /Thierfeldt and Deckert 1995/, stainless steel /NKA 1990/ and zircaloy.

Fe-55 ***(Half-life = 2.7 years)***

Fe-55 is an activation product from reactions between thermal neutrons and Fe-54 and fast neutrons and Ni-59 /Thierfeldt and Deckert 1995/.

Co-60 ***(Half-life = 5.3 years)***

Co-60 is formed from Co-59 by irradiation with thermal neutrons and from Ni-60 and Cu-63 by fast neutrons /Thierfeldt and Deckert 1995/. Sources for Co-60 is cobalt as an impurity in the nickel used in steel alloys.

Ni-59 ***(Half-life = $7.5 \cdot 10^4$ years)***

Ni-59 is formed from Ni-58 by irradiation with thermal neutrons and from Ni-60 by irradiation with fast neutrons. Sources for Ni-59 are austenitic steel in the reactor and activation of nickel dissolved in the coolant and in corrosion particles deposited on the core /Thierfeldt and Deckert 1995/. The major sources in the reactor are the metal parts of stainless steel (≈ 10%) and Inconel (50-60%). In addition, nickel is found as an impurity in zircalloy (≈ 40 ppm) and in reactor fuel (≈ 20 ppm).

Ni-63 ***(Half-life = 96 years)***

Ni-63 is formed from Ni-62 by irradiation with thermal neutrons and from Zn-66 and Cu-63 by fast neutrons. Sources for Ni-63 are austenitic steels in the core, activation of dissolved Ni-62 in the coolant and in corrosion particles in the core /Thierfeldt and Deckert 1995/.

Se-79 ***(Half-life = $6.5 \cdot 10^4$ years)***

Se-79 is a fission product from uranium. Se-79 is also formed from Se-78 by reaction with thermal neutrons and from Kr-82 and Br-79 by reaction with fast neutrons /Thierfeldt and Deckert 1995/.

Sr-90 ***(Half-life = 29 years)***

Sr-90 is a fission product from uranium. Sr-90 can also be formed by activation of zirconium in steel or zircaloy, but to a lesser extent /Thierfeldt and Deckert 1995/.

Zr-93 ***(Half-life = $1.5 \cdot 10^6$ years)***

Zr-93 is an activation product produced by neutron-gamma reaction of zirconium containing materials (fuel, cladding, guide tubes etc) Zr-93 is also a fission product from uranium.

Nb-93m ***(Half-life = 14 years)***

Nb-93m is formed due to activation of Nb-93 in metals by fast neutrons. Nb-93m is also a fission product from uranium.

Nb-94 ***(Half-life = $2.0 \cdot 10^4$ years)***

Nb-94 is an activation product formed by reactions between thermal neutrons and Nb-93. Nb-93 is a stable isotope and exist as a minor constituent in stainless steel and in trace amounts in nickel based alloys and in the fuel. Nb-94 is also a fission product from uranium.

Mo-93 ***(Half-life = $3.5 \cdot 10^3$ years)***

Mo-93 is formed from Mo-92 by irradiation with thermal neutrons. In addition, formation from Ru-96 and Mo-94 by fast neutrons is possible, but to a lesser extent. Ruthenium and molybdenum are present as impurities in the fuel. Molybdenum can be present as an alloy element in steels and in lubrications. Sources for Mo-93 are metallic materials (steels and zircaloy in the core region), and activation of molybdenum in dissolved form or as corrosion particles in the coolant /Thierfeldt and Deckert 1995/.

Tc-99 ***(Half-life = $=2.1 \cdot 10^5$ years)***

Tc-99 is formed by irradiation of Mo-98 with thermal neutrons and as a fission product. Sources for Tc-99 are uranium fission in the fuel and irradiation of molybdenum as an alloy element in steels and zircaloy /Thierfeldt and Deckert 1995/.

Pd-107 ***(Half-life = $6.5 \cdot 10^6$ years)***

Pd-107 is formed by reactions between thermal neutrons and Pd-108 and by reactions between fast neutrons and Cd-110 and Ag-107. Pd-107 is also a fission product from uranium /Thierfeldt and Deckert 1995/.

Ag-108m ***(Half-life = $1.3 \cdot 10^2$ years)***

Ag-108m is an activation product from silver and occurs due to activation of spent fuel assemblies. It is also a fission product from the fuel, /Kjellbert 1990/. Silver does also occur as an impurity in steel, although at very low concentrations.

Cd-113m ***(Half-life = 14 years)***

Cd-113m is formed by reaction between thermal neutrons and Cd-112 and by reactions between fast neutrons and Cd-113. Cd-113m is also a fission product from uranium /Thierfeldt and Deckert 1995/. Cd-113m can be found in PWR control rods /Thegerström and Hård 1981/.

Sn-126 ***(Half-life = $1.0 \cdot 10^5$ years)***

Sn-126 is a fission product from uranium.

Sb-125 ***(Half-life = 2.8 years)***

Sb-125 is produced both as a fission product and as an activation product /Thegerström and Hård 1981/.

I-129 ***(Half-life = $1.6 \cdot 10^7$ years)***

I-129 is mainly a fission product from uranium. The sources for I-129 are the fuel elements and uranium material impurities. The formation from Te-128 by thermal neutrons and from Xe-129 with fast neutrons is of minor importance, as these elements are usually not present in construction materials in nuclear power plants /Thierfeldt and Deckert 1995/.

Cs-134 ***(Half-life = 2.1 years)***

Cs-134 is a fission product from uranium. It is indirectly produced by neutron capture by the fission product Cs-133. Measured activities in fuel contaminated waste are of the same order as Cs-137 /Thegerström and Hård 1981/.

Cs-135 ***(Half-life = $2.3 \cdot 10^6$ years)***

Cs-135 is a fission product from uranium. Cs-135 is found in spent fuel elements and in materials containing uranium as impurities /Thierfeldt and Deckert 1995/.

Cs-137 ***(Half-life = 30 years)***

Cs-137 is a fission product from uranium.

Ba-133 ***(Half-life = 11 years)***

Ba-133 is formed by reactions between thermal neutrons and Ba-132 and by reactions between fast neutrons and Ce-136 and Ba-134 /Thierfeldt and Deckert 1995/. The main source is barium as an impurity in steel.

Pm-147 ***(Half-life = 2.6 years)***

Pm-147 is mainly formed by fission, but can also, to a lesser extent, be formed as an activation product in the spent fuel assemblies /Kjellbert 1990/.

Sm-151 ***(Half-life = 90 years)***

Sm-151 is formed by reactions between thermal neutrons and Sm-150 and by reactions between fast neutrons and Eu-151. Sm-151 is also a fission product from uranium /Thierfeldt and Deckert 1995/.

Eu-152 ***(Half-life = 13 years)***

Eu-152 is formed by reactions between thermal neutrons and Sm-151 and by reactions between fast neutrons and Gd-152 and Eu-153. Eu-152 is also a fission product from uranium /Thierfeldt and Deckert 1995/. Europium is present in steel /NKA 1990/.

Eu-154 ***(Half-life = 8.8 years)***

Eu-154 is formed by reactions between thermal neutrons and Eu-153 and by reactions between fast neutrons and Gd-154. Eu-154 is also a fission product from uranium /Thierfeldt and Deckert 1995/.

Eu-155 ***(Half-life = 5.0 years)***

Eu-155 is formed by reactions between thermal neutrons and Sm-151 and by reactions between fast neutrons and Gd-155. Eu-155 is also a fission product from uranium /Thierfeldt and Deckert 1995/.

Ho-166m ***(Half-life = $1.2 \cdot 10^3$ years)***

Ho-166m is both a fission product and an activation product that can be found in a spent fuel assembly /Kjellbert 1990/.

Radionuclides in the natural decay chains

The radionuclides in the natural decay chains, as shown in Figure 2-1, have been considered throughout this report.

Uranium is a major constituent of reactor fuel. In addition uranium may occur as an impurity in many materials e.g. steel /Thierfeldt and Deckert 1995/ and consequently these radionuclides may occur also in other irradiated materials. This has, however, not been considered for the SFL 3-5 waste.

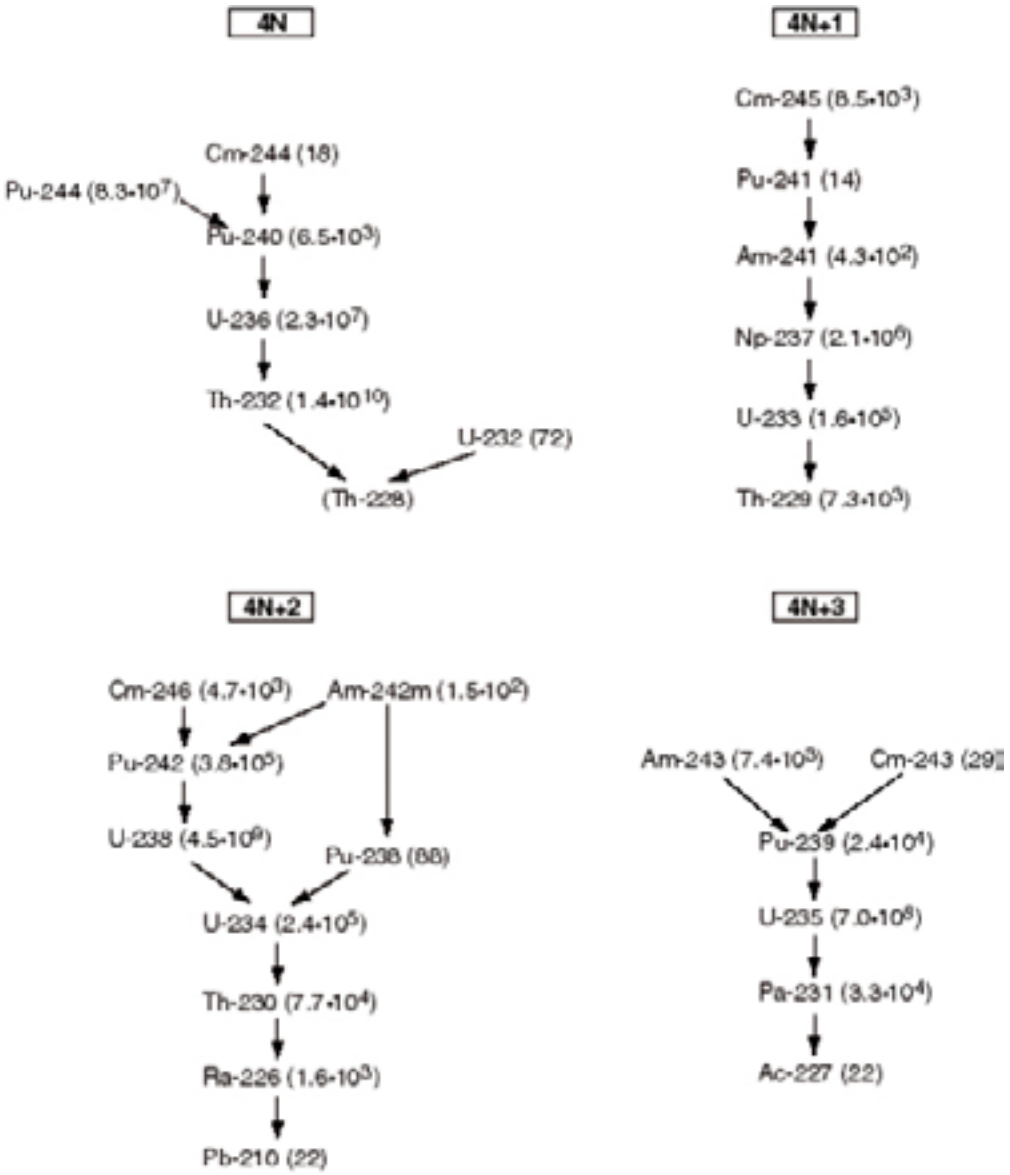

Figure 2-1. *Radionuclides in the natural decay chains.*

3 Information sources

In this chapter a summary is given of measurements and/or calculations that have been used in this report to evaluate contamination levels of key radionuclides and correlation factors for difficult to measure radionuclides in SFL 3-5 waste. The evaluation and the selection of data are given in Chapter 4.

Several studies have been undertaken to investigate the possibility to correlate difficult to measure radionuclides to key radionuclides that are easy to measure. The conclusion from many of the investigations is that it is a difficult task that must be done with great care. Some aspects that must be considered are:

- The estimated correlation factors may be waste, plant or reactor type specific.
- The time aspect must be taken into account, especially for short-lived radionuclides, in the evaluation of correlation factors from measured and calculated radionuclide concentrations.
- The material composition and its content of impurities determine the concentration of radionuclides especially at neutron activation.

Measurements and calculations performed on different types of contaminated water systems and wastes are described in the following sections. The information has been divided into the following groups:

- Contaminated water systems and surface contamination (section 3.1)
- Neutron activation (section 3.2)
- Other information including waste from Research, Medicine and Industry (RMI) (section 3.3).

3.1 Studies on contaminated water systems and surface contamination

3.1.1 Studies on contaminated water systems in Sweden

The content of transuranics in the low-level trash waste from Clab has been estimated. The amount of transuranics and the amount of Co-60 in the coolant water has been measured and a relation between the total alpha content and Co-60 has been evaluated /Grahn 1987/. In the report results from nuclide specific alpha analysis (Pu-239+240, Am-242, Pu-238+241, Cm-244 and Cm-242) in the coolant during cooling of transport casks with spent fuel from different Swedish nuclear power plants (Oskarshamn 1, Ringhals 2, Forsmark 1, and Barsebäck 1) are given. The activity content in the trash waste is then calculated from its Co-60 content and the evaluated relation between the alpha content and Co-60 in the coolant.

The activity of Co-60 and transuranics (Pu-238+Am-241, Pu-239+240, Cm-242, Cm-243, Cm-244, Am-243) in filters and spent ion-exchange resins in Clab has been calculated. The calculations are based correlations between transuranics and Co-60 evaluated from measurements during cooling of transportation casks. The content of the radionuclides are calculated for the internal cooling circuit, the clean-up system for process- and floor drainage water, the cooling and clean-up system in the receiving pools and the cooling and clean-up system in the storage pools during the years 1988-1990 /Axelsson 1989, 1990, 1991/.

A continuous evaporator was developed and used for collection of integral samples of reactor coolant water from Ringhals 1 during the 1983/84 operation period /Liljenzin and Johnson 1985/. Analytical processes were developed for the determination of Sr-90 and transuranics (Pu-238, Pu-239+240, Am-241 and Cm-244) in these samples and in samples from spent ion-exchange resins. The results from the analyses were compensated for decay and the reported activities in the

samples from the ion-exchange resins were from the time for the yearly reactor shutdown (1st of July) and the activities in the water samples were from the end of the sampling period. Internal correlations among the measured radionuclides were studied. Some correlation between Sr-90 and gamma-emitters, mainly Ag-108m but also Co-60, was observed. No correlation could be observed between gamma-emitters and Pu-239.

The activity content of Sr-90 and the transuranics (Pu-238, Pu-239+240, Am-241 and Cm-244) in spent ion-exchange resins and in the cleaning system for the reactor coolant water and the fuel pool water for the years 1988-1992 has been estimated /Aronsson 1990, Ericson 1991, Sjöström 1992, 1993/. The activity content has been calculated with the help of analyses and mathematical models. Quarterly measurements of the Co-60 and transuranics concentrations in the reactor coolant and fuel pool water at all four units at Ringhals form the basis for the estimates. The calculated activities in the ion-exchange resins are given in the report, whereas the measured data were left out. However, the uptake of activity in the ion-exchange resins is 100% for all radionuclides in the applied mathematical model /Carlsson 1996/.

Measurements of the content of Co-60, Cs-137, Ni-59, Ni-63 and Tc-99 in water samples from the reactors and fuel pools in the Swedish nuclear power units (Ringhals 1–4, Oskarshamn 1–3, Barsebäck 1–2) and in Clab has been performed and compiled /Roos 1994/. The data sets were used to evaluate ratios between easy to measure radionuclides (Co-60 and Cs-137) and radionuclides that are more difficult to measure. The aim was to increase the knowledge of how these ratios can be applied to estimate more difficult to measure radionuclides in reactor waste aimed for SFR 1.

The activity content in two serial coupled ion-exchange resins in the primary circuit in Oskarshamn 3 have been analysed mainly with respect to transuranics and gamma-emitters (Co-60, Sb-125, Cs-134, Cs-137, Pu-239+240, Pu-238, Am-241, Cm-244, and Cm-242). In addition, some more short-lived radionuclides were reported /Skålberg 1987/. The results from the analyses were compensated for decay and the reported activities in the samples from the ion-exchange resins were from the time of yearly reactor shutdown (1st of July). As a remark it was mentioned that the ion-exchange resin works as a filter for particles in addition to its ion-exchange capacity.

An investigation by /Chen et al. 1992/ deals with analyses of the alpha-emitting isotopes Pu-238 and Pu-239+240 in primary reactor coolant water. The objective of the study was to develop methods to determine transuranic radionuclides in the primary reactor coolant water and to estimate the efficiencies of the ion-exchange system and mechanical filters. Samples of coolant water from the primary circuit of Ringhals 2 were collected before and after the ion-exchange filter system to obtain data on the plutonium levels at these points of measuring.

3.1.2 Studies on surface contamination in Sweden

Surfaces that have been in contact with primary coolant water will be contaminated on the surfaces by so called CRUD (Chalk River Unidentified Deposits). The surface contamination is composed of activated corrosion products from structural materials in contact with the reactor coolant water and fission products from leaking fuel elements.

Decontamination of two steam generators from the Swedish Ågesta reactor has been performed at Studsvik. The activities of alpha-emitters (plutonium and uranium) and beta-emitters (Co-60, Cs-137) have been measured in the decontamination solutions, as well as the concentration of tritium /Brodén 1993, 1996/. The content of fission products in the CRUD is probably higher in the Ågesta reactor than in CRUD from modern Swedish power reactors since the Ågesta reactor have had problems with fuel leakage.

The amount of alpha active transuranics (Pu-238, Pu-239+240, Am-241, Cm-242 and Cm-244) and beta emitters (Co-60, Sb-125) in CRUD scraped from fuel in Ringhals, Barsebäck and Oskarshamn has been measured and reported by /Fridemo et al. 1985a, 1985b/. All reported activities were given for a reference time (date of reactor closure), which means that the activities are compensated for deviations in date for the performed measurements and analyses.

The surface contamination on the systems outside the reactor tank in Oskarshamn 2 has been calculated by ABB Atom /Lundgren 1991/ for an extended operation of the plant until year 2010. The surface contamination on primary systems of the reactor has been predicted with the computer code BKM-CRUD (ABB Atom code). Two different fuel leakage scenarios were considered in these calculations, one standard pinhole scenario and a more severe fuel leakage that released the fission products and actinides every 10 years. The report gives derived conversion factors for the evaluation of the contamination levels in the secondary systems. The calculated contamination levels are compared with measurements and show reasonable agreement.

Contamination by fission products can be measured on reactor system surfaces with a mobile analyser for detection of CRUD in piping (MADAC). Such measurements have been performed in Barsebäck 1 and 2 on a number of the reactor systems /Ernström 1996/. The database mainly contains information on short-lived radionuclides, but some information about contamination levels (Bq/m2) of Co-60 and Sb-125 was given.

Nuclide specific analyses of correlations between easily measured radionuclides and difficult to measure ones have been performed by /Thierfeldt and Deckert 1995/. Data were gathered for different waste samples from Sweden but also from other countries e.g. Germany and Japan. The waste samples were taken from reactor coolants, evaporator bottoms, solid wastes, CRUD, filter cakes, concentrates, resins, lubrication, and coolant from the fuel pond. Correlation factors were evaluated and discussed for H-3, C-14, Cl-36, Ca-41, Ni-59, Ni-63, Sr-90, Mo-93, Tc-99, I-129, Cs-135, U-234, Np-237, Pu-238, Pu-239+240, Pu-241, Am-241. In addition, the physical and chemical backgrounds behind the proposed correlations were elucidated.

The activities of transuranics in scrape samples from fuel rods have been measured at different axial positions at certain time intervals on all power units in Sweden /Granath 1983, Westerberg 1986, Fridemo 1989/. The radionuclides analysed were Pu-238, Pu-239+240, Am-241, Cm-242, and Cm-244. There were large variations in surface concentration levels (Bq/m^2) between the power units, but also within one power unit. The activity of the activated corrosion product (Co-60) in scrape samples from fuel rods were reported in /Troberg 1983, 1984, Rundlöf 1985abcd/. The scrape samples were taken from fuel rods taken out from the reactor during the yearly reactor shutdown.

3.1.3 Studies on surface contamination in other countries

The contamination in a PWR reactor has been the subject of a study by /Smith et al. 1985/. Average contamination levels after 5, 25 and 100 years from reactor shutdown were proposed. The contamination of different radionuclides has been established by comparing experimental values with theoretical ones.

The surface contamination and radionuclide distribution at seven nuclear power plants (BWR's and PWR's) have been investigated to provide a database /Robertson et al. 1984/. Emphasis has been placed on measuring the long-lived radionuclides, which are of special concern for a low-level waste management standpoint. The measured activity concentrations were corrected for decay to date of reactor shutdown for the reactors that have been shutdown or to sampling date for operating reactors.

A comprehensive database for regulatory assessment of radiological factors associated with reactor decommissioning and waste disposal has been generated in the US /Robertson et al. 1993/. Radioactive material from the actual dismantling of a light-water breeder reactor, Shippingport Station, and other commercial reactors in the US was used for radionuclide and stable element analyses. Excellent maintenance and operation of the Shippingport reactor as well as very few significant fuel leakages resulted in radionuclide contamination of primary piping systems and other components that was significantly lower than in other commercial nuclear power stations. The reported radionuclide concentrations on primary and secondary coolant piping were the activities of September 1988, whereas the reactor finished operation in October 1982.

The correlation factor between Cl-36 and Cs-137 has been evaluated based on measurements in reactor coolant water and resins from the reactor coolant cleanup system in Swiss nuclear power plants /Xinqi et al. 1991/.

3.2 Studies on neutron activation

The radionuclide content in materials exposed to irradiation will be influenced by several factors. The concentration of a particular radionuclide at a given location in a reactor depends on the concentration of the parent isotope, the amount of major constituents, the amount of impurities, the cross section of the specie, the duration of the exposure of the neutron flux, the distance to the radiation source etc.

3.2.1 Studies on activated components in Sweden

The neutron induced activity in reactor internals has been calculated, with the computer code AKTGAMMA, for a BWR 3000 /Lundgren 1982/. This type of reactor is installed in Oskarshamn 3 and Forsmark 3. It has been assumed that the reactor produced 7000 EFPH (effective full power hours) each year during an operation period of 40 years. The average fuel burn-up was set to 32 MWd/kg U. The induced activities were reported at certain times after reactor shutdown. Further, the data on material compositions were obtained from material analyses. The calculations were based on the material analyses available at that time. However, more recent analyses indicate that small amounts of material impurities, which were not included in the older analyses, should have been considered. The used computer code was originally developed for calculations of gamma-emitting nuclides only. The radionuclide database has, however, been extended with a selected number of beta-emitters, whereas, no alpha-emitters were included.

Neutron induced activity has been calculated for the PWR reactor Ringhals 2 by /Højerup 1993/. The induced activity was reported at certain times after reactor shutdown for sections with different vertical positions in relation to the core. The neutron flux distributions in the reactor for different energy levels were calculated first followed by the activation calculations. Neutron activation was given for 20, 30 and 40 years of irradiation.

In an old decommissioning study /SKB 1979/ the neutron induced activity and the CRUD contamination on the reactor tank and internal parts are reported for Oskarshamn 2 after 40 years of operation. Data are given for a limited number of radionuclides such as Fe-55, Co-60, Ni-59 and Ni-63.

The neutron induced activity in the storage cassettes used in Clab has been calculated and reported by /Elkert 1993/. The radionuclides reported are H-3, Mn-54, Fe-55, Co-60, Ni-59, Ni-63, Nb-93m and Nb-94.

3.2.2 Studies on induced activity in other countries

The activity of decommissioning waste consisting of components outside the active core of the two BWR nuclear reactors (Asea Atom type) at the Olkiluoto Power Plant in Finland has been calculated /Anttila and Wasastjerna 1989/. The activation and subsequent cooling of the components have been analysed with the ORIGEN-S computer program. Data for the core grid, the moderator tank, and the moderator tank cover have been used to evaluate correlation factors for the induced activity. The life-time of the reactors was set to 40 years and the average load factor was 0.9, which means an irradiation time of 36 effective full-power years. The activation was calculated at discharge and at certain time intervals.

/Smith et al. 1978/ calculated the activities of the radionuclides present in the reactor vessel, the internal structures and in the surrounding shielding enclosure for a US reference PWR. The reactor life-time was 40 years and the load factor 0.70, which corresponds to 30 effective full-power years. The activation was calculated with the computer code ORIGEN. The neutron induced

activity at reactor shutdown were given for the shroud, core barrel, thermal shields, inner cladding of the vessel, and the lower 5 m of vessel wall at the time for reactor shutdown.

3.3 Other information, including waste from RMI

3.3.1 Waste in Sweden

/Thegerström and Hård 1981/ gives a compilation of the activity content in the waste that was predicted to be stored in SFR 1 at that time. The radionuclides of interest and their activity content were reported for operational wastes, core components, decommissioning wastes and wastes from Studsvik. The induced activity was reported separated from surface contamination.

Since 1980, the activity measurements performed during production of RMI waste packages at Studsvik have been registered. The register contains no data on packages produced before 1980. A study on transuranics in drums with ashes has been performed at Studsvik /Andersson 1996/. Measurements of the activities of Am-241, Pu-239+240 and Cs-137 gave the following correlations, which are applied for ash-drums to SFR 1:

- The activity content of Pu-239+240 can be assumed to be 1% of the activity of Cs-137.
- The activity of Am-241 can be assumed to be 4% of the activity of Cs-137.

/Gustafsson 1995/ has made a decommissioning plan for the test reactor R2 at Studsvik. The decommissioning plan includes e.g. results from activation calculations.

The activity of activation products, heavy radionuclides and fission products in a reference BWR fuel assembly (38000 MWd/tU) were calculated with the computer code ORIGEN /Kjellbert 1990/.

3.3.2 Studies on waste in other countries

A model radioactive waste inventory has been developed for Swiss waste disposal projects /Alder and McGinnes 1994/. The waste categories included are operational waste, changeable non-fuel reactor core components, decommissioning waste and reprocessing waste as well as waste from medicine, industry and research. The database comprises data for 80 waste types and the activity content of a large number of radionuclides is reported. The database describes the conditioned and packed waste and does not separate surface contamination from induced activity. The activities are reported for specified reference times (≤ 5 years).

During dismantling of the US Shippingport Station (light-water breeder reactor core, typical of a commercial pressurised water reactor core) a number of piping and hardware samples (BWR's and PWR's) were analysed as part of a reactor ageing study /Robertson et al. 1993/. Radiological characterisations of the corrosion films deposited on the inside surfaces of the pipes were performed to provide important information of the radionuclide concentrations and distributions. In the same study, samples from fuel disassembly hardware, i.e. neutron activated zircaloy-4; Inconel-X750 and Type-348 stainless steel were analysed for long-lived neutron activation products. The reported radionuclide concentrations on primary and secondary coolant piping were the activities of September 1988, whereas the reactor finished operation in October 1982.

The stocks of radioactive wastes in the United Kingdom as existing on 1 April 1994, together with forecasts of radioactive wastes arising in the future has been reported /UK Nirex Ltd. 1996/. The report gives forecasts of e.g. the activities in decommissioning waste from a PWR (Sizewell B).

3.4 Summary of investigations

The information used for evaluation of correlation factors for surface contamination and neutron induced activity are summarised in Tables 3-1 and 3-2 respectively.

Table 3-1. Literature referred to in the evaluation of correlation factors in CRUD between radionuclides that are difficult to measure and radionuclides that are easy to measure. Underlined radionuclides are easy to measure (key nuclides). Available information that has not been included in this evaluation is within (brackish).

Reference	Type	Measured data		Calculated data		Type of unit
Contaminated water systems and surface contamination in Sweden						
Aronsson P O, 1990 Ericson M, 1991 Sjöström T, 1992 Sjöström T, 1993	Measurements of quarterly concentration of Sr-90 and transuranics in the reactor coolant and fuel pool water, have formed the basis for the calculations of the activity content of these in spent ion-exchange resins. (The measured data were not included in the reported)			(Sr-90) Pu-238 Pu-239+240 Am-241	Cm-244	Ringhals 1 (BWR) Ringhals 2-4 (PWR)
Axelsson A, 1989 Axelsson A, 1990 Axelsson A, 1991	Measurements of Co-60 in water in the storage pool and the internal cooling circuit. Activity of transuranics in filters and spent ion-exchange resins has been calculated	Co-60		(Pu-238+Am-241) Pu-239+240 Am-243	Cm-243 Cm-244	Clab
Brodén K, 1993, 1996	The activity in the decontamination solutions from decontamination of two steam generators from Ågesta has been measured.	H-3 Co-60 Cs-134 Cs-137 Eu-152	Eu-154 Eu-155 Pu-238 Pu-239 (uranium)			Ågesta (heavy water)
Chen Q et al. 1992	Analyses of alpha-emitting isotopes in primary reactor coolant water	Pu-238 Pu-239+240				Ringhals 2 (PWR)
Ernström R, 1996	MADAC measurements	Co-60 Sb-125				Barsebäck 1 (BWR) Barsebäck 2 (BWR)
Fridemo L et al. 1985a Fridemo L et al. 1985b	The amount of radionuclides in CRUD samples from fuel was measured for a number of samples	Co-60 Sb-125 Pu-238 Pu-239+240 Am-241 Cm-244				Ringhals 2 (PWR) Ringhals 1 (BWR) Barsebäck (BWR) Oskarshamn (BWR)
Grahn P H, 1987	The amount of transuranics has been measured together with the activity of Co-60 in the coolant water in Clab	Co-60 Pu-239+240 Am-243	(Pu-238+241) Cm-244			Clab

Reference	Type	Measured data		Calculated data		Type of unit
Contaminated water systems and surface contamination in Sweden						
Kjellbert N, 1990	The activity of activation products, heavy radionuclides and fission products in a reference BWR fuel assembly were calculated with the computer code ORIGEN			> 50 nuclides		Fuel assembly, BWR
				Be-10	Ag-108m	
				C-14	Cd-113m	
				Cl-36	Sn-126	
				Fe-55	I-129	
				Co-60	Cs-134	
				Ni-59	Cs-135	
				Ni-63	Cs-137	
				Se-79	Ba-133	
				Sr-90	Pm-147	
				Zr-93	Sm-151	
				Nb-93m	Eu-152	
				Nb-94	Eu-154	
				Mo-93	Eu-155	
				Tc-99	Ho-166m	
				Pd-107		
				Nuclides in natural decay chains		
Liljenzin J O and Johnson S, 1985	The concentration of Sr-90 and transuranics was measured in reactor coolant water and in samples from spent ion-exchange resins (Ringhals 1 during the 1983/84 operation period)	Co-60	Pu-238			Ringhals 1 (BWR)
		Sr-90	Pu-239			
		Cs-134	Am-241			
		Cs-137	Cm-244			

Reference	Type	Measured data		Calculated data		Type of unit
Contaminated water systems and surface contamination in Sweden						
Lundgren K, 1991	The radionuclide inventories in contaminated BWR systems outside the reactor tank are predicted with the computer code BKM CRUD			Co-60	Pu-238	Oskarshamn 2 (BWR)
				(Fe-55)	Pu-239	
				Ni-59	Pu-240	
				Ni-63	Pu-241	
				Sr-90	Am-241	
				Cs-134	Cm-244	
				Cs-137		
Roos P, 1994	Measurements of the concentration in water samples from the reactors and fuel pools.	Co-60	Tc-99	Cs-135 (theoretical value)		Ringhals 1 (BWR)
		Ni-59	Cs-134			Ringhals 2–4 (PWR)
		Ni-63	Cs-137			Oskarshamn 1–3 (BWR)
						Barsebäck 1–2 (BWR)
						Clab
Skålberg M, 1987	The activity content in two serial coupled ion-exchange resins in the primary circuit has been analysed.	Co-60	Pu-238			Oskarshamn 3 (BWR)
		Sb-125	Pu-239+240			
		Cs-134	Am-241			
		Cs-137	Cm-244			
Thegerström C and Hård E, 1981	A compilation of the activity content in waste that was estimated to be stored in SFR at that time.			C-14	Cs-134	Swedish operational waste, core components, decommissioning waste and waste from Studsvik
				Fe-55	Cs-135	
				Co-60	Cs-137	
				Ni-59	Pu-238	
				Ni-63	Pu-239	
				Nb-94		
				Cd-113m		
				I-129		

Reference	Type	Measured data		Calculated data		Type of unit
Contaminated water systems and surface contamination in Sweden						
Thierfeldt S and Deckert A, 1995	Data from studies and compilations performed in various countries were used to evaluate correlations. The samples used were taken from reactor coolant, evaporator bottoms, solid waste, CRUD, filter cake, concentrates, resins, lubrication and coolant from the fuel pond.	Co-60 Ni-59 Ni-63 Sr-90 Tc-99 Sb-125 Cs-134 Cs-137 U-235 U-236 U-238	Np-237 Pu-238 Pu-239+240 (Pu-241) Am-241 Am-243 Cm-243 Cm-244 H-3 (Ger.) C-14 (Ger.,Jap)			Data gathered from Sweden (Swedish database). In addition information from Germany and Japan.
Alder J C and McGinnes D F, 1994	A model radioactive waste inventory has been developed for Swiss waste disposal projects.			>50 nuclides Co-60 Se-79 Sr-90 Tc-99 Pd-107 Cd-113m Sn-126	 I-129 Cs-134 Cs-135 Cs-137 Ba-113 Pm-147 Sm-151	Swiss reference waste inventory
Robertson D E et al. 1993	During dismantling of the US Shippingport Station a number of piping and hardware samples (BWR's and PWR's) were analysed.	C-14 Fe-55 Co-60 Ni-59 Ni-63 Nb-93m	Nb-94 Pu-238 Pu-239+240 Am-241 Cm-244			US PWR and BWR

Reference	Type	Measured data		Calculated data		Type of unit
Contaminated water systems and surface contamination in Sweden						
Robertson D E et al. 1984	The residual radionuclide concentrations, distribution and inventories at seven nuclear power plants (BWR's and PWR's) have been investigated. Emphasis has been placed on measuring the long-lived radionuclides.	Fe-55	I-129			US PWR and BWR
		Co-60	Cs-134			
		Ni-59	Cs-137			
		Ni-63	Pu-238			
		Sr-90	Pu-239+240			
		Nb-94	Am-241			
		Tc-99	Cm-244			
Smith G M et al. 1985	The contamination in a PWR reactor has been the subject of this study. The contamination of different radionuclides has been established by comparing experimental values with theoretical ones.			(Fe-55)	Cs-135	US PWR
				Co-60	Cs-137	
				Ni-59	Pm-147	
				Ni-63	Sm-151	
				Sr-90	Pu-238	
				Zr-93	Pu-239	
				Nb-94	Pu-240	
				Tc-99	Pu-241	
				Sb-125	Am-241	
				Cs-134	Cm-244	
Xinqi L et al., 1991	Determination of the Cl-36 content in reactor cooling water and active resins from Swiss nuclear power plants.	Cl-36				
		Cs-137				

Table 3-2. Literature referred to in the evaluation of correlation factors in neutron activated metals between radionuclides that are difficult to measure and radionuclides that are easy to measure. Underlined radionuclides are easy to measure (key nuclides).

Reference Information from Sweden	Type	Measured data	Calculated data		Type of unit
Elkert J, 1993	The neutron induced activity in the storage cassettes (steel and boron steel) from Clab has been calculated.		H-3	Ni-63	Clab
			Fe-55	Nb-93m	
			Ni-59	Nb-94	
			Co-60		
Højerup C F, 1993	Neutron induced activity has been calculated for the PWR reactor Ringhals 2.		Ni-59		Ringhals 2 (PWR)
			Fe-55		
			Ni-63		
			Co-60		
Lundgren K, 1982	The induced activity in reactor internals has been calculated for a BWR 3000.		C-14	Co-60	BWR 3000
			Fe-55	Nb-93m	
			Ni-59	Nb-94	
			Ni-63	Tc-99	
SKB, 1979	In an old decommissioning study the neutron induced activity and the CRUD contamination on the reactor tank and internal parts are reported for Oskarshamn 2.		Fe-55	Co-60	Oskarshamn 2 (BWR)
			Ni-59		
			Ni-63		
Gustafsson L, 1995	Decommissioning of a research reactor in Studsvik.		C-14	Co-60	R2 (Studsvik)
			Fe-55	Mo-93	
			Ni-59	Nb-93m	
			Ni-63	Tc-99	

Reference Information from Sweden	Type	Measured data	Calculated data		Type of unit
Kjellbert N, 1990	The activity of activation products, heavy radionuclides and fission products in a reference BWR fuel assembly were calculated with the computer code ORIGEN		Ni-63 Co-60		Fuel assembly (BWR)
Thegerström C and Hård E, 1981	A compilation of activity content in wastes that will be stored in SFR 1.		H-3 Fe-55 Ni-59	Ni-63 Co-60	Boron plates
Alder J C and McGinnes D F, 1994	A model radioactive waste inventory has been developed for Swiss waste disposal projects		H-3 C-14 Cl-36 Fe-55 Ni-59 Ni-63	Co-60 Nb-93m Nb-94 Mo-93 Tc-99	Swiss model waste inventory
Anttila M, and Wasastjerna F,1989	The induced activity of decommissioning waste of BWR nuclear reactors has been analysed with the ORIGEN-S computer program. Data for the core grid, the moderator tank, and the moderator tank cover have been used in the SFL 3-5 evaluation.		H-3 C-14 Cl-36 Fe-55 Ni-59	Ni-63 Co-60 Nb-93m Mo-93 Tc-99	Finish BWR
Robertson D E et al. 1993	During dismantling of the US Shippingport Station a number of piping and hardware samples (BWR's and PWR's) were analysed as part of reactor ageing.	C-14	Co-60 Nb-93m		US PWR and BWR
Smith R I, et al. 1978	The radionuclides present in activation of a reference PWR were calculated with the computer code ORIGEN.		Fe-55 Ni-59 Ni-63 Co-60	Nb-93m Nb-94 Mo-93	PWR
UK Nirex Ltd, 1996.	The radionuclide inventory for a PWR, Sizewell B is reported.		Cl-36 Co-60 *)		

*) Activities for more nuclides are given in the report.

4 Evaluation and selection of data

Data from the investigations briefly described in Chapter 3 and summarised in Table 3-1 are evaluated in this chapter. The aim has been to estimate the activity for a consistent set of radionuclides (see section 2.1) for similar wastes when waste type specific information is limited or unavailable. The main part of the radionuclide activity in SFL 3-5 waste originates from nuclear power facilities and has been divided into two categories: surface contamination (CRUD) and neutron induced activity. In addition, there are waste from Research, Medicine and Industry.

For the purpose of this study Co-60, Cs-137, and Pu-239+240 have been selected as key radionuclides for estimates of the activity of other radionuclides. The gamma-emitters Co-60 and Cs-137 are often readily measured with ordinary plant equipment and therefore used as key radionuclides to which other radionuclides that are more difficult to measure can be correlated. Activation products are often correlated to Co-60 and fission products to Cs-137. In addition, measurements of Ce-144, Pu-239 and Pu-240 can be used for correlation of transuranics. The activity of Pu-239 and Pu-240 is generally reported together since the energies of their alpha particles have the same character and can therefore not easily be separated in the analyses / Thierfeldt and Deckert 1995, Ghysels 1993/.

The activity content of radionuclides in waste with surface contamination has been estimated by the use of:

- Correlation factors between difficult to measure radionuclides and easy to measure key radionuclides.
- Contamination levels of the key radionuclides on surfaces in contact with primary coolant water.
- The relation between the contamination level on surfaces in contact with primary coolant water and on surfaces in contact with other contaminated water.

The activity content of radionuclides in waste with neutron induced activity has been estimated by the use of:

- Correlation factors between difficult to measure radionuclides and the easy to measure key radionuclide Co-60.
- The activity content of the used key radionuclide Co-60 has generally been calculated and reported in literature for all the specific waste categories considered for SFL 3-5.

The strategy for the selection of correlation factors has been to compile reported data from measurements and then to calculate correlation factors from these data. The correlation factors evaluated from all reported data have been compiled in diagrams. The correlation factors evaluated from the different measurements vary over a wide interval for most of the radionuclides, often many orders of magnitude. From the performed evaluation one correlation factor representative for each radionuclide has been selected. The selected correlation factor is aimed to be representative for waste one year after discharge from the nuclear facility.

The time aspect may play an important role for estimates of correlation factors. Examples of important aspects are duration of exposure, the reference time for reported measurements and the assumed times in reported calculations. The exact reference time is not specified in all literature studied. This limitation is most important for the evaluation of correlation factors for short-lived radionuclides and for correlation of radionuclides to the relatively short-lived key radionuclide Co-60. The approach here has been to use the most conservative correlation factor evaluated if data are available at different times from the same source.

The greatest importance has been attached to the measured data, especially measurements from Swedish nuclear facilities, in the evaluation of correlation factors. One exception is the cor-

relation factors for the nuclides in the natural decay chains, where data from calculations of the inventory in a BWR spent fuel assembly has been used even though measurements of activities in Swedish nuclear facilities have been available for some of the nuclides. This has been done to achieve a consistent composition of these nuclides in the surface contamination.

Another limitation is that one general correlation factor has been selected for each radionuclide. The potential differences between different facilities and/or waste streams have not been considered. In addition, this study does not include a statistical evaluation of the correlation factors to prove if there is a correlation between the key nuclides and the other radionuclides.

Even though, constant correlation factors have been used for the estimates of the reference radionuclide inventory the results from the safety assessment will indicate where uncertainties in the used correlation factors are of major importance for the potential exposure of humans and the environment.

4.1 Surface contamination

The surface contamination, CRUD, on surfaces that have been in contact with the same water as nuclear fuel assemblies, comprises of activated corrosion products as well as fission products and heavy nuclides from leaking fuel.

The approach has been to first define a contamination level for key radionuclides on surfaces that have been in contact with primary reactor water and secondly, to define correlation factors between difficult to measure radionuclides and key radionuclides.

The radionuclide inventory for different waste types can then be calculated from measurements of key radionuclides and the correlation factors to the key nuclides. When no measurements of key radionuclides are available the radionuclide inventory can be estimated by making assumptions of the degree of contamination (%) in relation to primary surfaces, the contamination level of key nuclides (Bq/m^2) on primary surfaces and the surface areas of the considered waste (m^2).

4.1.1 Contamination levels of key radionuclides

The selection of contamination levels of the key radionuclides (Co-60, Cs-137 and Pu-239+240) on surfaces in contact with primary reactor coolant water is discussed in this section. Primary reactor coolant water is in contact with the fuel assemblies. The selection of contamination levels has mainly been based on measurements and investigations performed on Swedish nuclear reactors.

The activity of corrosion products in scrape samples from fuel rods from the different Swedish power reactors have been analysed and reported in several studies /Rundlöf 1985abcd, Troberg 1983, 1984/. The scrape samples were taken from fuel rods taken out from the reactor during the yearly reactor shutdown. From the reported data the activity of Co-60 in the CRUD seems to be within the interval of 1−20 GBq/m^2.

A summary of performed scrape tests on fuel rods indicate that the normal activity of Co-60 in CRUD in Swedish BWR's (at 20 kEFPH) is within the interval of 10−35 GBq/m^2 /Fridemo and Lundgren 1990/. The amount of Co-60 in the reactor systems does not change much from one year to another. This means that the production and deposition of fresh Co-60 compensate the decay /Aronsson 1994/.

Measurements with a mobile analyser for detection of CRUD in piping (MADAC) in different systems in Barsebäck 1 and 2 in different systems have been performed during the years 1977−1994 /Ernström 1996/, see Figures 4-1–4-2. The system with the highest contamination level of Co-60 is the cooling system for the closed reactor. The contamination level of Co-60 in this system varies between 0.07−2 GBq/m^2 in Barsebäck 1 and 0.1−4 GBq/m^2 in Barsebäck 2 during the time period. There is only one point of measuring of the contamination of Cs-137 in the same system in Barsebäck 2 which is 0.01 GBq/m^2 (year 1977).

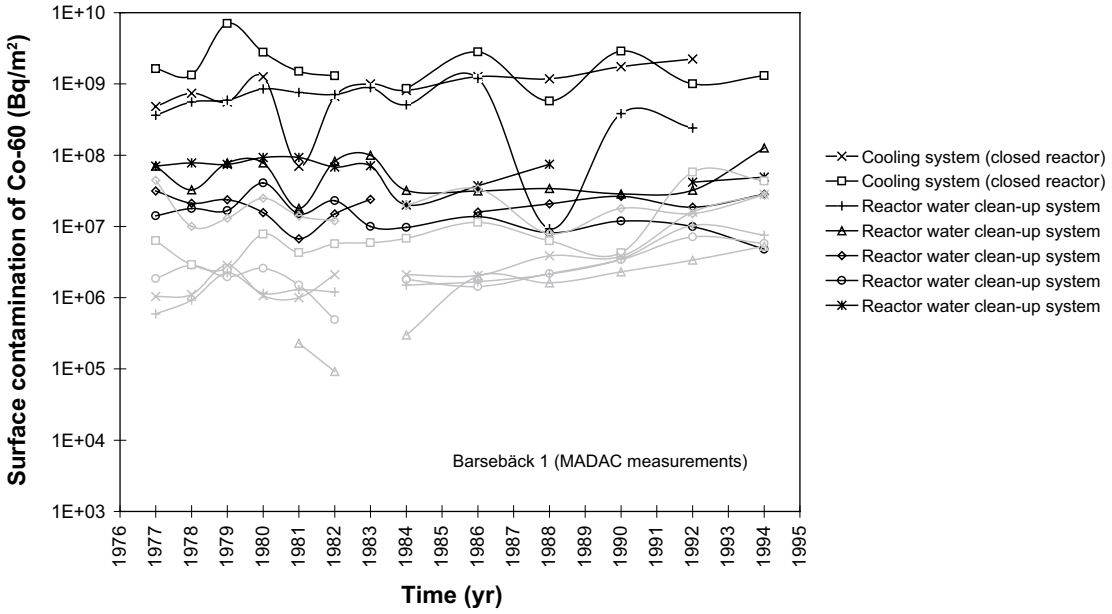

Figure 4-1. *Measured surface contamination (MADAC measurements) in Barsebäck 1 /Ernström 1995/.*

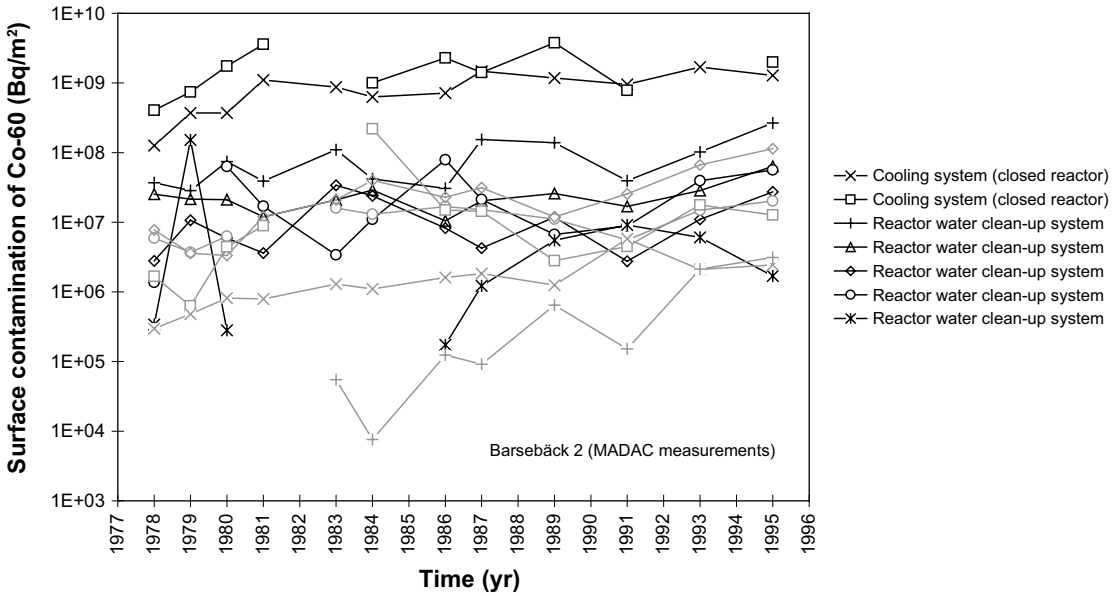

Figure 4-2. *Measured surface contamination (MADAC measurements) in Barsebäck 2 /Ernström 1995/.*

In an old decommissioning study /SKB 1979/, the surface contamination levels on primary surfaces in a reactor after 40 years of operation has been estimated. Determinations using a mobile analyser for detection of CRUD gave a Co-60 contamination level of 0.37 GBq/m^2 (10 mCi/m^2). After 40 years of operation and one year of shutdown it was conservatively estimated that this value will have increased to 1.85 GBq/m^2 (50 mCi/m^2). The surface contamination level of fission products on primary surfaces was reported to be < 10% of the Co-60 activity, i.e. < 0.185 GBq/m^2.

For an extended operation of Oskarshamn 1 to year 2010 the surface contamination level of Co-60 on primary surfaces has been calculated to be 1 GBq/m^2 /Lundgren 1991/.

According to measurements made on two Swiss PWR reactors (Beznau 1 and 2) the average contamination of the primary circuit by Co-60 was estimated to be 15 GBq/m^2 towards the end of the reactor's working life. The average activities of the radionuclides Cs-137 and Pu-239+240

were from the same reference estimated to be 10% and 0.02%, respectively, of the Co-60 activity 5 years after reactor shutdown /Smith et al. 1985/.

The surface contamination level of Pu-239+240 has been measured in scrape samples taken at different axial positions from fuel rods. This type of tests is being performed at certain time intervals on all power units in Sweden /Westerberg 1986, Granath 1983, Fridemo 1989/. The scrape samples were taken from fuel rods taken out from the reactor during the yearly reactor shutdown. The reported values vary within an interval of 0.1−30 kBq/m^2.

Selected contamination levels on primary surfaces

The surface contamination level of Co-60 on primary surfaces is selected to be 5 GBq/m^2 after 40 years of reactor operation. This value was selected mainly from the MADAC measurements in Barsebäck 1 and 2 (see Figures 4-1 and 4-2) and represents the contamination level in the most contaminated system. The measured data cover a time-period of 18 years and the surface contamination has been relatively constant during this time. This indicates that the production and deposition of fresh Co-60 compensate the decay. The selected contamination level is therefore assumed to be representative also at reactor closure. The data found in literature on contamination levels of Co-60 for different surfaces cover an interval of 1−35 GBq/m^2. The reported contamination levels on primary surfaces vary between the different power units, and in addition the contamination levels within one unit may vary with time.

The available information on Cs-137 contamination levels is relatively sparse. If it is assumed that the activity of Cs-137 is 10% of the Co-60 activity, the data evaluated for Co-60 would result in a Cs-137 contamination level of 0.1−3.5 GBq/m^2. However, one measurement on Barsebäck 2 /Ernström 1996/ indicates a level of the order of 0.01 GBq/m^2. An evaluation of the Swedish measurements on reactor internal parts in the data base compiled by /Thierfeldt and Deckert 1995/ resulted in scaling factors between Cs-137 and Co-60 in the range 0.001 to 0.1, with a median value of 0.06, see Figure 4-3 (reactor internals). Based on the median value (0.06) for the scaling factor and the contamination level defined for Co-60 (5 GBq/m^2), the contamination level of Cs-137 is selected to be 0.3 GBq/m^2.

As a representative average the contamination level for Pu-239+240 is selected to be $5 \cdot 10^{-6}$ GBq/m^2 (5 kBq/m^2), which is within the interval evaluated from scrape samples on fuel rods. This value is also supported by the scaling factors to Co-60 and Cs-137 that can be evaluated from the compilation of radionuclide data on reactor internals from Swedish reactors /Thierfeldt and Deckert 1995/, see Figure 4-3. These scaling factors is calculated to be in the range 10^{-6} to 10^{-3} for Cs-137 and in the range 10^{-7} to 10^{-6} for Co-60, see Figure 4-3.

The selected contamination level for key radionuclides on primary surfaces are given in Table 4-1 together with scaling factors between the key radionuclides calculated from the selected contamination levels. These scaling factors will be used only when information about Cs-137 and Pu-239+240 activities is lacking.

Table 4-1. Selected contamination levels of key radionuclides on primary surfaces in GBq/m^2. In addition scaling factors are calculated from the selected contamination levels.

Key radionuclide	Contamination level (GBq/m^2)	Scaling factor to Co-60	to Cs-137
Co-60	5	–	–
Cs-137	0·3	$6 \cdot 10^{-2}$	–
Pu-239+240	$5 \cdot 10^{-6}$	$1 \cdot 10^{-6}$	$1.7 \cdot 10^{-5}$

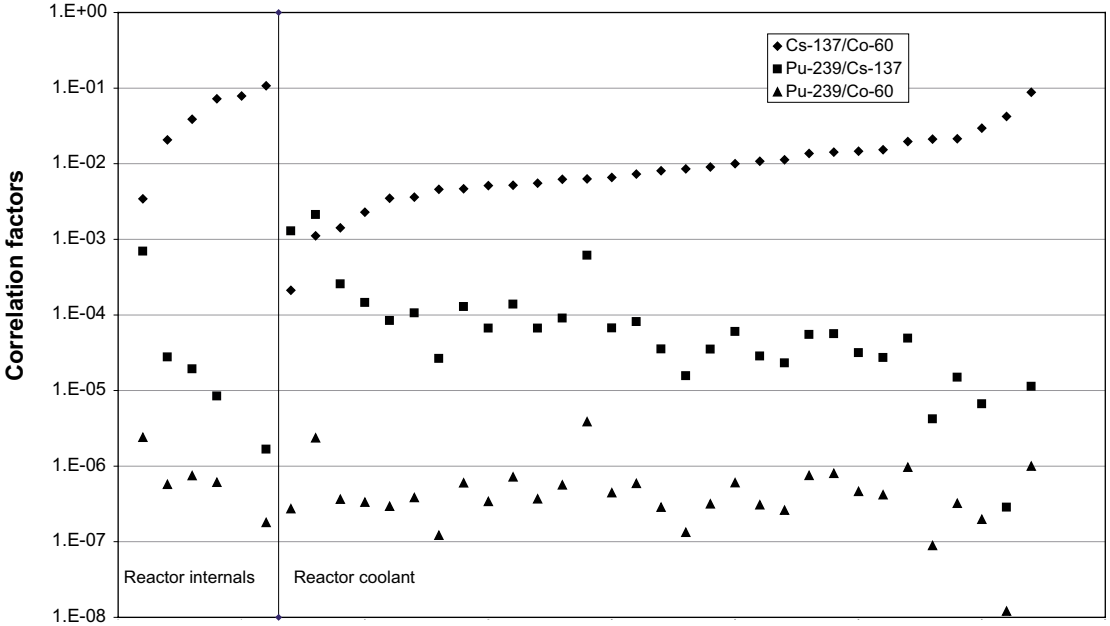

Figure 4-3. *Scaling factors between the selected key radionuclides (Co-60, Cs-137, and Pu-239+240) in reactor materials and reactor coolant.*

Selected contamination levels on secondary surfaces

An approach to estimate the contamination on secondary systems in a power plant from data on primary systems is reported by /Lundgren 1991/. The activity on surfaces in the secondary systems of a reactor was evaluated using a relative contamination level. For each secondary system a relative contamination level (i.e. relative to the primary system surfaces) was defined. The total surface contamination in secondary systems was then calculated from the:

- Surface contamination on the primary systems.
- Relative contamination levels.
- Surface areas of the secondary systems.

The relative contamination level in the fuel pools of a BWR reactor (Oskarshamn 2) was reported to be 0.03 /Lundgren 1991/, i.e. 3% of the contamination on primary systems. Here a relative contamination level of 3% of the contamination on primary systems has been selected for all other systems, to be deposited in SFL 3-5, which have been in contact with potentially contaminated water.

4.1.2 Correlation factors for surface contamination

The assumed CRUD composition is based on a compilation of available relevant information in the literature, see Chapter 3. Data are gathered from measurements of activity in scrape samples from fuel rods and decontamination solutions. It is further assumed that the composition of nuclides in surface contamination and in reactor coolant water is about the same. This means that measurements performed on reactor coolant water and ion-exchange resins in different reactor systems can be used to evaluate the composition of radionuclides in the CRUD. The assumed composition of the surface contamination is given as correlation factors, i.e. a relative value for all different radionuclides to a key radionuclide (Co-60, Cs-137 or Pu-239+240).

For some of the radionuclides in Table 2-2 no measurements are reported and the correlation factors have been estimated from values reported for a reference BWR spent fuel assembly /Kjellbert 1990/ and applicable information from other countries.

The evaluation and selection of correlation factors for each radionuclide is discussed separately with a short summary of evaluated factors from the literature. The description of each radionuclide begins with a short unnumbered table that gives the radionuclide name, the half-life, the selected correlation factor, the selected key radionuclide, and the main information source used for the selection of the correlation factor.

Radionuclides correlated to Co-60 and Cs-137

The activation products are mainly correlated to Co-60 and the fission products mainly to Cs-137.

H-3	Selected CRUD correlation factor
	$1 \cdot 10^{-4}$ H-3/Co-60
Half-life = 12 years	Data on induced activity

Data from Germany gathered by /Thierfeldt and Deckert 1995/ show that there is no correlation between H-3 and Cs-137 or Co-60. Instead, H-3 can be found in wastes if they contain moisture that is contaminated with water from the primary coolant loop /Thierfeldt and Deckert 1995/.

Data from measurements on decontamination solutions from the cleaning of two steam generators from Ågesta /Brodén 1996/ were used to calculate a correlation factor between H-3 and Co-60. The solution from cleaning of the first steam generator gave a correlation factor of 0.7, whereas, the solution from cleaning of the second steam generator gave a correlation factor of $3 \cdot 10^{-6}$.

No information on contamination of primary reactor surfaces by H-3 is reported. For the purpose of this study the same correlation factor between H-3 and Co-60 as applied for induced activity in metals is selected, i.e. $1 \cdot 10^{-4}$ (see section 4.2).

The selected correlation factor is not based on any measurements of surface contamination in Swedish power plants. However, it can be assumed that tritium plays a very limited role in the contamination of steel and concrete. Consequently wastes that consist of contaminated metal should contain only negligible amounts of tritium /Thierfeldt and Deckert, 1995/.

Be-10	Selected CRUD correlation factor
	$6 \cdot 10^{-10}$ Be-10/Co-60
Half-life = $1.6 \cdot 10^{6}$	Data for spent fuel assembly

No measured data have been found for this radionuclide in the studied literature. The correlation factor between Be-10 and Co-60 in a reference BWR spent fuel assembly /Kjellbert 1990/ can be calculated to be $6 \cdot 10^{-10}$ after a five year decay period and $3 \cdot 10^{-10}$ at fuel discharge. The correlation factor between Be-10 and Cs-137 can be calculated to be $3 \cdot 10^{-11}$ both at fuel discharge and after a five year decay period.

The correlation factor between Be-10 and Co-60 in a reference BWR spent fuel assembly after a five years decay period is used as being representative for the CRUD in the SFL 3-5 waste. The correlation to Co-60 results in a higher activity content on the waste surfaces than the correlation to Cs-137 evaluated from the fuel assembly would, supposing the selected contamination levels of Co-60 (5 GBq/m^2) and Cs-137 (0.3 GBq/m^2) in the CRUD.

No measured data on contamination on primary surfaces or in primary coolant are available and consequently the selected correlation factor is based on assumptions.

C-14	Selected CRUD correlation factor
	$1 \cdot 10^{-3}$ C-14/Co-60
Half-life = $5.7 \cdot 10^{4}$	Data for operational waste

The correlation between C-14 and Co-60 is reported in /Thegerström and Hård 1981) to be 10^{-3} in Swedish operational waste. This value is based on the assumption that 1% of C-14 in the coolant water will be found in the operational waste. As a comparison the correlation between C-14 and Co-60 in a reference BWR spent fuel assembly can be calculated to be $4 \cdot 10^{-4}$ after a five year decay period and $2 \cdot 10^{-4}$ at fuel discharge from the activity calculations reported by /Kjellbert 1990/. Correlation factors evaluated from the large database by /Thierfeldt and Deckert 1995/ that cover information from Germany and Japan show a wide distribution (from 10^{-4} to 10^{6}). /Robertson et al. 1993/ reported radionuclide concentrations, analysed from primary and secondary coolant piping from the US nuclear power station Shippingport (light-water breeder reactor core, typical of a commercial pressurised water reactor core). These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between C-14 and Co-60, and the correlation factors vary within a large interval, $5 \cdot 10^{-8}$–$6 \cdot 10^{-1}$, with a median value of $2 \cdot 10^{-5}$.

The correlation factor between C-14 and Co-60 is selected to be $1 \cdot 10^{-3}$ from data on Swedish operational waste.

The correlation factor for C-14 is very uncertain. No measurements from Swedish nuclear power units are available and other available data vary with many orders of magnitude. The chemical species of C-14 are not constant in the water systems and, consequently, the correlation factors show high dispersion within different wastes /Ghysels 1993/.

Cl-36	Selected CRUD correlation factor
	$1 \cdot 10^{-5}$ Cl-36/Cs-137
Half-life = 3.0·105 years	Swiss measurements on resins and reactor coolant water

The correlation factor between Cl-36 and Cs-137 has been evaluated based on measurements in reactor coolant water and resins from the reactor coolant cleanup system in Swiss nuclear power plants /Xinqi et al. 1991/. The correlation factor reported for a reactor coolant water sample from a Swiss PWR was $5.8 \cdot 10^{-5}$. The correlation factors reported for resins were $1.4 \cdot 10^{-6}$ and $2.2 \cdot 10^{-6}$ in PWR resins and $1.7 \cdot 10^{-5}$ in BWR resins. The water chemistry has a major influence on the Cl-36 formation.

The correlation factor between Cl-36 and Cs-137 is selected to be $1 \cdot 10^{-5}$, based on the Swiss data /Xinqi et al. 1991/ described above.

The correlation between Cl-36 and Co-60, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Kjellbert 1990/, is $1 \cdot 10^{-5}$ after a five year decay period and $5 \cdot 10^{-6}$ at fuel discharge. This value would result in a higher contamination of Cl-36 on the surfaces of the SFL 3-5 waste than the selected correlation factor between Cl-36 and Cs-137, supposing the selected contamination levels of Co-60 (5 GBq/m^2) and Cs-137 (0.3 GBq/m^2) in the CRUD. However, the correlation between Cl-36 and Co-60 in a spent fuel assembly is judged to be even more uncertain than the measured data in the Swiss power plant.

Fe-55	Selected CRUD correlation factor
	1 Fe-55/Co-60
Half-life = 2.7 years	Data for operational waste

The average correlation between Fe-55 and Co-60 used to calculate the activity in Swedish operational waste was reported to be 0.7 /Thegerström and Hård 1981, Robertson et al. 1993/ reported radionuclide concentrations, analysed from primary and secondary coolant piping from the US nuclear power station Shippingport (light-water breeder reactor core, typical of a commercial pressurised water reactor core). These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Fe-55 and Co-60, and the correlation factors vary within the interval $7 \cdot 10^{-2}$–40 with a median value of 0.3. The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven nuclear power units (BWR's and PWR's) in the US /Robertson et al. 1984/. The average activities reported from these measurements were used to calculate a correlation factor of 5. The correlation factor between Fe-55 and Co-60 calculated from the activities in a reference BWR spent fuel assembly /Kjellbert 1990/ is 1 at discharge and 0.7 after a decay period of five years.

The correlation factor between Fe-55 and Co-60 is selected to be 1, mainly based on the correlation factor used for Swedish operational waste /Thegerström and Hård 1981/.

The selected correlation factor between Fe-55 and Co-60 is not supported by any specific measurements of the activity of Fe-55 in Swedish power plants. However, due to the short half-life of Fe-55 the selection is not critical for the long-term safety of a repository since the activity content of Fe-55 will certainly have decayed significantly at the time of closure of the SFL 3-5 repository.

Ni-59	Selected CRUD correlation factor
	$1 \cdot 10^{-3}$ Ni-59/Co-60
Half-life = $7.5 \cdot 10^{4}$ years	See Figure 4-4

The content of Ni-59 in water from Swedish PWR reactors has been measured and reported /Roos 1994/ (the same data are included in the Swedish database of Thierfeldt and Deckert, 1995/. These data were used to calculate correlation factors between Ni-50 and Co-60 (6 data sets). The correlation factors fall within the interval $4 \cdot 10^{-4}$–$4 \cdot 10^{-3}$ with a median value of $1 \cdot 10^{-3}$. /Lundgren 1991/ has calculated the inventories of activated corrosion products in the pool water system and in the fuel pools in a BWR. The reported data were used to calculate a correlation factor between Ni-59 and Co-60 of $7 \cdot 10^{-4}$ one year after shutdown. A correlation factor of 10^{-3} was evaluated based on information from Swedish operational waste /Thegerström and Hård 1981/. The correlation factors calculated from data reported for Swedish waste or power units are compiled in Figure 4-4.

The contamination level in a US reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. By using these data a correlation factor of 10^{-4} after five years of decay could be calculated.

Radionuclide concentrations analysed from primary and secondary coolant piping from the US nuclear power station Shippingport was reported by /Robertson et al. 1993/. These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Ni-59 and Co-60, and the factors vary within the interval $3 \cdot 10^{-3}$–0.6, with a median value of $6 \cdot 10^{-3}$. The concentration of radionuclide on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activities reported from these measurements is $1 \cdot 10^{-3}$.

Based on the distribution of correlation factors depicted in Figure 4-4 (Swedish data) the correlation factor between Ni-59 and Co-60 is selected to be $1 \cdot 10^{-3}$. The correlation factor between Ni-59 and Co-60 calculated from the activities in a reference BWR spent fuel assembly /Kjellbert 1990/ is $5 \cdot 10^{-4}$ at discharge and $1 \cdot 10^{-3}$ after a decay period of five years. The selected correlation factor is also within the interval of correlation factors evaluated from US power units as described above which, however, indicates that the interval is wide and the correlation may be underestimated with more than two orders of magnitude.

Ni-63	Selected CRUD correlation factor
	0.2 Ni-63/Co-60
Half-life = 96 years	see Figure 4-5

The content of Ni-63 and Co-60 was measured in reactor coolant water of Swedish BWR's /Roos 1994/ and from these data a correlation factor has been calculated for each of the 48 data sets. The calculated correlation factors fall within the interval 10^{-2}–5 with a median value of 10^{-1}. The Swedish database of /Thierfeldt and Deckert 1995/ comprises the same data as described above. The inventory of activated corrosion products in pool water system and fuel pools system in a BWR was calculated by /Lundgren 1991/. From these data a correlation factor between Ni-63 and Co-60 of 0.1 one year after shutdown could be calculated. Data for Swedish operational waste /Thegerström and Hård 1981/ were used to calculate a correlation factor of 0.1. The correlation factors evaluated from Swedish information sources are compiled in Figure 4-5.

The contamination level in a US reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. By using these data a correlation factor of 0.02 after five years of decay can be calculated. Radionuclide concentrations analysed from primary and secondary coolant piping, from the US nuclear power station Shippingport was reported by /Robertson et al. 1993/. These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Ni-63 and Co-60, and the correlation factors vary within the interval 0.3–9 with a median value of 0.9. The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activities reported from these measurements was 0.1.

Based on the distribution of correlation factors depicted in Figure 4-5 (Swedish data) the correlation factor between Ni-63 and Co-60 is selected to be 0.2. The correlation factor between Ni-63 and Co-60 calculated from the activities in a reference BWR spent fuel assembly /Kjellbert 1990/ is 0.08 at discharge and 0.2 after a decay period of five years. The selected correlation factor between Ni-63 and Co-60 is mainly based on measurement in reactor coolant, and is well supported by other gathered information. The ratios evaluated from all discussed data are within the interval 0.01–9.

Se-79	Selected CRUD correlation factor
	$4 \cdot 10^{-6}$ Se-79/Cs-137
Half-life = $6.5 \cdot 10^{4}$ years	spent fuel assembly

No information on measurements of Se-79 was found in the studied literature. The relation between calculated activities of Se-79 and Cs-137 in a reference BWR spent fuel assembly /Kjellbert 1990/ was calculated to be $4 \cdot 10^{-6}$ at fuel discharge and after a decay period of five year. The total activity of Se-79 and Cs-137 was reported for all the Swiss reactor waste /Alder and McGinnes 1994/. From these total activities a correlation factor of $8 \cdot 10^{-6}$ could be calculated.

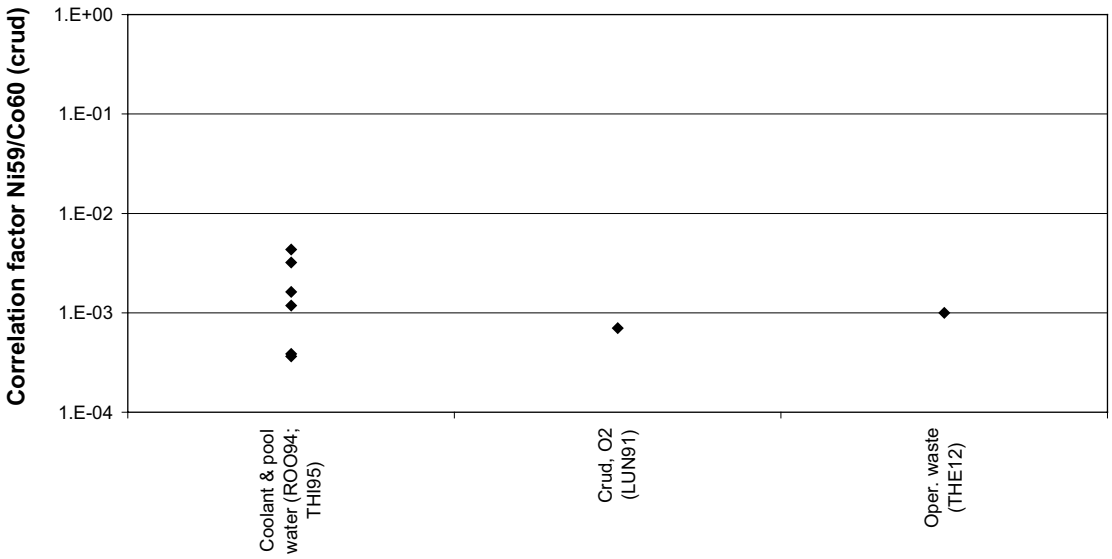

Figure 4-4. *Correlation factors evaluated between Ni-59 and Co-60 for surface contamination (CRUD).*

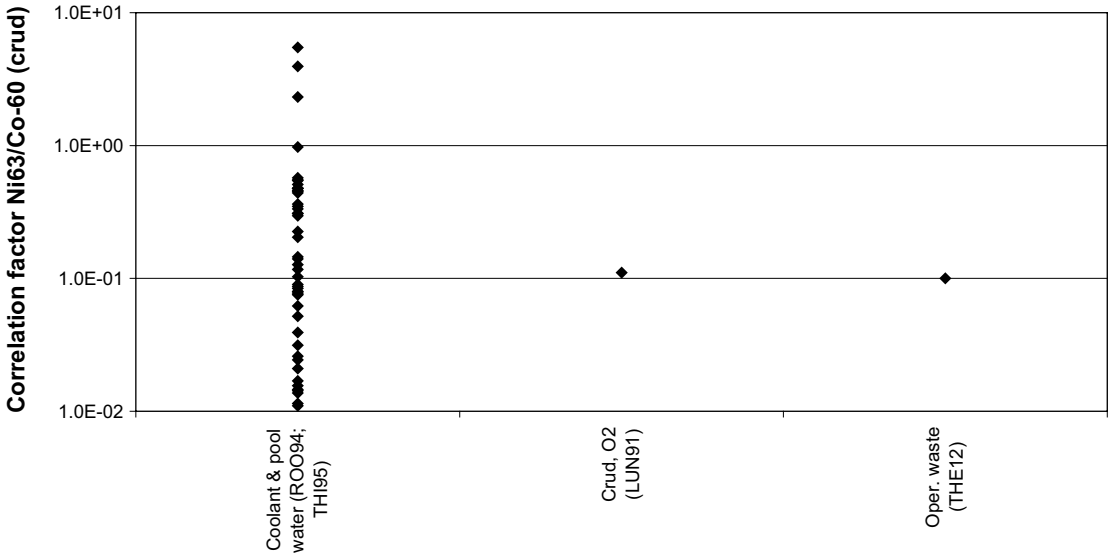

Figure 4-5. *Correlation factors evaluated between Ni-63 and Co-60 for surface contamination (CRUD).*

The correlation between Se-79 and Cs-137 is selected to be $4 \cdot 10^{-6}$ from the relation in a Swedish reference BWR spent fuel assembly.

No measurements on the contamination of Se-79 on primary surfaces or in reactor coolant are available. That means that the selected correlation factor is very uncertain.

Sr-90	Selected CRUD correlation factor
	0.1 Sr-90/Cs-137
Half-life = 29 years	see Figure 4-6

The concentration of Sr-90 and Cs-137 has been measured in reactor coolant water in Ringhals 1 /Liljenzin and Johnson 1985/. From these data a correlation factor within the interval of $2 \cdot 10^{-3}$–$3 \cdot 10^{-1}$ was evaluated, see Figure 4-6. The median value was 10^{-2}. The database used by /Thierfeldt and Deckert 1995/ comprises the same Swedish database except for some data points. /Lundgren 1991/ has calculated the inventories of fission products in the pool water system in a BWR. These data were used to calculate a correlation factor between Sr-90 and Cs-137 of 1, one year after shutdown.

From a study reporting contamination in a US PWR /Smith et al. 1985/, a correlation factor of 0.5 after five year of decay was calculated. The same correlation factor, 0.5, was calculated based on the total activity reported for all the Swiss reactor waste /Alder and McGinnes 1994/. The concentration of radionuclide on piping exposed to primary reactor coolant has been measured at seven US nuclear power stations (BWR's and PWR's) by /Robertson et al. 1984/. The correlation factor between Sr-90 and Cs-137 was calculated to be 0.06 from the average activities reported.

The activities in a reference BWR spent fuel assembly have been calculated at fuel discharge /Kjellbert 1990/ and after a five year decay period /Kjellbert 1990, Smith et al. 1985/. The corresponding correlation factor was evaluated to be 0.7.

The correlation factor between Sr-90 and Cs-137 is selected to be 0.1. This value is ten times higher than the median correlation factor value evaluated from the measurements in Ringhals 1, but lower than most ratios evaluated from the other references including also the spent fuel assembly.

The interval of correlation factors from the Swedish measurements is $2 \cdot 10^{-3}$–$3 \cdot 10^{-1}$, which covers almost all other data reported for Sr-90. The numbers of data points are sparse, but the selected value seems reasonable.

Zr-93	**Selected CRUD correlation factor**
	$1 \cdot 10^{-6}$ Zr-93/Co-60
Half-life = $1.5 \cdot 10^{6}$ years	spent fuel assembly

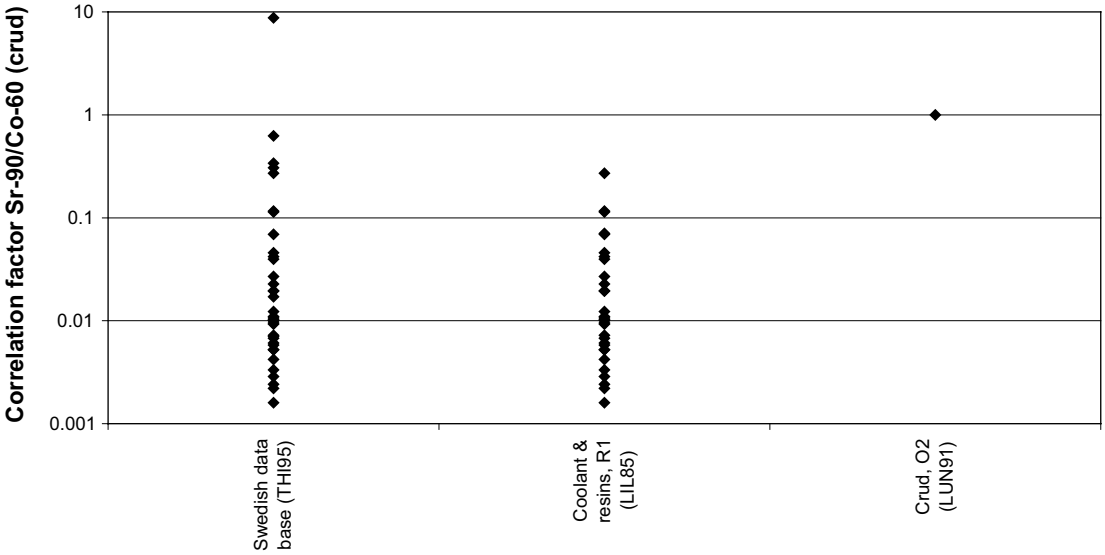

Figure 4-6. *Correlation factors evaluated between Sr-90 and Cs-137 for surface contamination (CRUD).*

No measurements on contamination of Zr-93 in primary systems or reactor coolant have been found in the studied literature. The main source for Zr-93 is activation of zirconium containing materials and activation and reactions in the fuel.

Information about contamination in a US reference PWR, reported by /Smith et al. 1985/, was used to calculate a correlation factor of $1 \cdot 10^{-6}$ after five years of decay. The correlation factor between Zr-93 and Co-60 is selected to be 10^{-6} based on this single reference.

Zr-93 in a spent fuel assembly is mainly formed due to fission in the fuel. The correlation between Zr-93 and Cs-137 was calculated to be $2 \cdot 10^{-5}$ at fuel discharge and after a decay period of 5 years from the activities in a reference BWR spent fuel assembly as reported by /Kjellbert 1990/. This value would result in roughly the same contamination (Bq/m^2) of Zr-93 on the surfaces of the SFL 3-5 waste as the selected correlation factor between Zr-93 and Co-60 will, supposing the selected contamination levels of Co-60 (5 GBq/m^2) and Cs-137 (0.3 GBq/m^2) in the CRUD.

No measurements of the contamination of Zr-93 on primary surfaces or in reactor coolant are available and the selected correlation factor is based on one single data that makes the selected value very uncertain.

Nb-93m	Selected CRUD correlation factor
	$1 \cdot 10^{-3}$ Nb-93m/Co-60
Half-life = 14 years	correlation to Nb-94

No measured data on contamination of Nb-93 in primary systems or reactor coolant of Swedish nuclear power units have been found.

The correlation between Nb-93m and Co-60, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Kjellbert 1990/ is $3 \cdot 10^{-6}$ at fuel discharge and $2 \cdot 10^{-5}$ after a five year decay period.

/Robertson et al. 1993/ reported radionuclide concentrations, analysed from primary and secondary coolant piping from the US nuclear power station Shippingport. These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Nb-93m and Co-60, and the correlation factors vary within the interval $2 \cdot 10^{-3}$–3, with a median value of $7 \cdot 10^{-3}$.

The correlation factor between Nb-93m and Co-60 is selected to $1 \cdot 10^{-3}$. The selected value is at the lower end of the interval evaluated from the US references, but the selected value is in agreement with the correlation factor selected for neutron induced activity (see section 4.2).

Nb-94	Selected CRUD correlation factor
	$1 \cdot 10^{-5}$ Nb-94/Co-60
Half-life = $2.0 \cdot 10^{4}$ years	Data for spent fuel assembly

No measured data from Swedish nuclear power reactors have been found in the studied literature. The only Swedish information is an average correlation factor of 10^{-6} between Nb-94 and Co-60 assumed for operational waste /Thegerström and Hård 1981/. The correlation between Nb-94 and Co-60, evaluated from the activity data calculated for a reference BWR spent fuel assembly /Kjellbert 1990/ is $2 \cdot 10^{-5}$ at fuel discharge and $5 \cdot 10^{-5}$ after a five year decay period.

Radionuclide concentrations, analysed from primary and secondary coolant piping from the US nuclear power station Shippingport were reported by /Robertson et al. 1993/. These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Nb-94 and Co-60. These correlation factors vary within the interval $2 \cdot 10^{-5}$–0.1 with a median value of $6 \cdot 10^{-5}$. The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven US nuclear power stations (BWR's and PWR's) /Robertson et al. 1984/. The correlation factor calculated from the average activity reported from these measurements was $3 \cdot 10^{-5}$. The reported contamination levels in a US reference PWR established by comparing experimental and theoretical values /Smith et al. 1985/ were used to calculate a correlation factor of $1 \cdot 10^{-5}$ after a five year decay period.

The correlation factor between Nb-94 and Co-60 is selected to be $1 \cdot 10^{-5}$. The selected value is at the lower end of the interval evaluated from the US references, but the selected value is in agreement with the correlation factor selected for neutron induced activity (see section 4.2).

Mo-93	**Selected CRUD correlation factor**
	$5 \cdot 10^{-6}$ Mo-93/Co-60
Half-life = $3.5 \cdot 10^{3}$ years	Induced activity

No information on measured data on the activity of Mo-93 in CRUD has been found in the literature. Therefore, the same correlation factor to Co-60 as evaluated for induced activity is selected (see section 4.2). This correlation factor is $5 \cdot 10^{-6}$. The correlation between Mo-93 and Co-60 was calculated to be $4 \cdot 10^{-8}$ at fuel discharge and $8 \cdot 10^{-8}$ after a 5 years decay period from reported activities in a reference BWR spent fuel assembly /Kjellbert 1990/. This value is lower than the selected correlation factor, but is not judged to be representative for the composition of surface contamination.

The selected correlation factor is not supported by any relevant measurements and is therefore very uncertain.

Tc-99	**Selected CRUD correlation factor**
	$5 \cdot 10^{-3}$ Tc-99/Cs-137
Half-life = $2.1 \cdot 10^{5}$ years	see Figure 4-7

The content of Tc-99 and Cs-137 was measured in reactor coolant water from Swedish BWR's and PWR's /Roos 1994/. Calculated correlation factors based on these data (45 points of measuring) fall within an interval of $5 \cdot 10^{-5}$–1, and the median value is $5 \cdot 10^{-3}$. By using the Swedish database in /Thierfeldt and Deckert 1995/ to calculate correlation factors an interval of $8 \cdot 10^{-7}$–$1 \cdot 10^{-2}$ was obtained, with a median value of $2 \cdot 10^{-4}$. The ratio between Tc-99 and Cs-137 was reported to be $1 \cdot 10^{-4}$ in spent fuel in /Thegerström and Hård 1981/.

Correlation factors evaluated from the Swedish information sources are compiled in Figure 4-7.

The contamination level in a US reference PWR, established by comparing experimental and theoretical values, was reported in /Smith et al. 1985/. These data were used to calculate a correlation factor of 10^{-4} after five year of decay. The concentration of radionuclides on piping exposed to primary reactor coolant have also been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor between Tc-99 and Cs-137 calculated from the average activities reported from these measurements was $3 \cdot 10^{-3}$. The average activity of Tc-99 in all reactor internals, reported in the Swiss model inventory /Alder and McGinnes 1994/, has been used to calculate an average correlation factor between Tc-99 and Cs-137 of $5 \cdot 10^{-3}$.

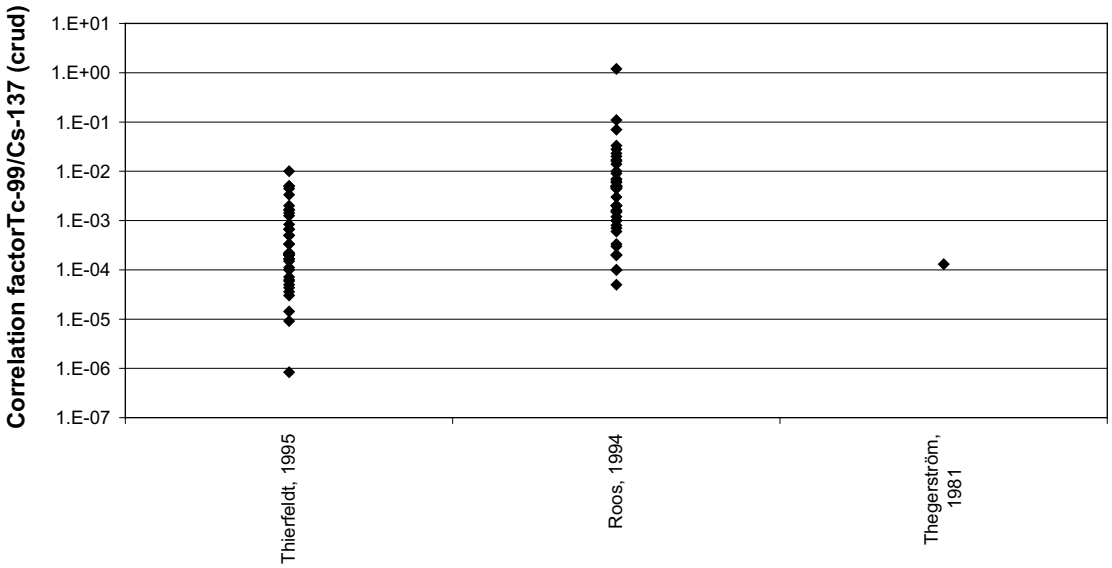

Figure 4-7. *Correlation factors evaluated between Tc-99 and Cs-137 for surface contamination (CRUD).*

The selected value for the correlation factor between Tc-99 and Cs-137 is $5 \cdot 10^{-3}$ based on the Swedish measurements, but also well supported by both the US and Swiss data.

The correlation between Tc-99 and Cs-137, was calculated to be $1 \cdot 10^{-4}$ from reported activities in a reference BWR spent fuel assembly at fuel discharge and after a decay period of 5 years /Kjellbert 1990/. This value supports the selected correlation factor since the leakage of technetium from the fuel is reported to be 10% of the activity leakage of Cs-137 /Thegerström and Hård 1981/.

Pd-107	**Selected CRUD correlation factor**
Half-life = $6.5 \cdot 10^{6}$ years	$1 \cdot 10^{-6}$ Pd-107/Cs-137 spent fuel assembly

No information on measured contamination of Pd-107 has been found in the literature. The correlation factor between Pd-107 and Cs-137, calculated for a reference BWR spent fuel assembly /Kjellbert 1990/, is $1 \cdot 10^{-6}$ at fuel discharge and after a five year decay period. The average activity of Pd-107 in all reactor internals, reported in the Swiss model inventory /Alder and McGinnes 1994/, has been used to calculate an average correlation factor between Pd-107 and Cs-137 of $1 \cdot 10^{-6}$.

The correlation factor between Pd-107 and Cs-137 is selected to be $1 \cdot 10^{-6}$, based on data for a spent fuel assembly.

The selection of the correlation factor for Pd-107 is not based on any measurements of contamination on primary surfaces and is therefore judged to be very uncertain.

Ag-108m	Selected CRUD correlation factor
	$6 \cdot 10^{-5}$ Ag-108m/Co-60
Half-life = $1.3 \cdot 10^{2}$ years	spent fuel assembly

No data on measured surface contamination of Ag-108m have been found in literature. The correlation factor between Ag-108m and Co-60, evaluated from activity data reported for a reference BWR spent fuel assembly /Kjellbert 1990/ was $3 \cdot 10^{-5}$ at fuel discharge and $6 \cdot 10^{-5}$ after a five year decay period,. From the same data the correlation factor between Ag-108m and Cs-137 was calculated to $3 \cdot 10^{-10}$.

The correlation factor between Ag-108m and Co-60 in a spent fuel assembly is selected as representative for the CRUD on SFL 3-5 waste. This correlation results in more surface contamination of Ag-108m than the correlation to Cs-137, supposing that the selected contamination levels of Co-60 (5 GBq/m^2) and Cs-137 (0.3 GBq/m^2) are correct.

The correlation for Ag-108m is not based on any measurements of the contamination on primary surfaces or in reactor coolant and is therefore very uncertain.

Cd-113m	Selected CRUD correlation factor
	$6 \cdot 10^{-4}$ Cd-113m/Cs-137
Half-life = 14 years	Spent fuel assembly

No measured data on Cd-113m have been found in the literature studied. The correlation between Cd-113m and Cs-137 was calculated to be $6 \cdot 10^{-4}$ at fuel discharge and $5 \cdot 10^{-4}$ after a 5 years decay period from activities reported for a reference BWR spent fuel assembly /Kjellbert 1990/.

In the Swiss model inventory /Alder and McGinnes 1994/, Cd-113m was reported in one single waste type named “Brennelementkasten”. Further, it was reported that this waste type contains considerable amount of CRUD. From these data a correlation factor between Cd-113m and Cs-137 of $3 \cdot 10^{-4}$ could be calculated. The content of Cd-113m in Swedish operational waste is reported to be negligible /Thegerström and Hård 1981/.

The correlation factor between Cd-113m and Cs-137 is selected to be $6 \cdot 10^{-4}$ based mainly on data for a spent fuel assembly, but also in agreement with the Swiss information.

Sn-126	Selected CRUD correlation factor
	$5 \cdot 10^{-7}$ Sn-126/Cs-137
Half-life = $1.0 \cdot 10^{5}$ years	Swiss model inventory, reactor internals

No measured data for Sn-126 have been found in the literature studied. The average activities of Sn-126 and Cs-137 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ have been used to calculate an average correlation factor of $5 \cdot 10^{-7}$. This value is selected as the correlation factor for the SFL 3-5 waste, since it is judged that the major activity of both these radionuclides originates from fuel leakages. The same correlation factor was obtained when the reported activity data for specific Swiss operational waste types containing ion-exchange resins “Harze” /Alder and McGinnes 1994/ were used.

The correlation between Sn-126 and Cs-137 is calculated to be $8 \cdot 10^{-6}$ at fuel discharge and $9 \cdot 10^{-6}$ after a 5 years decay period from activities reported for a reference BWR spent fuel assembly /Kjellbert 1990/. This value supports that the selected value is at least reasonable since the leakage of Sn-126 from the fuel is reported to be lower than 0.1% of the leakage of Cs-137 /Thegerström and Hård 1981/.

Sb-125	Selected CRUD correlation factor
	0.1 Sb-125/Co-60
Half-life = 2.8 years	see Figure 4-8

The available data from measurements in Swedish reactors can only be correlated to Co-60 since there is no comparable information on Cs-137. Data reported from scrape tests on fuel rods from Swedish BWR's /Fridemo et al. 1985/ were used to evaluate an interval for the correlation factor between Sb-125 and Co-60 of 0.06−7, with a median value of 4. Data from MADAC-measurements in Barsebäck 1 and 2 /Ernström 1996/ were used to calculate an interval of the correlation factor of 0.01−1, with a median value of 0.1. Analyses of the activity in primary circuit ion-exchange resins in Oskarshamn 3 /Skålberg 1987/ were used to calculate an interval of the correlation factor between Sb-125 and Co-60 of 0.005−0.3, with a median value of 0.01. These measurements on ion-exchange resins were also included in the database of /Thierfeldt and Deckert 1995/, but the data were not discussed in the report. Correlation factors evaluated from Swedish information sources are given in Figure 4-8.

From data reported on contamination in a US reference PWR /Smith et al. 1985/ a correlation factor of 0.1 after five year of decay was calculated.

The correlation factor between Sb-125 and Co-60 is selected to be 0.1, based mainly on the measured Swedish data given in Figure 4-8. The concentration of Sb-125 is often at least ten times lower than the concentration of Co-60 in operational waste /Thegerström and Hård 1981/. However, due to the short half-life of Sb-125 the selection is not critical for the long term-safety of a repository since the activity content of Sb-125 will certainly have decayed significantly at the time of closure of the SFL 3-5 repository.

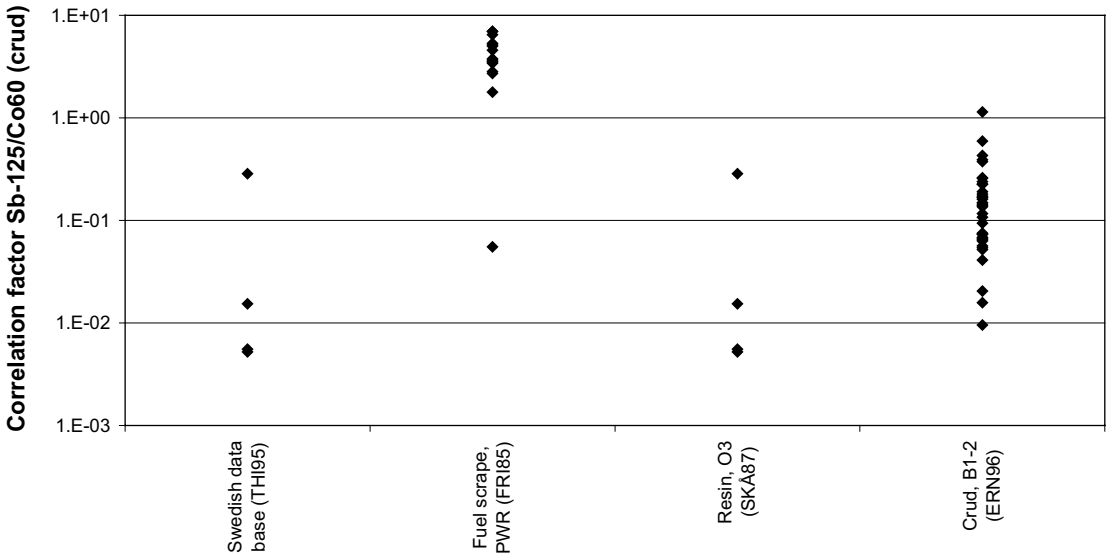

Figure 4-8. *Correlation factors evaluated between Sb-125 and Co-60 for surface contamination (CRUD).*

I-129	Selected CRUD correlation factor
	$3 \cdot 10^{-7}$ I-129/Cs-137
Half-life = $1.6 \cdot 10^{7}$ years	spent fuel assembly

From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between I-129 and Cs-137 was calculated to be $3 \cdot 10^{-7}$ at fuel discharge and after a 5 years decay period. The activities of I-129 and Cs-137 were reported for core components and internal parts in /Thegerström and Hård 1981/. From these data a correlation factor of $4 \cdot 10^{-7}$ could be calculated. In the same reference /Thegerström and Hård 1981/ the correlation factor in ion-exchange resins was reported to be $3 \cdot 10^{-7}$.

The average activity of I-129 in all reactor internals that was reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate an average correlation factor between I-129 and Cs-137 to be $2 \cdot 10^{-4}$. The concentration of radionuclides on piping exposed to primary reactor coolant has been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activity reported from these measurements was $3 \cdot 10^{-5}$.

The correlation factor between I-129 and Cs-137 is selected to be $3 \cdot 10^{-7}$, based mainly on data from the spent fuel assembly, but also supported by the old data used for Swedish radioactive waste /Thegerström and Hård 1981/.

In /Thierfeldt and Deckert 1995/ activity data from Germany and Japan are given. These data were used to calculate correlation factors between I-129 and Cs-137 and the ratios vary within a wide range, $2 \cdot 10^{-8}$−10. Iodine and cesium have similar transport properties in reactor systems and release mechanisms from reactor fuel, but they have different retention properties in the filter systems. In addition, different correlation behaviour may exist for the ranges of low and high specific activities /Thierfeldt and Deckert 1995/.

Cs-134	Selected CRUD correlation factor
	1 Cs-134/Cs-137
Half-life = 2.1 years	see Figure 4-9

The concentration of Cs-134 and Cs-137 has been measured in reactor coolant water in Ringhals 1 /Liljenzin and Johnson 1985/. From these data (42 points of measuring) an interval of 0.06−14 were evaluated for the correlation factor with a median value of 1. /Thierfeldt and Deckert 1995/ used the same database. The content of Cs-134 and Cs-137 was also measured in reactor coolant water in Forsmark (3 points of measuring) /Roos 1994/. Correlation factors calculated from these data fall within an interval of 0.4−0.6, with a median value of 0.5. /Skålberg 1987/ reported data from analyses of ion-exchange resins in Oskarshamn 3 that gave a correlation factor of 0.4. Evaluation of the radionuclide concentrations, measured in decontamination solutions from cleaning of a steam generator from Ågesta /Brodén 1993/ was used to calculate a correlation factor of 0.07. The time for the measurements was unknown to the author, and the low value may be due to decay of Cs-134. The inventory of fission products in pool water system in a BWR was calculated in /Lundgren 1991/ for a fuel leakage scenario. Evaluation of the results gave a correlation factor between Cs-134 and Co-60 of 0.7 one year after shutdown. In /Thegerström and Hård 1981/ it was stated that measured activities of Cs-134 and Cs-137 in Swedish waste were about the same. Correlation factors evaluated from Swedish data are compiled in Figure 4-9.

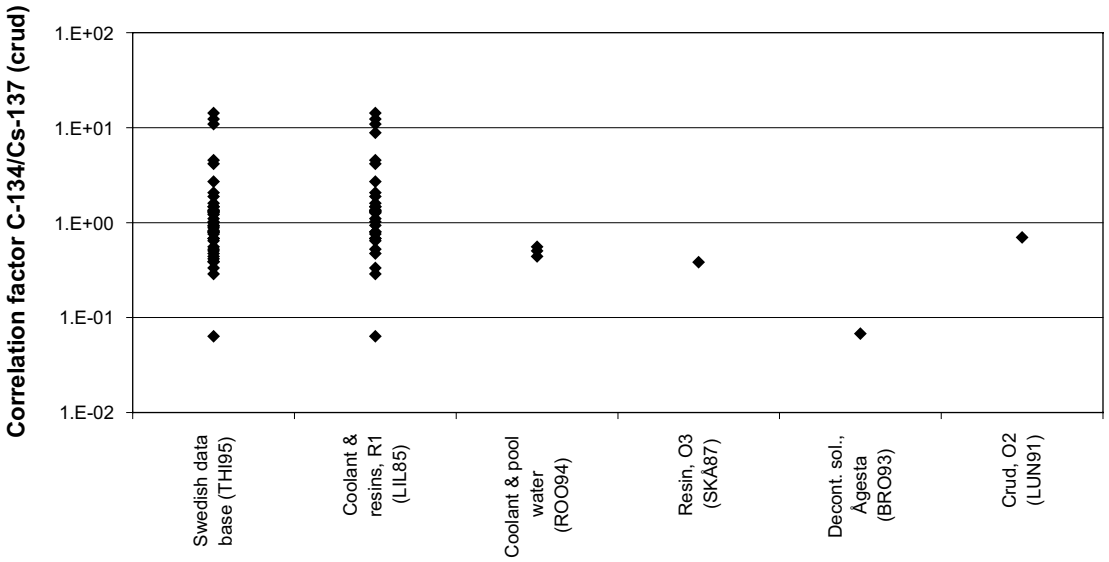

Figure 4-9. *Correlation factors evaluated between Cs-134 and Cs-137 for surface contamination (CRUD).*

Evaluation of contamination data, reported for a US reference PWR /Smith et al. 1985/, gave a correlation factor of 0.4 after five year of decay. The concentration of radionuclides on piping exposed to primary reactor coolant has been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activity reported from these measurements was 0.5. The average activity of Cs-134 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate an average correlation factor between Cs-134 and Cs-137 to 0.02. The time for the reported data was specified as "the end year of the waste package production period".

The correlation factor between Cs-134 and Cs-137 is selected to be 1, based mainly on the data compiled in Figure 4-9. However, the other references support the selected value. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Cs-134 and Cs-137 was calculated to be 2 at fuel discharge and 0.3 after five years of decay.

Cs-135	Selected CRUD correlation factor
Half-life = $2.3 \cdot 10^6$ years	$5 \cdot 10^{-6}$ Cs-135/Cs-137 spent fuel assembly

Cs-135 and Cs-137 are both single neutron fission products and a simple linear correlation between them should be expected /Thierfeldt and Deckert 1995, Thegerström and Hård 1981/ report a correlation factor of $3 \cdot 10^{-6}$ in operational waste (ion-exchange resins). /Thierfeldt and Deckert 1995/ reported some measurements (number of 5) from which an average correlation factor of $8 \cdot 10^{-6}$ could be calculated. Evaluation of contamination data from a US reference PWR /Smith et al. 1985/ gave a correlation factor of $3 \cdot 10^{-6}$ after five years of decay. The average activity of Cs-135 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate the average correlation factor between Cs-135 and Cs-137 to be $7 \cdot 10^{-6}$. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Cs-135 and Cs-137 was calculated to be $4 \cdot 10^{-6}$ at fuel discharge and $5 \cdot 10^{-6}$ after a five year decay period. A theoretical value for the ratio between Cs-135 and Cs-137 has been reported to be $1 \cdot 10^{-5}$ by /Roos 1994/.

From the compiled data the correlation factor between Cs-135 and Cs-137 was selected to be $5 \cdot 10^{-6}$. The selected value is supported by a reported theoretical value as well as by the correlation calculated for a spent fuel assembly**.**

Ba-133	**Selected CRUD correlation factor**
	$1 \cdot 10^{-5}$ Ba-133/Co-60
Half-life = 11 years	Swiss model inventory, reactor internals

The information on contamination levels of Ba-133 is very sparse. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Ba-133 and Cs-137 was calculated to be $1 \cdot 10^{-6}$ at fuel discharge and after a five year decay period. The average activity of Ba-133 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate the average correlation factor between Ba-133 and Co-60 to be $1 \cdot 10^{-5}$. The average activities reported in the Swiss model inventory for Ba-133 and Co-60 are the sum of surface deposited activity and activation. It is expected that the activity of Co-60 in reactor internals is dominated by activation. This means that the evaluated average correlation factor is very uncertain.

The correlation factor between Ba-133 and Co-60 is selected to be $1 \cdot 10^{-5}$, which is the highest value of those given above.

The selected correlation factor for Ba-133 is rough but, due to the relatively short half-life of Ba-133, the selection is not very important for the long-term safety of a repository.

Pm-147	**Selected CRUD correlation factor**
	0.9 Pm-147/Cs-137
Half-life = 2.6 years	spent fuel assembly

No information on measured contamination levels of Pm-147 has been found in the literature studied. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Pm-147 and Cs-137 was calculated to be 0.9 at fuel discharge and 0.3 after a five year decay period. The average activities of Pm-147 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ have been used to calculate an average correlation factor between Pm-147 and Cs-137 of 0.02. Evaluation of contamination data from a US reference PWR /Smith et al. 1985/ gave a correlation factor of 1 five years after shutdown.

The correlation factor between Pm-147 and Cs-137 is selected to be 0.9, based on data for a spent fuel assembly.

The selection of the correlation factor of Pm-147 is not very sensitive since its half-life is short, and Pm-147 will probably have decayed to Sm-147 (half-life = $1 \cdot 10^{11}$ years) at the time of closure of the SFL 3-5 repository.

Sm-151	**Selected CRUD correlation factor**
	$3 \cdot 10^{-3}$ Sm-151/Cs-137
Half-life = 90 years	spent fuel assembly

No data on measured contamination levels of Sm-151 were found in the literature studied. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Sm-151 and Cs-137 was calculated to be $3 \cdot 10^{-3}$ at fuel discharge and after a five year decay period. The average activity of Sm-151 in all reactor internals that are reported in the Swiss model inventory /Alder and McGinnes 1994/ has been used to calculate an average correlation factor between Sm-151 and Cs-137 of $2 \cdot 10^{-4}$. Evaluation of contamination data from a US reference PWR /Smith et al. 1985/ gave a correlation factor of $1 \cdot 10^{-3}$ five years after shutdown.

The correlation factor between Sm-151 and Cs-137 is selected to be $3 \cdot 10^{-3}$, based on data for a spent fuel assembly. This is also the highest value of those given above.

		Selected CRUD correlation factor	
Eu-152	Half-life = 13 years	$7 \cdot 10^{-5}$	Eu-152/Cs-137
Eu-154	Half-life = 8.8 years	0.1	Eu-153/Cs-137
Eu-155	Half-life = 5.0 years	0.07	Eu-154/Cs-137
			spent fuel assembly

From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between the europium isotopes and Cs-137 is $7 \cdot 10^{-5}$ for Eu-152, 0.1 for Eu-154, and 0.07 for Eu-155 at fuel discharge. The correlation factors are lower after five year decay period, $6 \cdot 10^{-5}$ for Eu-152, 0.09 for Eu-154, and 0.04 for Eu-155. The only measured data from which correlation factors for the relatively short-lived europium isotopes can be evaluated are activity measurements on decontamination solutions from cleaning of one of the Ågesta steam generators /Brodén 1993/. The correlation factors between the europium isotopes and Cs-137 calculated from these measurements are 0.07 for Eu-152, 0.03 for Eu-154, and 0.01 for Eu-155.

The correlation factor between europium isotopes and Cs-137 are selected from the spent fuel assembly, $7 \cdot 10^{-5}$ for Eu-152, 0.1 for Eu-154, and 0.07 for Eu-155. The selected values will not be important for the long-term safety since all isotopes have relatively short half-life (< 14 years).

Ho-166m	Selected CRUD correlation factor	
	$4 \cdot 10^{-6}$	Ho-166m/Co-60
Half-life = $1.2 \cdot 10^{3}$ years	spent fuel assembly	

No data on measured contamination levels of Ho-166m were found in the literature studied. From the reported radionuclide inventory in a reference BWR spent fuel assembly /Kjellbert 1990/ the correlation between Ho-166m as an activation product and Co-60 was calculated to be $2 \cdot 10^{-6}$ at fuel discharge and $4 \cdot 10^{-6}$ after a five year decay period. The correlation factor between Ho-166m and Co-60 was selected to be $4 \cdot 10^{-6}$. This is the correlation factor in a fuel assembly after 5 years of decay.

Radionuclides correlated to Pu-239+240

The radionuclides in the natural decay chains (see Figure 2-1 in Chapter 2) are all being correlated to Pu-239+240.

Measured values for the transuranics are relatively sparse. Therefore, the general approach for the selection of correlation factors for the transuranics has been to use the factors that can be calculated from data reported for a reference BWR spent fuel assembly (38 000 MWd/tU) /Kjellbert 1990/. Data are given for the time of fuel discharge and after a five year decay period. The correlation factor used is evaluated from data including five years of decay, since for some radionuclides the correlation factor is higher at that time, see Figure 4-10. The selected correlation factors are summarised in Table 4-2.

For some of the actinides measured data are available and correlation factors evaluated from the performed measurements are discussed below. The calculated data for a BWR spent fuel assembly falls within the interval of measured data except for Cm-243 and U-235. However to have a consistent set of correlation factors the calculated data have been selected as base for the evaluation and selection of correlation factors for the nuclides in the natural decay chains.

Table 4-2. Selected correlation factors to Pu-239+240 for radionuclides.in the natural decay chains. The selected values are identical to the correlation factors evaluated from the activities in a reference BWR spent fuel assembly after 5 years of decay.

Radionuclide	Half-life (years)	Selected CRUD correlation factor Radionuclide/(Pu-239+240)
Pb-210	22	$2 \cdot 10^{-11}$
Ra-226	$1.6 \cdot 10^{3}$	$2 \cdot 10^{-10}$
Ac-227	22	$4 \cdot 10^{-9}$
Th-229	$7.3 \cdot 10^{3}$	$3 \cdot 10^{-10}$
Th-230	$7.7 \cdot 10^{4}$	$9 \cdot 10^{-8}$
Th-232	$1.4 \cdot 10^{10}$	$1 \cdot 10^{-13}$
Pa-231	$3.3 \cdot 10^{4}$	3.10^{-8}
U-232	72	$3 \cdot 10^{-5}$
U-233	$1.6 \cdot 10^{5}$	$2 \cdot 10^{-8}$
U-234	$2.4 \cdot 10^{5}$	$1 \cdot 10^{-3}$
U-235 [1)]	$7.0 \cdot 10^{8}$	$2 \cdot 10^{-5}$
U-236 [1)]	$2.3 \cdot 10^{7}$	$3 \cdot 10^{-4}$
U-238 [1)]	$4.5 \cdot 10^{9}$	$4 \cdot 10^{-4}$
Np-237 [1)]	$2.1 \cdot 10^{6}$	$4 \cdot 10^{-4}$
Pu-238 [1)]	88	4
Pu-239 [1) 2)]	$2.4 \cdot 10^{4}$	key radionuclide
Pu-240 [1) 3)]	$6.5 \cdot 10^{3}$	key radionuclide
Pu-241 [1)]	14	$1 \cdot 10^{2}$
Pu-242	$3.8 \cdot 10^{5}$	$3 \cdot 10^{-3}$
Pu-244	$8.3 \cdot 10^{7}$	$7 \cdot 10^{-10}$
Am-241 [1)]	$4.3 \cdot 10^{2}$	1
Am-242m	$1.5 \cdot 10^{2}$	$1 \cdot 10^{-2}$
Am-243 [1)]	$7.4 \cdot 10^{3}$	$3 \cdot 10^{-2}$
Cm-243 [1)]	29	$2 \cdot 10^{-2}$
Cm-244 [1)]	18	3
Cm-245	$8.5 \cdot 10^{3}$	$3 \cdot 10^{-4}$
Cm-246	$4.7 \cdot 10^{3}$	$8 \cdot 10^{-5}$

[1)] Measured data are available.

[2)] The activity contribution of Pu-239 is 1/3 to the total activity of Pu-239+240.

[3)] The activity contribution of Pu-240 is 2/3 to the total activity of Pu-239+240.

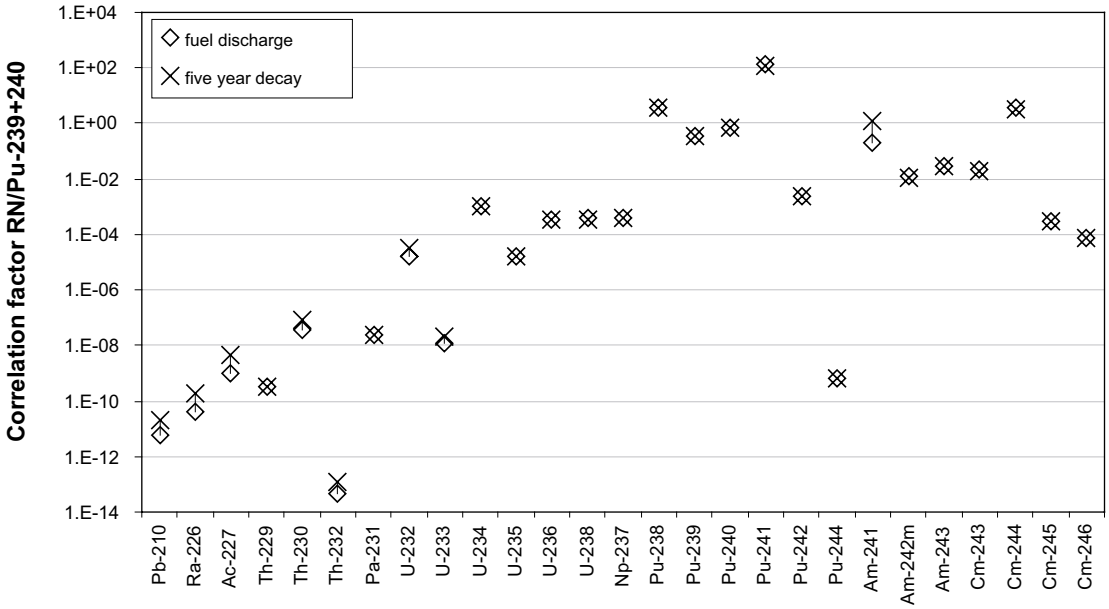

Figure 4-10. *Correlation factors between radionuclides in the natural decay chains and Pu-239+240 evaluated from calculated activities in a spent fuel assembly. Correlation factors are evaluated at fuel discharge and after a five year decay period.*

U-235	**Selected CRUD correlation factor**
	$2 \cdot 10^{-5}$ U-235/(Pu-239+240)
Half-life = $7.0 \cdot 10^{8}$ years	spent fuel assembly

The Swedish database in /Thierfeldt and Deckert 1995/ comprise measured concentrations in reactor coolant in Forsmark and Oskarshamn (number of 4). These data were used to evaluate an interval of the correlation factors of $4 \cdot 10^{-4}$–$6 \cdot 10^{-3}$, with a median value of $2 \cdot 10^{-3}$ (see Figure 4-11).

The correlation factor between U-235 and Pu-239+240 is selected to be $2 \cdot 10^{-5}$. This value is the ratio that can be calculated from the reported activities in a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/. The selected correlation factor has been selected to have a consistent composition of the actinides in the surface contamination although it is lower than the correlation factors evaluated from the measured data.

U-236	**Selected CRUD correlation factor**
	$3 \cdot 10^{-4}$ U-236/(Pu-239+240)
Half-life = $2.3 \cdot 10^{7}$ years	spent fuel assembly

Measured concentrations in reactor coolant in Oskarshamn (number of 3), included in the Swedish database in /Thierfeldt and Deckert 1995/, were used to evaluate the correlation factor between U-236 and Pu-239+240. These data were used to evaluate an interval of the correlation factors of $3 \cdot 10^{-4}$–$3 \cdot 10^{-3}$, with a median value of $7 \cdot 10^{-4}$ (see Figure 4-11).

The correlation factor between U-236 and Pu-239+240 is selected to be $3 \cdot 10^{-4}$. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

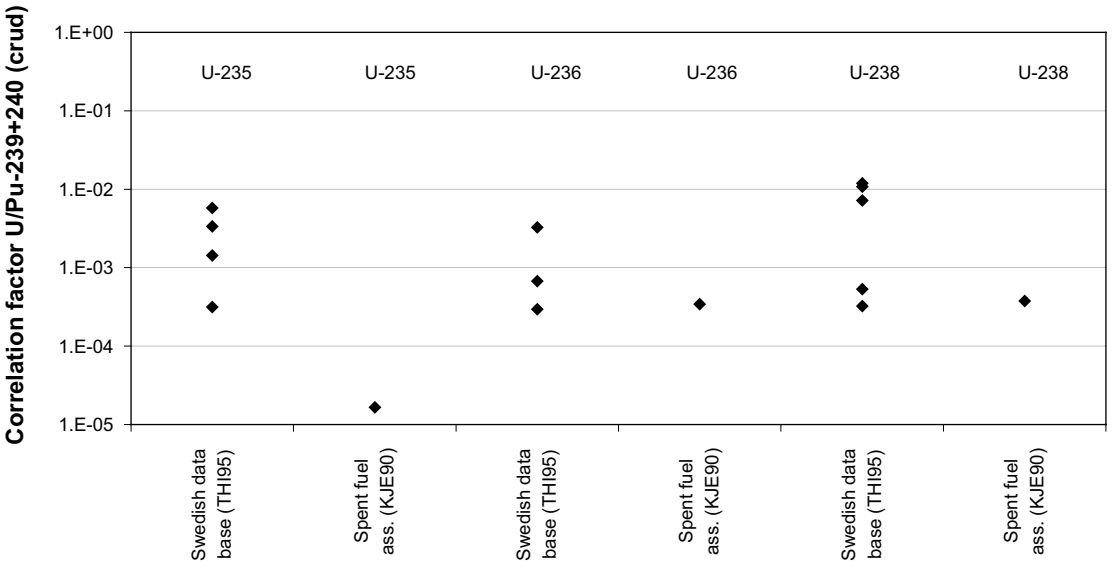

Figure 4-11. *Correlation factors evaluated between uranium isotopes and Pu-239+240 for surface contamination (CRUD).*

U-238	Selected CRUD correlation factor	
	$4 \cdot 10^{-4}$	U-238/(Pu-239+240)
Half-life = $4.5 \cdot 10^{9}$ years	spent fuel assembly	

Measured concentrations in reactor coolant in Oskarshamn and Forsmark (5 points of measuring), included in the Swedish database in /Thierfeldt and Deckert 1995/, were used to evaluate the correlation factor between U-238 and Pu-239+240. The correlation factor vary within an interval of $3 \cdot 10^{-4}$–$1 \cdot 10^{-2}$, with a median value of $7 \cdot 10^{-3}$ (see Figure 4-11).

The correlation factor between U-238 and Pu-239+240 is selected to be $4 \cdot 10^{-4}$. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

Np-237	Selected CRUD correlation factor	
	$4 \cdot 10^{-4}$	Np-237/(Pu-239+240)
Half-life = $2.1 \cdot 10^{6}$ years	spent fuel assembly	

Measured concentrations in reactor coolant in Oskarshamn and Forsmark (5 points of measuring), included in the database in /Thierfeldt and Deckert 1995/, were used to evaluate the correlation factor. The correlation between Np-237 and Pu-239+240 vary within an interval of $7 \cdot 10^{-4}$–$7 \cdot 10^{-2}$, with a median value of $5 \cdot 10^{-3}$.

The correlation factor between Np-237 and Pu-239+240 is selected to be $4 \cdot 10^{-4}$. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

Pu-238	Selected CRUD correlation factor
	4 Pu-238/(Pu-239+240)
Half-life = 88 years	spent fuel assembly

Measured data that were used to evaluate correlation factors between Pu-238 and Pu-239+240 were available and the correlation factors evaluated here are discussed and shown in Figure 4-12.

Data from continuos (4 times per year) measurements in Ringhals of transuranics in the reactor coolant water /Aronsson 1990, Ericson 1991, Sjöström 1992 and 1993/ were used to calculate correlation factors. The calculated intervals of the correlation factor for Ringhals 1-4 were:

Year	Correlation factors					
	R1-R4 (PWR+BWR)		R1 (BWR)		R2-R4 (PWR)	
	Interval	Median	Interval	Median	Interval	Median
1988	1.2–3.4	1.7	1.2–3.4	2.3	1.3–2.4	1.7
1989	10–360	49	46–52	49	10–360	42
1990	0.4–4	1	0.9–1.2	1.1	0.4–4	1.7
1991	18–2900	80	34–37	35	18–2900	124
1992	37–2170	430	37–930	480	97–2200	430

Evaluation of data from scrape tests on PWR fuel rods from Ringhals 2 /Fridemo et al. 1985a/ gave an interval of the correlation factor of 0.2−1, with a median value of 0.5. Scrape tests on BWR fuel rods from Barsebäck, Oskarshamn and Forsmark /Fridemo et al. 1985b/ were used to evaluate an interval of correlation factors of 0.03−49, with a median value of 4.

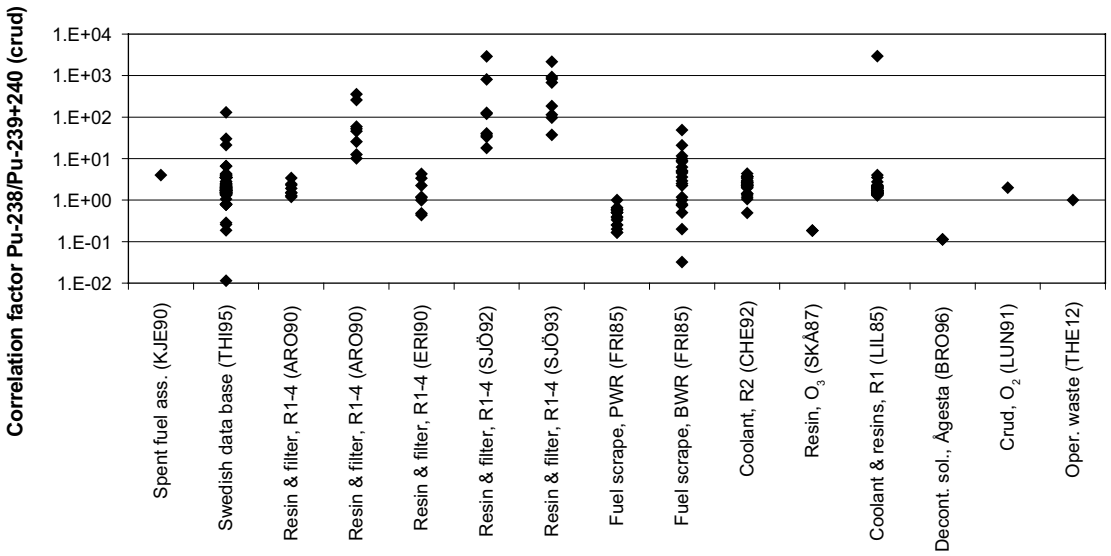

Figure 4-12. *Correlation factors evaluated between Pu-238 and Pu-239+240 for surface contamination (CRUD).*

Data from measurement of plutonium isotopes in the reactor coolant water in Ringhals 2 /Chen et al. 1992/ were used to calculate a correlation factor interval of 0.5−4, with a median value of 2.

Measured concentrations (2 points of measuring) in exchange resins in Oskarshamn 3 /Skålberg 1987/ were used to calculate a correlation factor of 0.2. Measurements of transuranics in reactor coolant water from Ringhals 1 (BWR) /Liljenzin and Johnson 1985/ were used to evaluate a correlation factor interval of 1.3−3000, with a median value of 1.7.

The correlation factor used for Swedish operational waste in /Thegerström and Hård 1981/ was 1. Data from measurements on decontamination solutions from the cleaning of two steam generators from Ågesta /Brodén 1996/ were used to calculate a correlation factor of 0.1.

The calculated inventory of activated corrosion products in pool water system in a BWR was reported in /Lundgren 1991/. The results were used to calculate a correlation factor of 2 one year after shutdown.

Evaluation of the Swedish database in /Thierfeldt and Deckert 1995/ resulted in a correlation factor interval of 0.01−130, with a median value of 2.

The reported contamination levels in a US reference PWR established by comparing experimental and theoretical values /Smith et al. 1985/ were used to calculate a correlation factor of 4 after 5 years of decay. /Robertson et al. 1993/ reported radionuclide concentrations, analysed from primary and secondary coolant piping from the US nuclear power station Shippingport. These data (7 points of measuring all dated 6 years after reactor shutdown) were used here to calculate correlation factors between Pu-238 and Pu-239+240. The correlation factors vary within the interval 0.05−2 with a median value of 1. The concentration of radionuclides on piping exposed to primary reactor coolant has also been measured at seven US nuclear power stations (BWR's and PWR's) /Robertson et al. 1984/. From these measurements the correlation factor calculated from the average activities was 1.

The correlation factor between Pu-238 and Pu-239+240 is selected to be 4. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

The data evaluated from Swedish measurements or calculations vary between 0.03 and 3000. The selected correlation factor is, however, in agreement with most of the calculated mean values evaluated from both Swedish measurements and information from other countries.

		Selected CRUD correlation factor	
Pu-239	Half-life = $2.4 \cdot 10^4$ years	1/3	Pu-239/(Pu-239+240)
Pu-240	Half-life = $6.5 \cdot 10^3$ years	2/3	Pu-240/(Pu-239+240)
		spent fuel assembly	

The activity of the key radionuclides Pu-239 and Pu-240 are often given as the total sum of the two, as they are difficult to separate in measurements and analyses.

The contribution to the total sum of the two is 1/3 of Pu-239 and 2/3 of Pu-240. This distribution is the distribution between the two that can be calculated from the reported activities in a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

Pu-241	Selected CRUD correlation factor
	100 Pu-241/(Pu-239+240)
Half-life = 14 years	spent fuel assembly

Very few data on surface contamination of Pu-241 were reported in the literature studied.

The inventory of activated corrosion products in the pool water system in a BWR, due to a fuel leakage, was calculated in /Lundgren 1991/. From the reported data a correlation factor of 167 one year after shutdown could be calculated.

The reported contamination levels in a reference PWR, established by comparing experimental and theoretical values /Smith et al. 1985/, were used to calculate a correlation factor of 100 after a five year decay period.

The correlation factor between Pu-241 and Pu-239+240 is selected to be 100. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

Am-241	Selected CRUD correlation factor
	1 Am-241/(Pu-239+240)
Half-life = $4.3 \cdot 10^2$ years	spent fuel assembly

The activity of Am-241 and Pu-239+240 in ion-exchange resins from Ringhals has been reported /Aronsson 1990, Ericson 1991, Sjöström 1992, 1993/. The reported data were calculated from measurements of Co-60. The reported activities were here used to evaluate correlation factors between Am-241 and Pu-239+240. The correlation factors were within the interval 0.09−38, with a median value of 1.

Activity information from scrape tests on fuel rods from Barsebäck, Oskarshamn and Ringhals (BWR) /Fridemo et al. 1985b/ were used to calculate correlation factors. The correlation factors varied within the interval 0.2−35, with a median value of 2. Evaluation of scrape tests on fuel rods from Ringhals (PWR) /Fridemo et al. 1985a/ gave correlation factors within the interval 0.1−1, with a median value of 0.6.

Data from measurements on reactor coolant water in Ringhals 1 /Liljenzin and Johnson 1985/ were used to evaluated correlation factors within the interval 0.006−42, with a median value of 0.3. These measurements were also included as a data subset in the Swedish database of /Thierfeldt and Deckert 1995/.

Measurements on ion-exchange resins in Oskarshamn 3 (2 points of measuring) /Skålberg 1987/ was used to calculate a correlation factor of $4 \cdot 10^{-3}$.

The inventory of activated corrosion products in pool water system in a BWR, due to a fuel leakage scenario, was calculated in /Lundgren 1991/. From the reported data a correlation factor of 0.2 one year after shutdown could be calculated.

The correlation factors evaluated from Swedish information are shown in Figure 4-13.

The reported contamination levels in a US reference PWR, established by comparing experimental and theoretical values /Smith et al. 1985/, were used to calculate a correlation factor of 1.2 after a five year decay period.

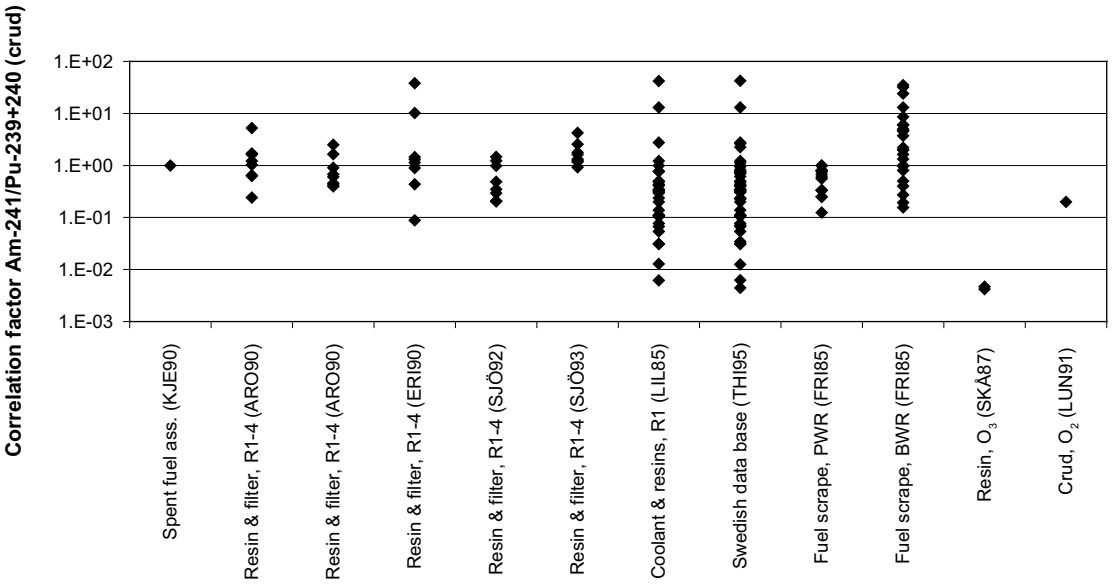

Figure 4-13. *Correlation factors evaluated between Am-241 and Pu-239+240 for surface contamination (CRUD).*

Radionuclide concentrations analysed from primary and secondary coolant piping from the US nuclear power station Shippingport was reported by /Robertson et al. 1993/. These data (7 points of measuring all dated 6 years after reactor shutdown) were used here to calculate correlation factors between Am-241 and Pu-239+240. The correlation factors vary within the interval 0.09−7, with a median value of 0.3. The concentration of radionuclide on piping exposed to primary reactor coolant has also been measured at seven nuclear power stations (BWR's and PWR's) in the US /Robertson et al. 1984/. The correlation factor calculated from the average activities reported from these measurements was 2.

The correlation factor between Am-241 and Pu-239+240 is selected to be 1. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly after a five year decay period /Kjellbert 1990/. The correlation factor calculated at fuel discharge is 0.2.

Am-243	Selected CRUD correlation factor
Half-life = $7.4 \cdot 10^3$ years	$3 \cdot 10^{-2}$ Am-243/(Pu-239+240) spent fuel assembly

The Swedish database in /Thierfeldt and Deckert 1995/ was used to evaluate correlation factors between Am-243 and Pu-239+240. The correlation factors varies within an interval of 0.006−3, with a median value of 0.08.

The activity of transuranics in waste from Clab has been calculated from measurements of the activity of Co-60 and correlation factors /Axelsson 1989, 1990, 1991, Grahn 1987/. The reported activities were used to calculate correlation factors between Am-243 and Pu-239+240 that were within the interval 0.02−0.7, with a median value of 0.2.

The correlation factors evaluated from the Swedish information are shown in Figure 4-14.

The correlation factor between Am-243 and Pu-239+240 is selected to be $3 \cdot 10^{-2}$. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel

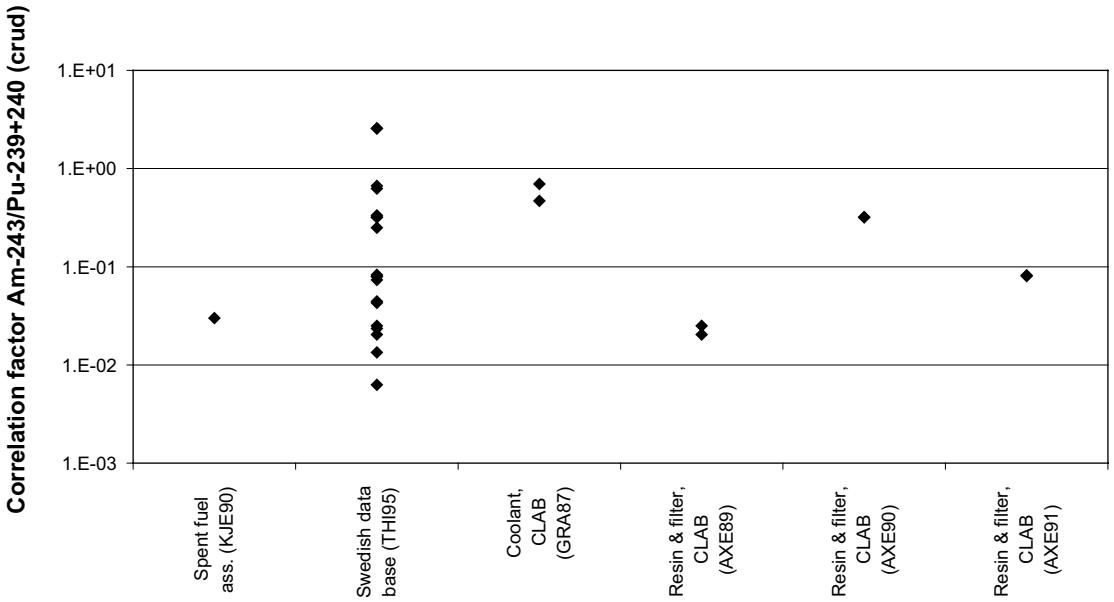

Figure 4-14. *Correlation factors evaluated between Am-243 and Pu-239+240 for surface contamination (CRUD).*

assembly at fuel discharge and after a five year decay period /Kjellbert 1990/. The selected correlation factor is within the range evaluated from measured data reported in /Thierfeldt and Deckert 1995/.

Cm-243	Selected CRUD correlation factor
Half-life = 29 years	$2 \cdot 10^{-2}$ Cm-243/(Pu-239+240) spent fuel assembly

The Swedish database in /Thierfeldt and Deckert 1995/ was used to evaluate correlation factors between Cm-243 and Pu-239+240. The correlation factors varies within an interval of 0.7–25, with a median value of 1.

The activity of transuranics in waste from Clab has been calculated from measurements of the activity of Co-60 and correlation factors /Axelsson 1989, 1990, 1991/. The reported activities were used to calculate correlation factors between Cm-243 and Pu-239+240 that were within the interval 0.9−3 with a median value of 1.

The correlation factors evaluated from the Swedish information are shown in Figure 4-15.

The correlation factor between Cm-243 and Pu-239+240 is selected to be $2 \cdot 10^{-2}$. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a 5 year decay period /Kjellbert 1990/. The correlation factor has been selected to have a consistent composition of the actinides in the surface contamination although it is lower than the correlation factors evaluated from the measured data.

Cm-244	Selected CRUD correlation factor
Half-life = 18 years	3 Cm-244/(Pu-239+240) spent fuel assembly

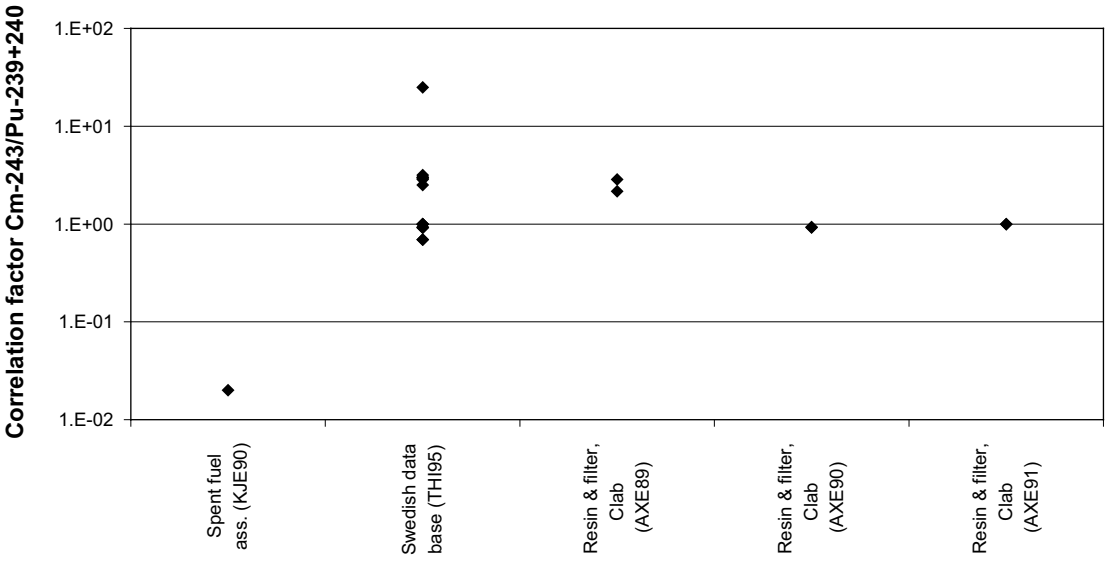

Figure 4-15. *Correlation factors evaluated between Cm-243 and Pu-239+240 for surface contamination (CRUD).*

The Swedish database in /Thierfeldt and Deckert 1995/ was used to evaluate correlation factors between Cm-244 and Pu-239+240. The correlation factors varies within an interval of $4 \cdot 10^{-4}$−17, with a median value of 0.4.

Data from scrape tests on fuel rods were reported by /Fridemo et al. 1985a, 1985b/. These data were used to calculate correlation factors for both a PWR (Ringhals) and BWR's (Barsebäck, Oskarshamn, Ringhals). The correlation factor interval was between 0.2 and 2 with median value 0.4 for the PWR, and for the BWR's between 0.03 and 260 with a median value of 30.

Measurements on ion-exchange resins in Oskarshamn 3 /Skålberg 1987/ was used to calculate a correlation factor of $7 \cdot 10^{-4}$.

The activity of Cm-244 and Pu-239+240 in ion-exchange resins from Ringhals has been reported /Aronsson 1990, Ericson 1991, Sjöström 1992, 1993/. The reported data were calculated from measurements of Co-60. The reported activities were here used to evaluate correlation factors between Cm-244 and Pu-239+240. The correlation factors evaluated were within the interval 0.06−7, with a median value of 0.8.

The activity of transuranics in waste from Clab has been calculated from measurements of the activity of Co-60 and correlation factors /Axelsson 1989, 1990, 1991, Grahn 1987/. The reported activities were used to calculate correlation factors between Cm-244 and Pu-239+240 that were within the interval 0.2−4, with a median value of 2.

Data from measurements on reactor coolant water in Ringhals 1 /Liljenzin and Johnson 1985/ were used to evaluate correlation factors within the interval $4 \cdot 10^{-4}$−11, with a median value of 0.2. Some of the measured data were also included in the Swedish database of /Thierfeldt and Deckert 1995/.

/Lundgren 1991/ calculated the inventory of activated corrosion products in the pool water system in a BWR, due to a fuel leakage. The reported data were used to calculate a correlation factor of 2 one year after shutdown.

The correlation factors evaluated from the Swedish measurements are shown in Figure 4-16.

Concentrations analysed from primary and secondary coolant piping from the US nuclear power station Shippingport has been reported by /Robertson et al. 1993/. These data (7 points of measuring all dated 6 years after reactor shutdown) were used to calculate correlation factors between Cm-244 and Pu-239+240. The correlation factors varies within the interval 0.02−0.5,

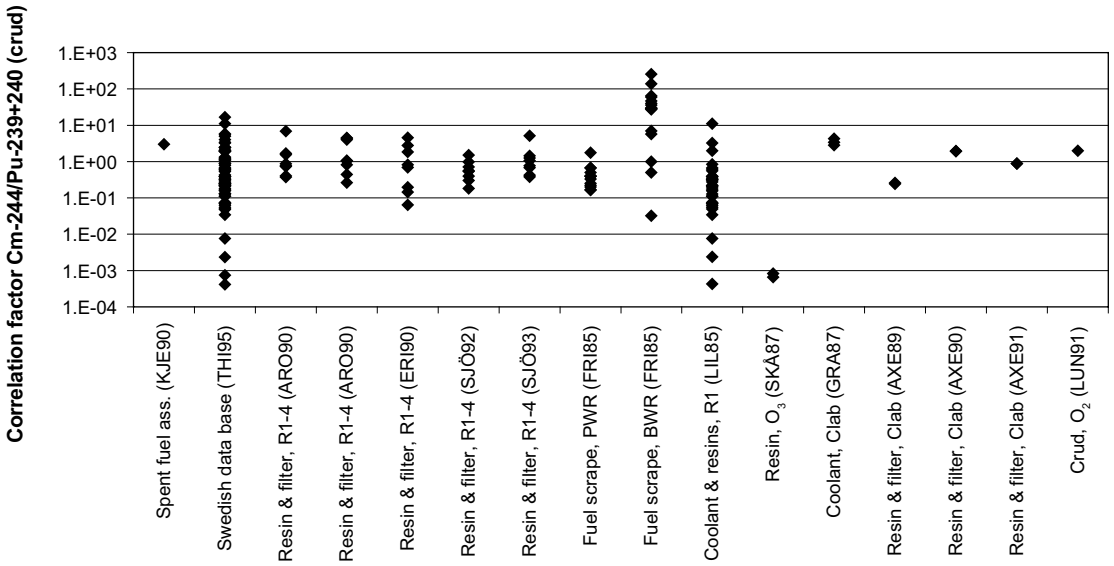

Figure 4-16. *Correlation factors evaluated between Cm-244 and Pu-239+240 for surface contamination (CRUD).*

with a median value of 0.03. The activity of radionuclides on piping exposed to primary reactor coolant has also been measured at seven US nuclear power stations (BWR's and PWR's) / Robertson et al. 1984/. A correlation factor of 0.7 was evaluated from the average activities reported from these measurements.

The reported contamination levels in a US reference PWR established by comparing experimental and theoretical values /Smith et al. 1985/ were used to calculate a correlation factor of 2 after a five year decay period.

The correlation factor between Cm-244 and Pu-239+240 is selected to be 3. This value is the ratio that can be calculated from the activities reported for a reference BWR spent fuel assembly at fuel discharge and after a five year decay period /Kjellbert 1990/.

4.2 Induced activity

In this study the approach has been to define correlation factors to Co-60 for the radionuclides in neutron activated metals. The correlation factors for neutron induced activity in the metals are evaluated from activity levels reported in literature.

The following radionuclides are expected to dominate the neutron induced activity in irradiated steel and other metals and consequently a correlation factor to a key nuclide has been evaluated for: H-3, C-14, Cl-36, Fe-55, Co-60, Ni-59, Ni-63, Nb-93m, Nb-94, Mo-93, Tc-99. No correlation factors have been evaluated for additional radionuclides e.g. Be-10, Zr-93, Ag-108m, Ba-113, Cd-113, Sb-125, Sm-151, Eu-152, Eu-154, and Eu-155. The activity content of these nuclides is included in the reference waste inventory for SFL 3-5 only if the activity is reported for a specific waste type.

For most of the radionuclides present in irradiated metals correlation factors to the key nuclide, Co-60, have been evaluated from reported data. The majority of these data are from calculations of the neutron activation of steel, while only a few data are from measurements or for other materials than steel. One correlation factor for each radionuclide is selected which will be used to estimate the activity content in irradiated steel materials when no specific information is at hand. No correlation factors for other materials than steel is evaluated here, but possible values are discussed when data are available for e.g. zircaloy, Inconel etc.

Since the selected key radionuclide Co-60 has a relatively short half-life (5.3 years), the time for the evaluation of the correlation factor is of significant importance. Most of the activities referred to are valid at reactor shutdown or after 1 or 5 years of decay. The correlation factors evaluated are therefore selected to be applicable for the SFL 3-5 waste after 1 year of decay.

The content of neutron activation products in the metals depends on the concentration of the parent isotope, the amount of major constituents, the amount of impurities, the cross section of the specie, the duration of the irradiation, the neutron flux and the distance to the radiation source.

H-3	Selected correlation factor for induced activity
	$1 \cdot 10^{-4}$ H-3/Co-60
Half-life = 12 years	see Figure 4-17

The activity of H-3 is reported in the Swiss model inventory /Alder and McGinnes 1994/. In the Swiss data the induced activity and surface contamination is reported as a sum. However, for the purpose of this study, it has been assumed that the activity content of H-3 in steel reactor waste is dominated by the contribution from induced activity. Activity data for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) were used to evaluate correlation factors. The average correlation factor between H-3 and Co-60 was around 10^{-4}. The activities reported in the Swiss model inventory include both induced activity and surface contamination and the time between discharge and conditioning is 5 years for the changeable non-fuel reactor core components and 1 or 2 years for the decommissioning waste. An average correlation factor between H-3 and Co-60 of $3 \cdot 10^{-2}$ was also evaluated from the reported total activity in Swiss reactor waste /Alder and McGinnes 1994/. The contribution of H-3 from waste containing other materials than steel, e.g. sludges and resins, is however judged to be significant.

Calculations of the induced activity in a Finish core grid at discharge /Antilla and Wasastjerna 1989/ were used to evaluate a correlation factor between H-3 and Co-60 of 10^{-8}. Correlation factors evaluated from the reported activity calculations for the Clab storage canisters (1 year

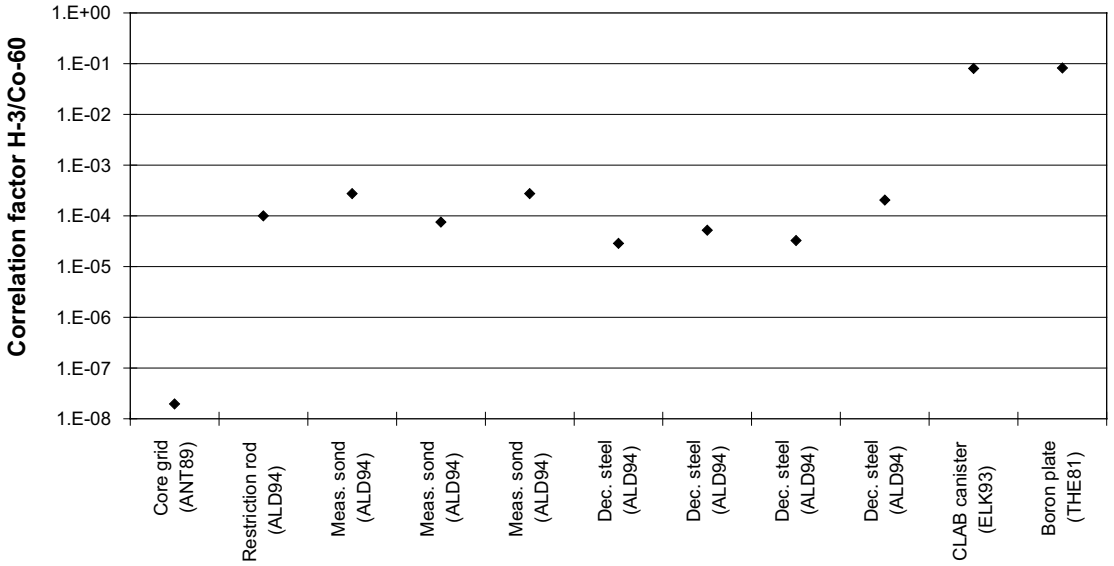

Figure 4-17. *Correlation factors evaluated between H-3 and Co-60 for induced activity.*

of decay) made of steel and boron steel /Elkert 1993/ is considerably higher, 10^{-1}. A correlation factor of 10^{-1} was calculated also from reported activities in boron plates /Thegerström and Hård 1981/.

From this wide span of evaluated correlation factors (see Figure 4-17) between H-3 and Co-60 a correlation factor of 10^{-4} is selected for steel materials, mainly based on the activity data from the Swiss steel reactor waste (flow restriction rods, measuring sonds, and medium active steel waste).

The correlation factor is not based on estimates from Swedish power plants and is therefor relatively uncertain. The correlation factor evaluated for the Finish core grid is almost three orders of magnitudes lower than the selected correlation factor.

C-14	Selected correlation factor for induced activity
	$5 \cdot 10^{-4}$ C-14/Co-60
Half-life = $5.7 \cdot 10^{3}$ years	see Figure 4-18

The induced activity in the core grid, the moderator tank and the core spray support in a Swedish BWR after 1 year of decay has been calculated and is reported in /Lundgren 1982/. The calculated activities were used to evaluate correlation factors between C-14 and Co-60 just above 10^{-3}. The activity in the core grid, moderator tank and moderator tank cover in a finish BWR at discharge was calculated and reported by /Antilla and Wasastjerna 1989/. The correlation factors calculated from these results were also just above 10^{-3}. Activity data reported for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) /Alder and McGinnes 1994/ have been used to evaluate correlation factors between C-14 and Co-60 that varies between $3 \cdot 10^{-5}$ and $2 \cdot 10^{-4}$. Reported activity data for a US PWR core barrel and shroud at shutdown /Smith et al. 1978/ were used to calculate correlation factors between C-14 and Co-60 of $2 \cdot 10^{-4}$ and $3 \cdot 10^{-4}$, respectively. /Robertson et al. 1993/ has reported measured activities of C-14 in spent fuel assembly hardware (steel). In addition, the correlation factor between C-14 and Co-60 was reported to vary from $6 \cdot 10^{-5}$ to $9.9 \cdot 10^{-4}$ /Robertson et al. 1993/. The reported average correlation factor, including information from all parts of the fuel assembly, varies between $4 \cdot 10^{-5}$ and $7 \cdot 10^{-3}$ with a geometric mean of $5 \cdot 10^{-4}$ /Robertson et al. 1993/.

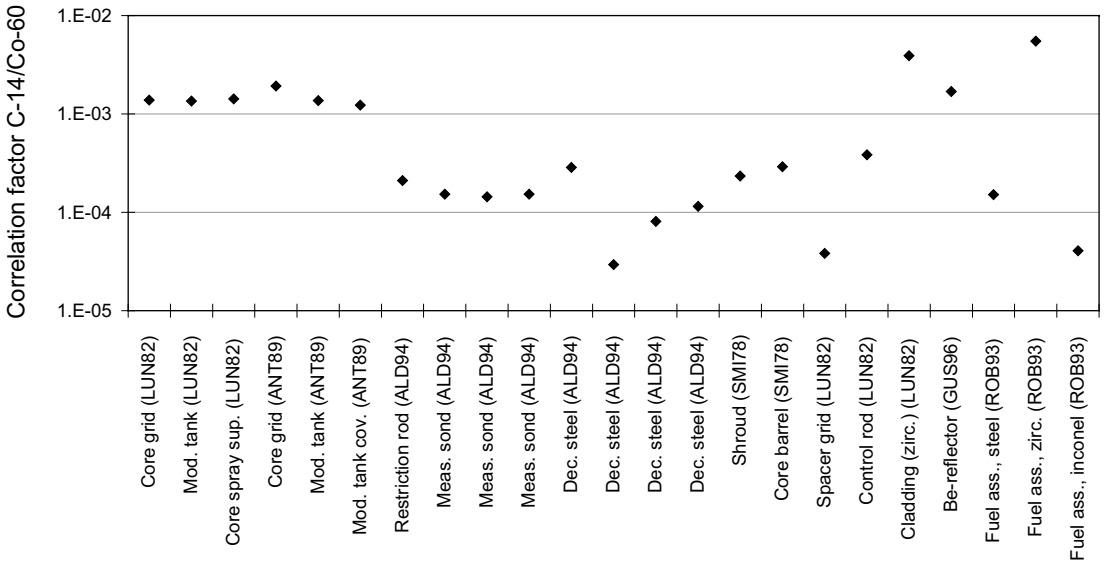

Figure 4-18. *Correlation factors evaluated between C-14 and Co-60 for induced activity.*

From the data compiled in Figure 4-18 the correlation factor between C-14 and Co-60 is selected to be $5 \cdot 10^{-4}$.

The correlation factor is selected for steel, but may be applied also for other metals such as zircaloy, Inconel and boron steel. The few references /Lundgren 1982, Thegerström and Hård, 1981, Elkert 1993, Gustafsson 1995/ giving data for these materials show that the correlation factor for these materials (see Figure 4-18) are roughly covered by the same variation interval as the correlation factors for steel.

Cl-36	Selected correlation factor for induced activity
	$7 \cdot 10^{-7}$ Cl-36/Co-60
Half-life = $3.0 \cdot 10^{5}$ years	see Figure 4-19

Information on induced activity of Cl-36 is relatively sparse. The induced activity of Cl-36 was calculated and reported for a Finish core grid in /Antilla and Wasastjerna 1989/. From this calculation a correlation factor between Cl-36 and Co-60 at discharge was calculated to be $4 \cdot 10^{-9}$. The activity content of Cl-36 is also reported in the Swiss model inventory /Alder and McGinnes 1994/. Median activity data for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) were used to evaluate correlation factors between Cl-36 and Co-60. The activities reported in the Swiss model inventory include both induced activity and surface contamination and the time between discharge and conditioning is 5 years for the changeable non-fuel reactor core components and 1 or 2 years for the decommissioning waste. The correlation factors are between $9 \cdot 10^{-7}$ and $9 \cdot 10^{-6}$ (see Figure 4-19). The influence of the time interval between discharge and conditioning on the correlation factors is that the correlation factors at discharge are slightly smaller than those evaluated, at the most a factor of 2 lower.

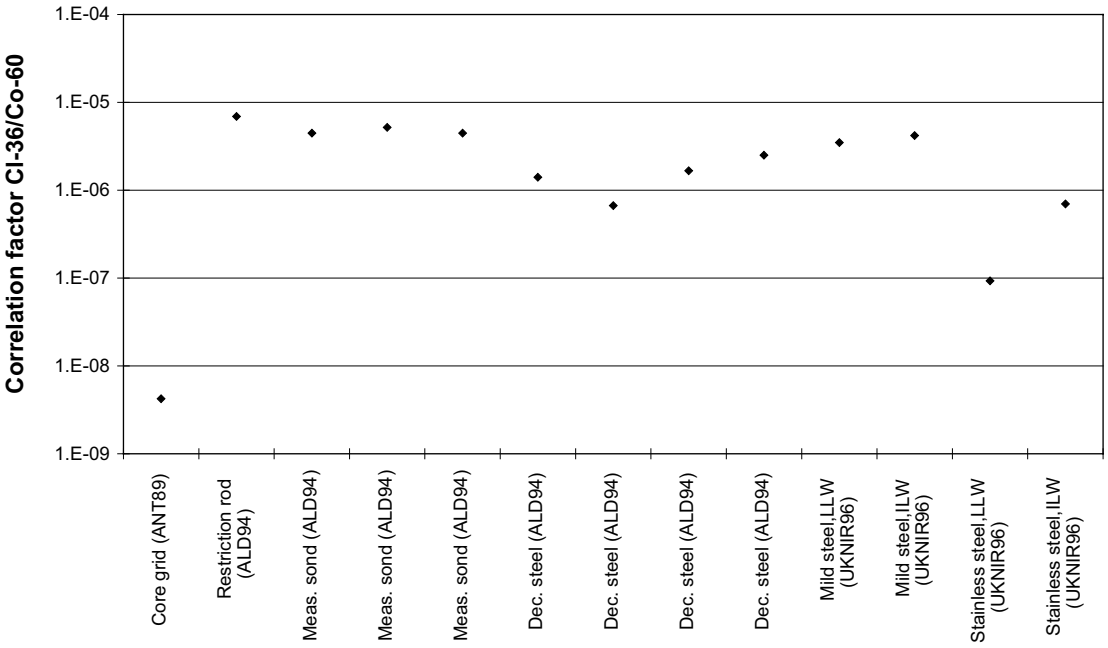

Figure 4-19. *Correlation factors evaluated between Cl-36 and Co-60 for induced activity.*

UK Nirex Ltd. has spent considerable efforts to estimate the content of Cl-36 in waste arising from nuclear facilities. The stocks of radioactive wastes in the United Kingdom as existing on 1 April 1994, together with forecasts of radioactive wastes arising in the future have been reported /UK Nirex Ltd. 1996/. The report gives e.g. forecasts of the activities in decommissioning waste from a PWR (Sizewell B). The induced activity in four representative waste types was selected to evaluate correlation factors between Cl-36 and Co-60 at reactor shutdown. The selected waste types were "mild steel, LLW", "mild steel, ILW", "stainless steel, LLW" and "stainless steel, ILW". The evaluated correlation factors are between $9 \cdot 10^{-8}$ and $4 \cdot 10^{-6}$ with a median value of $2 \cdot 10^{-6}$ (see Figure 4-19). The waste type "stainless steel, ILW" contributes with 93% of the Cl-36 content in the decommissioning waste from Sizewell B. The correlation factor between Cl-36 och Co-60 evaluated for this waste type is $7 \cdot 10^{-7}$ at reactor shutdown. The upper and lower band of the uncertainties in the reported activities in the four selected waste types are reported to be a factor of 100 and 10 for Cl-36 and a factor of 10 for both the upper and lower band for Co-60.

In the selection of the correlation factor between Cl-36 and Co-60, most weight has been assigned to the data reported by Nirex Ltd. The correlation factor between Cl-36 and Co-60 is selected to be $7 \cdot 10^{-7}$ from the waste type that dominate the Cl-36 content in the intermediate level stainless steel waste from the UK PWR Sizewell B.

Fe-55	**Selected correlation factor for induced activity**
	5 Fe-55/Co-60
Half-life = 2.7 years	see Figure 4-20

Induced activity of Fe-55 in internal parts has been reported in a number of references and correlation factors between Fe-55 and Co-60 have been calculated and compiled, see Figure 4-20. The references include calculated activities in internal parts from a Swedish BWR /Lundgren 1982, SKB 1979/ and a Swedish PWR /Højerup 1993/. Further, calculated activities for a Finish core grid, moderator tank and moderator tank cover /Antilla and Wasastjerna 1989/ are available, as well as data for a PWR shroud and a PWR core barrel from the US at reactor shutdown /Smith et al. 1978/. Activity data for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) /Alder and McGinnes 1994/ were also included in the evaluation. The activities reported in the Swiss model inventory include both induced activity and contamination. The correlation factors calculated from the activities reported in the different references vary within a relatively narrow interval, 1−10 (see Figure 4-20).

The correlation factor between Fe-55 and Co-60 is selected to be 5 from the compiled data.

Correlation factors evaluated for other materials than steel, e.g. zircaloy, Inconel and boron steel /Lundgren 1982, Thegerström and Hård 1981, Elkert 1993, Gustafsson 1995/, are all within the same interval as the correlation factors for steel.

Ni-59	**Selected correlation factor for induced activity**
	$5 \cdot 10^{-3}$ Ni-59/Co-60
Half-life = $7.5 \cdot 10^{4}$ years	see Figure 4-21

The induced activity of Ni-59 in internal reactor parts has been reported in a number of references and correlation factors between Ni-59 and Co-60 have been calculated, see Figure 4-21. The evaluation includes calculated activities in internal parts from a Swedish BWR /Lundgren

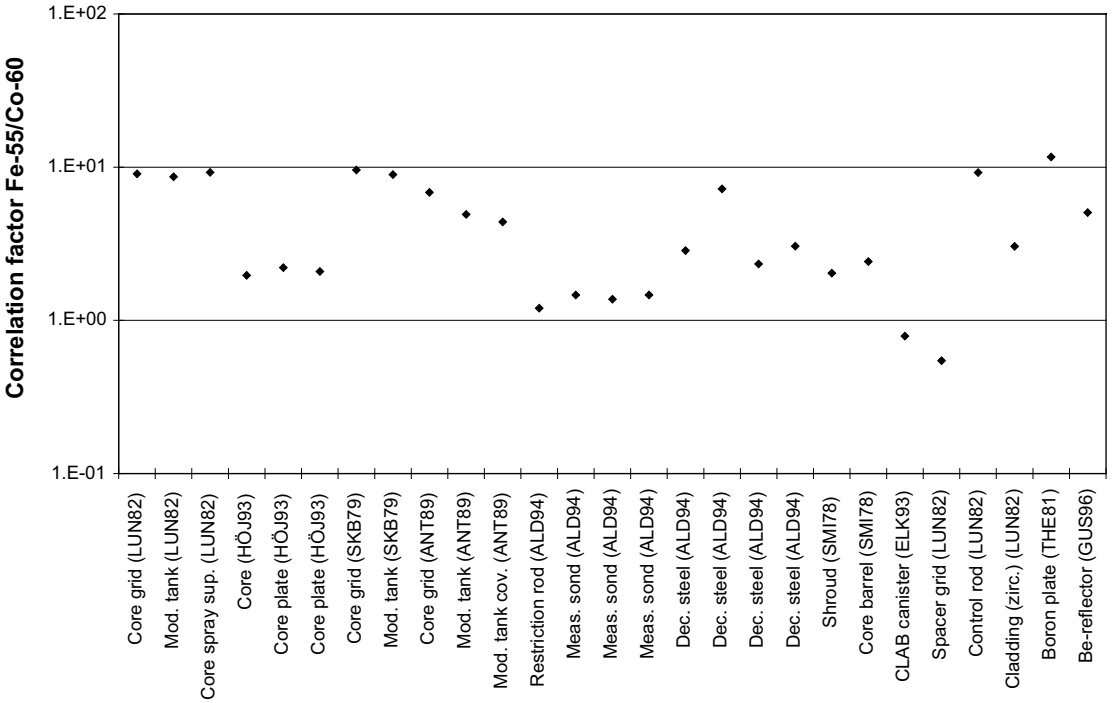

Figure 4-20. *Correlation factors evaluated between Fe-55 and Co-60 for induced activity.*

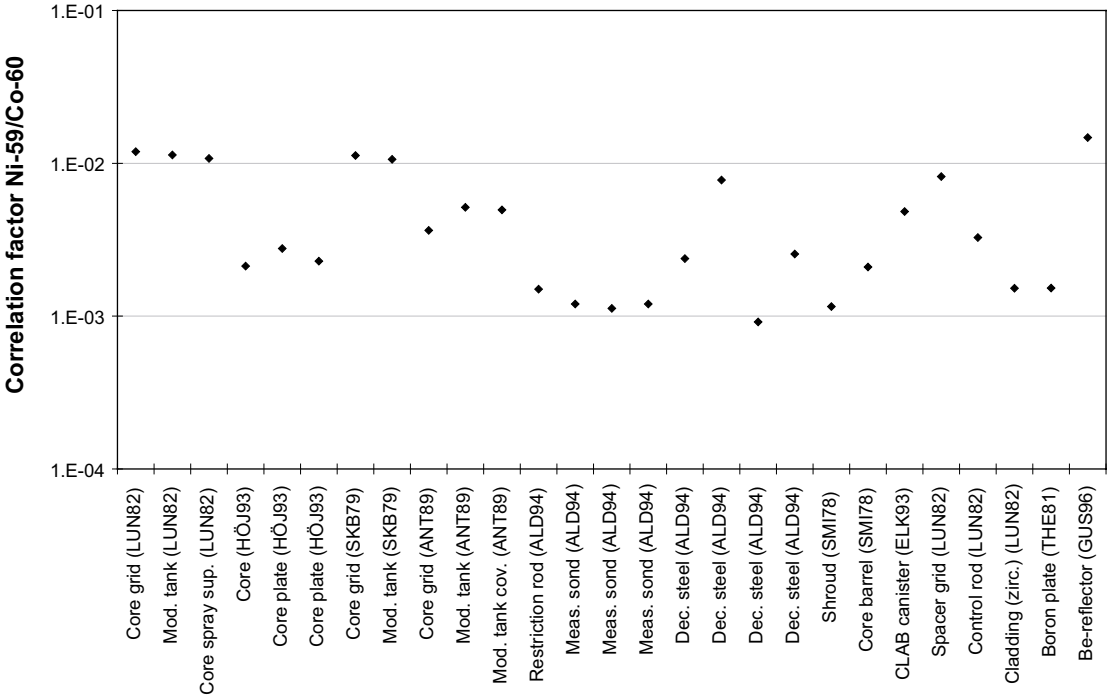

Figure 4-21. *Correlation factors evaluated between Ni-59 and Co-60 for induced activity.*

1982, SKB 1979/ and a Swedish PWR /Højerup 1993/. Further, calculated induced activities in a Finish core grid, a moderator tank and a moderator tank cover /Antilla and Wasastjerna 1989/ were included, as well as, data for a PWR shroud and a PWR core from /Smith et al. 1978/. Activity data for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) /Alder and McGinnes 1994/ were also included in the

evaluation. The activities reported in the Swiss model inventory include both induced activity and contamination. The correlation factors calculated from the activities reported in the different references vary within an interval of $9 \cdot 10^{-4}$–$1 \cdot 10^{-2}$ (see Figure 4-21).

The correlation factor between Ni-59 and Co-60 is selected to be $5 \cdot 10^{-3}$ from compiled data (see Figure 4-21).

The correlation factors evaluated for other materials than steel, e.g. zircaloy, Inconel and boron steel /Lundgren 1982, Thegerström and Hård 1981, Elkert 1993, Gustafsson 1995/, are within the same interval as the correlation factors for steel.

Ni-63	Selected correlation factor for induced activity
	1 Ni-63/Co-60
Half-life = $9.6 \cdot 10^{1}$ years	see Figure 4-22

The activity content of Ni-63 are reported in a number of references and the correlation factor between Ni-63 and Co-60 have been calculated from the reported activities, see Figure 4-22. The references include calculated activities in internal parts from a Swedish BWR /Lundgren 1982, SKB 1979/ and a Swedish PWR /Højerup 1993/. Furthermore, activities in a Finish core grid, a moderator tank and a moderator tank cover /Antilla and Wasastjerna 1989/ are included, as well as data for a PWR shroud and a PWR core from /Smith et al. 1978/. Activity data for a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) /Alder and McGinnes 1994/ were also included in the evaluation. The activities reported in the Swiss model inventory include both induced activity and contamination. The correlation factors calculated from the activities reported in the different references vary within an interval of 0.1−2 (see Figure 4-22).

The correlation factor between Ni-63 and Co-60 is selected to be 1. This may be somewhat high compared to the correlation factors evaluated from data in the references. The value is, however,

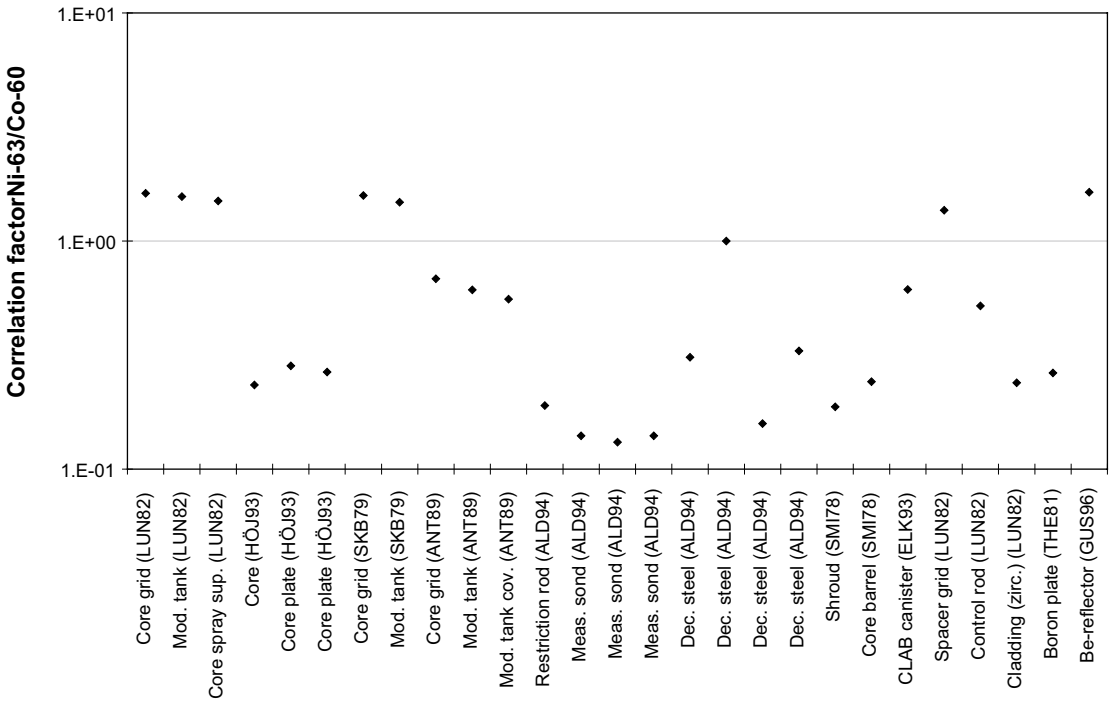

Figure 4-22. *Correlation factors evaluated between Ni-63 and Co-60 for induced activity.*

of the same magnitude as the ratio between Ni-63 and Ni-59 of 150 evaluated from the activities reported in a spent fuel assembly /Kjellbert 1990/.

The correlation factor selected for steel may be applied also for other metals such as zircaloy, Inconel and boron steel. The references giving data for these materials show that the correlation factors roughly have the same variation interval as the correlation factors for steel /Lundgren 1982, Thegerström and Hård 1981, Elkert 1993, Gustafsson 1995/ (see Figure 4-22).

Nb-93m	Selected correlation factor for induced activity	
	$1 \cdot 10^{-3}$	Nb-93m/Co-60
Half-life = 14 years	decommissioning	

Information on induced activity of Nb-93m is very sparse. The induced activity of Nb-93m has been calculated and reported for the core grid, moderator tank and moderator tank cover in a Finish BWR /Antilla and Wasastjerna 1989/. The correlation factors between Nb-93m and Co-60 could be calculated to be within the interval of $4 \cdot 10^{-7}$–$5 \cdot 10^{-7}$ from these data.

Activities measured in spent fuel disassembly hardware (stainless steel and Inconel) from Shippingport in US have been used to estimate the average ratio between Nb-93m and Nb-94, which was reported to be 98 /Robertson et al. 1993/. These measurements indicate that activity calculations tend to underestimate the activity of Nb-93m /Robertson et al. 1993/. Activity data for one Swiss steel waste type (medium active decommissioning waste) /Alder and McGinnes 1994/ including both induced activity and surface contamination was used to calculate a correlation factor between Nb-93 and Co-60 of $1 \cdot 10^{-3}$.

The correlation factor between Nb-93m and Co-60 is selected to be $1 \cdot 10^{-3}$, which means that the ratio between Nb-93m and Nb-94 is 100 (see Nb-94 below).

The activities calculated for Clab storage canisters, holding reference BWR spent fuel assemblies /Elkert 1993/, were used to evaluate a ratio between Nb-93m and Co-60 of $1 \cdot 10^{-2}$. Calculated activities for the cladding from a BWR results in a ratio between the two of $4 \cdot 10^{-4}$ /Lundgren 1982/ and a decommissioned beryllium reflector gave a correlation factor of $6 \cdot 10^{-7}$ /Gustafsson 1995/. In addition, the activities of Nb-93m and Nb-94 reported for the Clab storage canisters were used to evaluate a correlation factor of 40 between the two niobium isotopes, Nb-93m and Nb-94.

Nb-94	Selected correlation factor for induced activity	
	$1 \cdot 10^{-5}$	Nb-94/Co-60
Half-life = $2.0 \cdot 10^{4}$ years	see Figure 4-23	

/Lundgren 1982/ has calculated the induced activity in a core grid, a moderator tank and a core spray support in one Swedish BWR after one year of decay. These data were used to evaluate correlation factors between Nb-94 and Co-60 just above 10^{-5}. Calculated activities in a number of selected Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) /Alder and McGinnes 1994/ have been used to evaluate the correlation factor between Nb-94 and Co-60. The correlation factors vary within the interval $1 \cdot 10^{-6}$–$6 \cdot 10^{-6}$, whereas one data point gave a considerably higher value ($3 \cdot 10^{-4}$). The median value of the correlation factors calculated from the Swiss data is $5 \cdot 10^{-6}$. Calculated activity data for a US PWR core barrel and a shroud /Smith et al. 1978/ were used to calculate correlation factors between Nb-94 and Co-60 of $4 \cdot 10^{-6}$ and $8 \cdot 10^{-6}$, respectively.

The correlation factor between Nb-94 and Co-60 is selected to be $1 \cdot 10^{-5}$ from the data compiled in Figure 4-23.

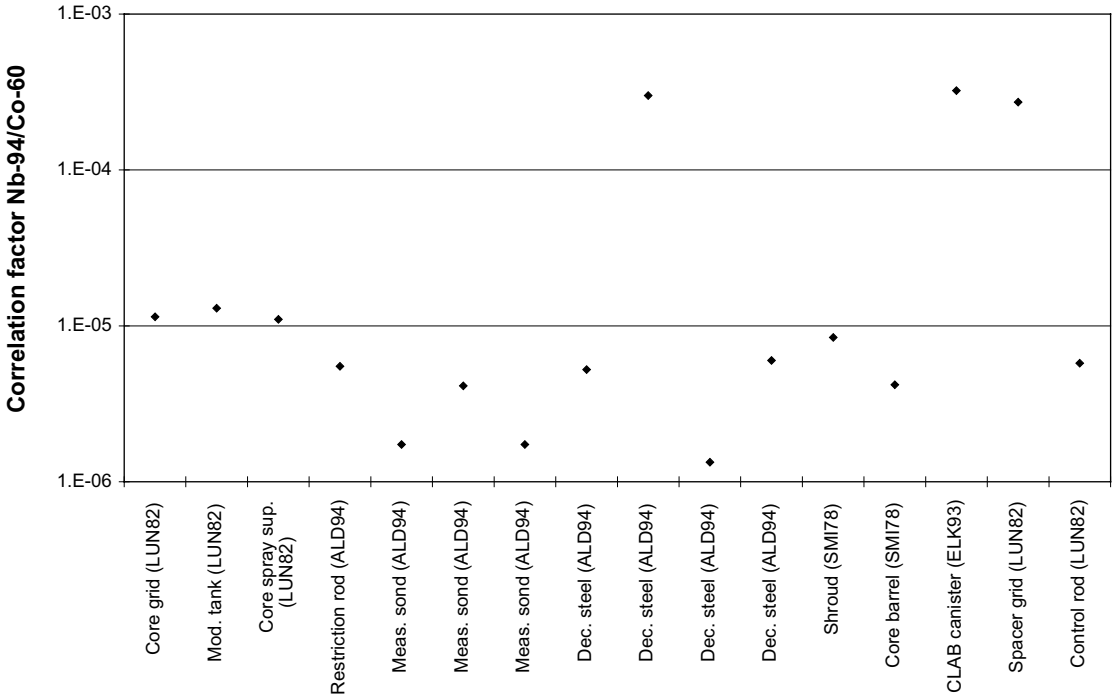

Figure 4-23. *Correlation factors evaluated between Nb-94 and Co-60 for induced activity.*

The results from activation calculations for Clab storage canisters, holding reference BWR spent fuel assemblies /Elkert 1993/, were used to calculate a ratio between Nb-94 and Co-60 of $3 \cdot 10^{-4}$. This value was not included in the selection of the correlation factor since the data are not from a nuclear power reactor. Calculated activities for BWR spacer grids made of Inconel were used to calculate a correlation factor of $3 \cdot 10^{-4}$ and the activities in BWR control rods (boron steel and zircaloy) gave a ratio between the two of $6 \cdot 10^{-6}$ /Lundgren 1982/.

Mo-93	Selected correlation factor for induced activity
	$5 \cdot 10^{-6}$ Mo-93/Co-60
Half-life = $3.5 \cdot 10^{3}$ years	see Figure 4-24

Calculated activities including both induced activity and contamination of Mo-93 are reported in the Swiss model inventory /Alder and McGinnes 1994/. The correlation factor between Mo-93 and Co-60 has here been evaluated for a selected number of Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste) and the correlation factor vary within the interval 10^{-6}–10^{-5}. /Antilla and Wasastjerna 1989/ have reported calculated activities for a Finish core grid, moderator tank and a moderator tank cover. These activities were used to evaluate correlation factors that vary within the interval $7 \cdot 10^{-7}$–$9 \cdot 10^{-7}$. Activities that are reported for a US PWR core barrel and a shroud /Smith et al. 1978/ gave similar correlation factors, $6 \cdot 10^{-7}$ and $8 \cdot 10^{-7}$, respectively. Calculated activity data for a decommissioned beryllium reflector from the Swedish Ågesta reactor gave a correlation factor of $1 \cdot 10^{-6}$.

The correlation factor between Mo-93 and Co-60 is selected to be $5 \cdot 10^{-6}$ from the evaluated correlation factors given in the Figure 4-24.

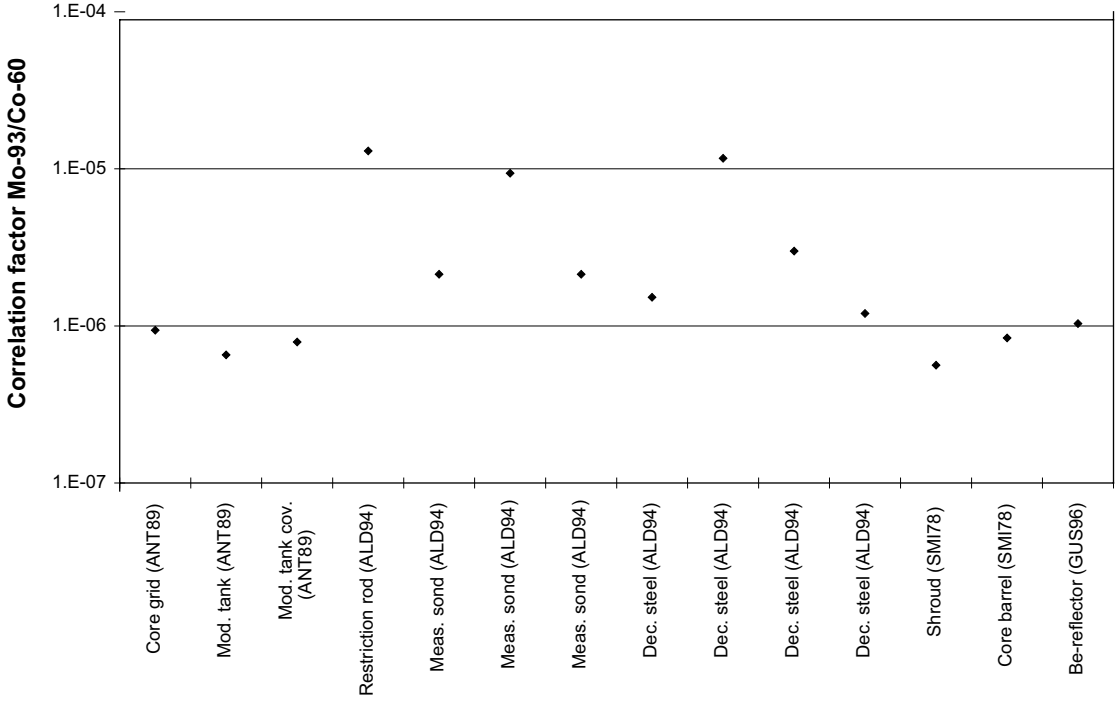

Figure 4-24. *Correlation factors evaluated between Mo-93 and Co-60 for induced activity.*

Tc-99	Selected correlation factor for induced activity
	5.10^{-7} Tc-99/Co-60
Half-life = $2.1 \cdot 10^5$ years	see Figure 4-25

Calculated induced activity in the core grid, the moderator tank and the core spray support in a Swedish BWR after a one year decay period reported in /Lundgren 1982/. These data were used to evaluate correlation factors between Nb-94 and Co-60 to be around 10^{-6}. The calculated induced activity reported for the core grid, moderator tank and moderator tank cover from a Finish BWR /Antilla and Wasastjerna 1989/ has been used to evaluate correlation factors between Tc-99 and Co-60 to be around 10^{-7}. Calculated activity data, including both induced activity and contamination of Tc-99, are reported in the Swiss model inventory /Alder and McGinnes 1994/. From these activities the correlation factor between Tc-99 and Co-60 has been evaluated to be within the interval $6 \cdot 10^{-8}$–$2 \cdot 10^{-6}$ for a selected number of Swiss steel waste types (flow restriction rods, measuring sonds, and medium active steel waste). The evaluated correlation factors are given in Figure 4-25.

The correlation factor between Tc-99 and Co-60 is selected to be $5 \cdot 10^{-7}$.

From data reported for BWR spacer grids (Inconel), control rods (boron steel and zircaloy), and cladding (zircaloy) /Lundgren 1982/ it seems reasonable to use the same correlation for these materials as for steel since the few data points reported fall within the variation interval of the data for steel (see Figure 4-25).

Additional radionuclides

The main contribution of **Be-10** is foreseen from control rods containing boron /Thegerström and Hård 1981/. No correlation between Be-10 and a key radionuclide has been evaluated for induced activity. This radionuclide is included in the SFL 3-5 inventory only for waste types with a reported activity content.

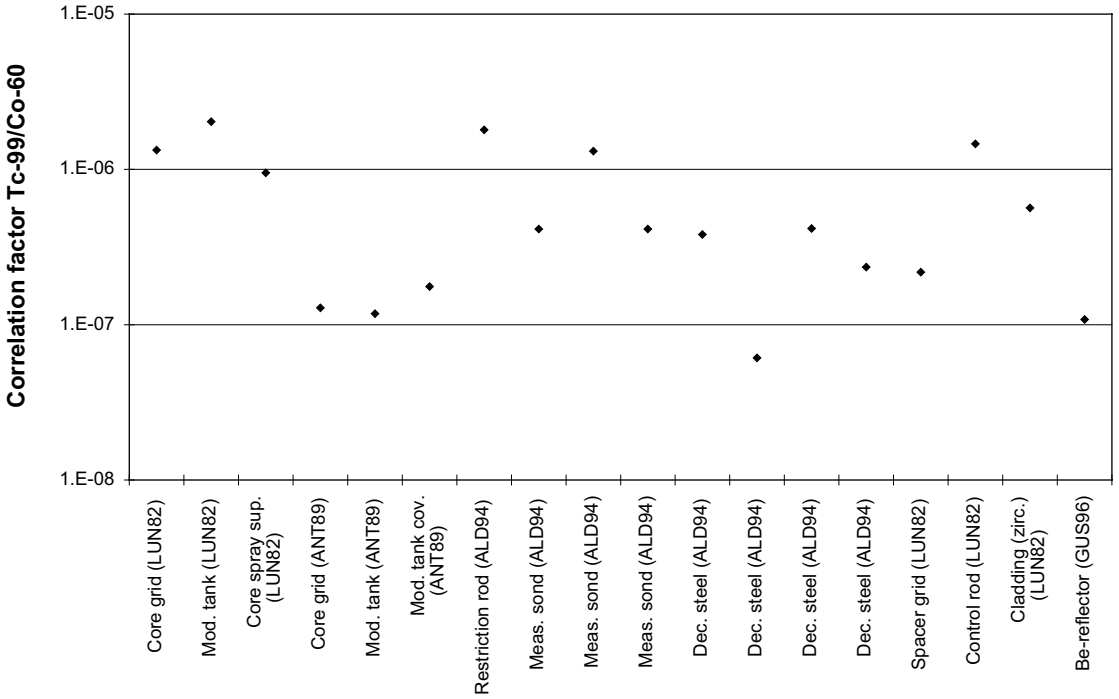

Figure 4-25. *Correlation factors evaluated between Tc-99 and Co-60 for induced activity.*

The major source for **Zr-93** is zircaloy (98% zirconium). A general correlation factor for neutron activated material is not evaluated. This radionuclide is included in the inventory of SFL 3-5 only when the activity is reported for a waste category.

The source for **Ag-108m** and **Cd-113** is mainly the control rods. Silver occurs as impurity in steel, but no data for Ag-108m are available in the literature studied. General correlation factors for Ag-108m and Cd-113 have therefore not been evaluated. These radionuclides are included in the reference activity inventory only for waste types for which the contents of Ag-108m and Cd-113 are reported.

Other radionuclides that may occur due to activation of impurities in steel are e.g. **Sb-125**, **Ba-113**, **Sm-151**, **Eu-152**, **Eu-154** and **Eu-155**. Unfortunately the information on these radionuclides are spars or lacking and no correlation factors have therefore been evaluated.

4.3 Summary of selected correlation factors

A summary of the selected correlation factors for surface contamination (CRUD) and neutron induced activity is given in Table 4-4.

Waste from nuclear facilities

The selected correlation factors (Table 4-4) are to be used in the estimates of the activity content in different wastes from operation, maintains and decommissioning of nuclear facilities when radionuclide specific measurements or calculations are not at hand. The correlation factors were evaluated to be applicable for the SFL 3-5 waste after 1 year of decay.

RMI-waste

To estimate the activity of radionuclides in RMI waste from Studsvik, for which no information from measurements or primary producers is at hand, correlation factors to nuclides for which data are available are used. For some waste categories correlation factors derived for ash-drums

to SFR 1 are applied to estimate the activity of Am-241 and Pu-239+240 from the Cs-137 activity instead of the general factors given in Table 4-4. RMI-waste specific correlation factors are given in Table 4-3.

Table 4-3. Correlation factors derived for ash-drums to SFR 1.

Radionuclide	Half-life (years)	Selected correlation factor Radionuclide/Cs-137
Pu-239+240	$2.4 \cdot 10^4$, $6.5 \cdot 10^3$	0.01
Am-241	$4.3 \cdot 10^2$	0.04

Table 4-4. Correlation factors selected for radionuclides in surface contamination and neutron induced activity in SFL 3-5 waste. The correlation factors are aimed to be representative for waste one year after discharge from the nuclear facility.

Radionuclide	Half-life (year)	Correlation factor			
		Induced activity	Surface contamination		
		key RN: Co-60	Key RN: Co-60	key RN: Cs-137	key RN: Pu-239+240
H-3	12	$1 \cdot 10^{-4}$	$1 \cdot 10^{-4}$		
Be-10	$1.6 \cdot 10^6$		$6 \cdot 10^{-10}$		
C-14	$5.7 \cdot 10^3$	$5 \cdot 10^{-4}$	$1 \cdot 10^{-3}$		
Cl-36	$3.0 \cdot 10^5$	$7 \cdot 10^{-7}$		1.10^{-5}	
Fe-55	2.7	5	1		
Co-60 (key RN)	5.3				
Ni-59	$7.5 \cdot 10^4$	$5 \cdot 10^{-3}$	$1 \cdot 10^{-3}$		
Ni-63	96	1	$2 \cdot 10^{-1}$		
Se-79	$6.5 \cdot 10^4$			$4 \cdot 10^{-6}$	
Sr-90	29			$1 \cdot 10^{-1}$	
Zr-93	$1.5 \cdot 10^6$		$1 \cdot 10^{-6}$		
Nb-93m	14	$1 \cdot 10^{-3}$	$1 \cdot 10^{-3}$		
Nb-94	$2.0 \cdot 10^4$	$1 \cdot 10^{-5}$	$1 \cdot 10^{-5}$		
Mo-93	$3.5 \cdot 10^3$	$5 \cdot 10^{-6}$	$5 \cdot 10^{-6}$		
Tc-99	$2.1 \cdot 10^5$	$5 \cdot 10^{-7}$		$5 \cdot 10^{-3}$	
Pd-107	$6.5 \cdot 10^6$			$1 \cdot 10^{-6}$	
Ag-108m	$1.3 \cdot 10^2$		$6 \cdot 10^{-5}$		
Cd-113m	14			$6 \cdot 10^{-4}$	
Sn-126	$1.0 \cdot 10^5$			$5 \cdot 10^{-7}$	
Sb-125	2.8		$1 \cdot 10^{-1}$		
I-129	$1.6 \cdot 10^7$			$3 \cdot 10^{-7}$	
Cs-134	2.1			1	
Cs-135	$2.3 \cdot 10^6$			$5 \cdot 10^{-6}$	
Cs-137 (key RN)	30				
Ba-133	11		$1 \cdot 10^{-5}$		
Pm-147	2.6			$9 \cdot 10^{-1}$	
Sm-151	90			$3 \cdot 10^{-3}$	
Eu-152	13			$7 \cdot 10^{-5}$	
Eu-154	8.8			$1 \cdot 10^{-1}$	
Eu-155	5.0			$7 \cdot 10^{-2}$	
Ho-166m	$1.2 \cdot 10^3$		$4 \cdot 10^{-6}$		

Radionuclide	Half-life (year)	Correlation factor			
		Induced activity	Surface contamination		
		key RN: Co-60	Key RN: Co-60	key RN: Cs-137	key RN: Pu-239+240
Pb-210	22				$2 \cdot 10^{-11}$
Ra-226	$1.6 \cdot 10^{3}$				$2 \cdot 10^{-10}$
Ac-227	22				$4 \cdot 10^{-9}$
Th-229	$7.3 \cdot 10^{3}$				$3 \cdot 10^{-10}$
Th-230	$7.7 \cdot 10^{4}$				$9 \cdot 10^{-8}$
Th-232	$1.4 \cdot 10^{10}$				$1 \cdot 10^{-13}$
Pa-231	$3.3 \cdot 10^{4}$				$3 \cdot 10^{-8}$
U-232	72				$3 \cdot 10^{-5}$
U-233	$1.6 \cdot 10^{5}$				$2 \cdot 10^{-8}$
U-234	$2.4 \cdot 10^{5}$				$1 \cdot 10^{-3}$
U-235	$7.0 \cdot 10^{8}$				$2 \cdot 10^{-5}$
U-236	$2.3 \cdot 10^{7}$				$3 \cdot 10^{-4}$
U-238	$4.5 \cdot 10^{9}$				$4 \cdot 10^{-4}$
Np-237	$2.1 \cdot 10^{6}$				$4 \cdot 10^{-4}$
Pu-238	88				4
Pu-239 (key RN)	$2.4 \cdot 10^{4}$				
Pu-240 (key RN)	$6.5 \cdot 10^{3}$				
Pu-241	14				$1 \cdot 10^{2}$
Pu-242	$3.8 \cdot 10^{5}$				$3 \cdot 10^{-3}$
Pu-244	$8.3 \cdot 10^{7}$				$7 \cdot 10^{-10}$
Am-241	$4.3 \cdot 10^{2}$				1
Am-242m	$1.5 \cdot 10^{2}$				$1 \cdot 10^{-2}$
Am-243	$7.4 \cdot 10^{3}$				$3 \cdot 10^{-2}$
Cm-243	29				2.10^{-2}
Cm-244	18				3
Cm-245	$8.5 \cdot 10^{3}$				3.10^{-4}
Cm-246	$4.7 \cdot 10^{3}$				8.10^{-5}

5 Uncertainties

Large uncertainties are involved in the use of general correlation factors and contamination levels to estimate the activity content in specific waste types. The use of correlation factors is one possibility to estimate a reference radionuclide inventory for the purpose of safety assessment studies, when waste-type specific information is not at hand. The outcome of the safety assessment will, however, give indications on which radionuclides it is of major concern to reduce the uncertainties in the reference inventory.

The largest uncertainty is related to the general correlation factors selected. The correlation factors evaluated for specific radionuclides vary within orders of magnitude, which gives an indication of the uncertainty interval. In addition, no statistical evaluations have been made in this study to prove that correlations between the key nuclides (Co-60, Cs-137 and Pu-239+240) and the other nuclides exist.

Surface contamination

Contamination levels of key radionuclides for materials in contact with primary coolant water and other water have been defined, based mainly on measurements in Swedish nuclear power units. Differences between PWR's and BWR's, in operational life-times, and in operational procedures result in uncertainties in the activity estimates for individual waste types. The estimated values may, however, be representative as an average for a group of waste types. The contamination level of Co-60 on surfaces in contact with primary coolant water is mainly based on measurements in a BWR, but is here applied to waste from both BWR's and PWR's. Data on contamination levels of Cs-137 is sparse. Reported data for reactor internals gives a scaling factor between Cs-137 and Co-60 of 6%. This value was selected in this study. In other studies, the Cs-137 activity is estimated to be less than 10% of the Co-60 activity. For the selection of contamination level of Pu-239+240 in different waste types both measured data and scaling factors to Co-60 and Cs-137 has been used. The major uncertainties in the selected contamination levels are related to the use of the same values for all BWR and PWR components. No consideration has been taken to the fuel leakage history, the coolant chemistry etc.

Waste from Clab and the encapsulation plant that has been in contact with the contaminated pool water have been assigned the same contamination level as reported for a secondary system in a reactor power unit. In addition, the composition of the surface contamination is assumed to be the same as the composition defined for primary reactor systems. These assumptions lead to uncertainties in the radionuclide inventory for these waste streams.

The reported radionuclide inventory calculated for a BWR spent fuel assembly has been the base for the selection of correlation factors for the radionuclides in the natural decay chains. This assumption is a simplification of the real conditions where the availability of radionuclides in the fuel and the water chemistry plays an important role for the surface contamination due to leakages from defect fuel rods. In addition, changes in operation conditions e.g. higher burn-up and increased enrichment of the fuel may have an influence on the composition of the surface contamination.

Induced activity

Measurements of induced activity in irradiated materials are sparse. In order to estimate the activity content, computer codes developed for estimating the composition of irradiated fuel are often being used. The suitability of these codes, if adapted to calculate the activity in other materials and at other locations, can be disputed. These circumstances may result in significant uncertainties in the radionuclide content in other irradiated materials than spent fuel. In the

calculations the assumed composition of the material including the content of impurities plays an important role. The main constituent of the metals are considered in neutron activation calculations, whereas, impurities in the materials are treated more randomly since the content of impurities is not specified or unknown. If impurities are considered the limits guarantied by the supplier, rather than the real values, are often the only values available. Consideration of impurities may lead to unexpected results and may introduce considerable activity of some radioactive isotopes. Despite these limitations, results from activation calculations have been used to define a set of correlation factors between the key radionuclide Co-60 and other nuclides.

RMI-waste

For some waste categories from Studsvik correlation factors derived for ash-drums to SFR 1 are applied to estimate the activity of Pu-239+240 and Am-241 from the Cs-137 activity. It is uncertain whether these correlation factors are applicable for the waste to SFL 3. For other waste types and other radionuclides, the correlation factors defined as being representative for induced activity and surface contamination in reactor systems are used. Since the waste in Studsvik origin from sources, different from power reactors, the correlation factors for surface contamination and induced activity in reactor systems may not be applicable in all cases.

Time aspects

For radionuclides with half-lives of the order of a few years the time aspect is important for the selection of correlation factors. The duration of exposure, the reference times for measurement and calculations influence the ratio between a relatively short-lived radionuclide and a more long-lived radionuclide with several orders of magnitude.

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