IAEA Nuclear Energy Series





DETERMINATION AND USE OF SCALING FACTORS FOR WASTE CHARACTERIZATION IN NUCLEAR POWER PLANTS

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FOREWORD

Consistent with the IAEA's published safety requirements for Predisposal Management of Radioactive Waste, Including Decommissioning (WS-R-2), radioactive waste disposal requires planned and systematic actions to provide confidence that the assessment of the radioactive inventory of the waste will satisfy given requirements for quality. However, many of the important long lived radionuclides contained in radioactive waste are difficult to measure (DTM) from the outside of waste packages using non-intrusive techniques because they are low energy, beta or alpha emitting nuclides. Identification of these DTM nuclides using complex radiochemical analysis is not practical for large numbers of waste packages.

This report presents international experience with the scaling factor methodology that, in many cases, can be applied to evaluate the radioactive inventory of DTM nuclides in waste packages. This methodology relies on establishing a correlation (scaling factor) between the DTM nuclides and easy to measure (ETM) nuclides. The inventory of the ETM nuclides in a waste package can be derived based upon external gamma radiation measurements carried out on the waste package, and the DTM nuclides can be estimated from the inventory of the ETM nuclides using established scaling factors.

Of specific interest is the extension of the applicability of the scaling factor methodology to the characterization of waste arising from the decommissioning of nuclear facilities, institutional waste and problematic/historical waste that needs to be retrieved from existing storage structures.

This report was prepared at two consultants meetings, in December 2006 and September 2007, and at one technical meeting in September 2007. In total, 18 experts from 16 Member States, and five consultants from five other Member States participated at different stages in the development process.

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The IAEA officers responsible for this report were Z. Drace and P.J.C. Dinner of the Division of Nuclear Fuel Cycle and Waste Technology.

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SUMMARY

The management of radioactive waste requires knowledge of the amounts and concentrations of specific radionuclides in the waste. The particular radionuclides of interest or concern may vary according to the safety case or other requirements of the waste management facility and/or the regulations of the Member State. Many of the radionuclides important for long term management of the waste are difficult to measure (DTM) from the outside of a waste package. Identification and quantification of these DTM nuclides require methods that, in general, involve analysis of waste samples using complex radiochemical analysis to separate the various radionuclides for measurement. This method of direct measurement is generally not practical to employ for large numbers of waste packages or for many heterogeneous waste streams.

An alternative approach that can be used in many cases is to exploit the relationship between some easy to measure (ETM) key nuclides, such as certain gamma emitters, and the DTM nuclides to derive information for the DTM nuclides of interest. The scaling factor (SF) method is such an approach that is widely used to evaluate these DTM nuclides. The SF method is based on developing a correlation between ETM and DTM nuclides. The activities of DTM nuclides in waste packages are then estimated by measuring the ETM nuclides based on gamma measurements from outside the package and applying the SFs to calculate the DTM activities.

The international standard ISO 21238:2007 gives guidelines for the general methodology for empirically determining SFs to evaluate the radioactivity of DTM nuclides in low and intermediate level radioactive waste (LILW) packages.

The objective of this report is to provide information on the international experience in the determination and use of SFs. This report shows actual examples of the ways in which SFs have been derived and applied in various Member States. Although the main focus of the ISO standard is on LILW from the operation of nuclear power plants, the SF technique is also applicable to other situations where it is desirable to infer the activity of DTM nuclides from easy to make measurements, such as for research reactors, nuclear fuel manufacturing plants, nuclear fuel reprocessing plants, decommissioning waste, historical waste, contaminated land, etc. In each case, the technique employed is similar, but consideration must be given to the unique aspects of the situation, such as the radionuclides of concern.

Development of SFs in Member States has been based on analysis of representative waste streams and/or on theoretical calculations and modelling of radionuclide production and transport mechanisms.

There is general consensus among Member States that the results derived from the application of SFs must be reasonable without being overly conservative. For example, if the estimate of a DTM nuclide is overly conservative (i.e. too high), it may prematurely result in the radionuclide capacity or authorized limit of a repository being 'expended' before the repository is physically full. The acceptable degree of conservatism will vary depending on the application.

Many Member States use the same set of key nuclides and apply similar sampling and analysis strategies. There is also an indication that Member States operating similar nuclear facilities (e.g. the same design of nuclear power plant) may be able to pool their data to develop common SFs for certain radionuclides and waste streams. This is especially useful for Member States with smaller nuclear programmes that may not be able on their own to support a large sampling and analysis programme to develop their own SFs or who lack the technical infrastructure to perform the complex measurements required.

The experience compiled from Member States indicates that the development and use of SFs is a widely accepted practice by waste generators, facility operators and regulators in many countries. While the details and scope of SF programmes differ in Member States, the basic techniques and methodologies are very similar. Other Member States are in various stages of developing waste characterization programmes based on SF techniques.

1. INTRODUCTION

1.1. BACKGROUND

Disposal of radioactive waste requires knowledge of its radioactivity content in terms of the concentrations of specific radionuclides. This information is used by the repository operator and/or regulator to track radionuclide inventories and concentrations, which are generally limited by the repository safety assessment and resulting operating licence. It is normally the responsibility of waste generators to evaluate and declare the activity content of their waste. The repository safety assessment typically identifies a relatively small set of long lived radionuclides that are more or less limited by the specific design of the disposal site. Many of the important long lived radionuclides contained in the radioactive waste are difficult to measure (DTM) from outside the waste packages using non-intrusive techniques because they are low energy, non-penetrating beta or alpha emitting nuclides (i.e. non-gamma emitters). Identification of these DTM nuclides requires methods that, in general, involve analysis of waste samples using complex chemical analysis to separate the various radionuclides for measurement. This method is not practical to employ for large numbers of waste packages.

An alternative approach that can be used in some cases is to use the knowledge of gamma emitters, obtained by external gamma measurements, to derive information for the DTM nuclides. The scaling factor (SF) method is one such approach and is widely used to evaluate these DTM nuclides. The SF method is based on developing a correlation between easily measurable gamma emitting nuclides (key nuclides) and DTM nuclides. The activities of DTM nuclides in waste packages are then estimated by measuring the gamma emitting nuclides and applying the SFs [1–7].

The international standard ISO 21238:2007 [8] provides a general methodology for empirically determining SFs to evaluate the radioactivity of DTM nuclides in low and intermediate level radioactive waste packages. This international standard gives general guidelines for the SFs used in the characterization of contaminated waste produced in water cooled nuclear power plants. This standard is also relevant to other types of reactor, such as gas cooled reactors (GCRs).

Of specific interest to the IAEA is the possibility of extending the applications of the SF methodology for characterization of waste arising from the decommissioning of nuclear facilities, institutional waste and problematic/historical waste that needs to be retrieved from existing storage structures. In response to this need, the IAEA prepared this technical report.

In addition to reinforcing and enlarging the concepts discussed in the ISO standard, this report includes international lessons learned in the application of SFs, the pros and cons of analysis on the application of SFs and guidance on how to use and implement the new ISO standard for the characterization of waste. These methodologies and lessons learned can be applied to other situations, such as the non-nuclear power plant wastes mentioned above.

1.2. OBJECTIVE

The objective of this report is to provide information and guidance on applying ISO 21238:2007 for the determination and use of SFs. This report shows actual examples of the ways in which SFs have been applied in various countries, and explains basic technical concepts and terms related to SF methods.

1.3. SCOPE

The SF method was developed from operating experience at nuclear power plants for low and intermediate level radioactive waste. However, this method can be applied to a range of conditions and types of waste and facilities such as:

- (a) Nuclear power plants, test reactors, fuel manufacturing and processing;
- (b) Facility operation and decommissioning period;
- (c) Contaminated waste, activated waste;
- (d) Historical waste.

This is because the basic methodologies described in the ISO standard and this report can be used to demonstrate a correlation between DTM nuclides and selected ETM gamma emitting nuclides in virtually any situation where DTM nuclides and ETM nuclides are co-produced. The method can be extended to other applications, but these are not discussed directly in this report.

1.4. STRUCTURE

This report consists of four technical sections and an annex. Section 2 describes the basis of SFs for DTM nuclides such as ¹⁴C, ⁶³Ni, etc., and ETM key nuclides such as ⁶⁰Co or ¹³⁷Cs.

In Section 3, the basic application methodology of SFs and a summary of each country's actual practices are described. A summary is shown comparing each of the main technical items of the SF method. (The details of each country's practices are described in the Annex.)

In Section 4, several examples of experience and knowledge are discussed; many important experiences are gathered from the development of the SF method in each country. Section 4 also discusses the various factors that may influence SFs, such as the effects of plant materials, fuel failures and waste treatment methods.

Section 5 describes the quality management requirements in each step of development and usage of SFs. It is important to keep quality control of sampling, radiochemical analysis and evaluation of SF values to ensure accuracy and reduce uncertainty. The key issues are discussed in this section.

1.5. KEY DEFINITIONS

- **activation product (AP).** A radionuclide produced by neutron activation. For example, in decommissioning waste comprising structural materials from a nuclear facility, APs might typically be found primarily within the matrix of the material.
- **auxiliary key nuclides.** These are nuclides that can be measured via radiochemical analysis or are scaled from primary key nuclides such as ⁶⁰Co and are used to calculate other DTM nuclides in a two step process.
- **correlation coefficient (CC).** A CC is a number between -1 and 1 that measures the degree to which two variables are linearly related. If there is perfect linear relationship, with positive slope between the two variables, we have a CC of 1; if there is positive correlation, whenever one variable has a high (low) value, so does the other. A CC of 0 means that there is no linear relationship between the variables. Typically values of the CC above 0.6 have been taken to indicate evidence of significant positive correlation.
- corrosion product (CP). A radionuclide produced by neutron activation of products of material corrosion that are transported through the reactor core. Example: ⁶⁰Co, ⁶³Ni. Note: Adapted from ISO 21238:2007 [8]. Example: ³H, ¹⁴C, ⁶⁰Co. Note: An activated CP is also an AP, but not all APs are CPs. Note: Adapted from Ref. [9].
- **difficult to measure (DTM) nuclides.** A radionuclide whose radioactivity is difficult to measure directly from the outside of the waste package by non-destructive assay means. Example: Alpha emitting nuclides, beta emitting nuclides and characteristic X ray emitting nuclides. Note: Adapted from ISO 21238:2007 [8].

dry active waste (DAW). Solid waste generated in various waste streams in a nuclear facility, including protective clothing, replaced equipment, parts, plastics, polyvinyl chloride sheets and high efficiency particulate air filters removed during plant operation and maintenance. Note: Adapted from ISO 21238:2007 [8].

IAEA-TECDOC-1492 (dry solid waste): All waste which was not generated as a result of liquid treatment

processes, including combustible solids, metal, plastics, concrete and similar dry wastes [11].

- **easy to measure (ETM) nuclides.** Gamma emitting nuclide whose radioactivity can be readily measured directly by non-destructive assay means.
- fission product (FP). A radionuclide produced either by fission or by the subsequent radioactive decay of radionuclides thus formed.
 Note: Adapted from ISO 921:1997 [10].
 Example: ¹³⁷Cs, ⁹⁰Sr.
 Glossary 2003 Edition: A radionuclide produced by nuclear fission [9].
- **heterogeneous waste.** Radioactive waste that does not meet the definition of homogeneous waste. This includes solid components and mixtures of solid components such as DAW and cartridge filters. Note: Adapted from ISO 21238:2007 [8].

Example: ${}^{239/240}$ Pu scaled from 60 Co used to calculate other actinides, based on their theoretical ratios to ${}^{239/240}$ Pu.

homogeneous waste. Radioactive waste that shows an essentially uniform distribution of activity and physical contents.

Example: Flowable wastes such as concentrates, solidified liquids and spent resins in which the radioactivity may reasonably be assumed to be uniformly distributed over the volume or flowable wastes uniformly mixed with a solid matrix.

Note: Adapted from ISO 21238:2007 [8].

key nuclide. A gamma emitting nuclide whose radioactivity is correlated with that of DTM nuclides and can be readily measured directly by non-destructive assay means.
 Note: Also called 'easy to measure nuclide' or 'marker nuclide'. Example: ⁶⁰Co and/or ¹³⁷Cs.

Note: Adapted from ISO 21238:2007 [8].

scaling factor (SF). A factor or parameter derived from a mathematical relationship used in calculating the radioactivity of a DTM nuclide from that of an ETM key nuclide as determined from sampling and analysis data.

Note: Adapted from ISO 21238:2007 [8].

transuranic nuclide. A radionuclide with atomic numbers above 92.

2. SCALING FACTOR BASICS

2.1. BASIC PHILOSOPHY

The radionuclides produced in a nuclear reactor are carried by the fluids that are flowing within the nuclear reactor plant systems. Some of the radionuclides are collected in radioactive waste such as ion exchange resins generated from reactor water purification systems, concentrated liquids produced by evaporating various

kinds of process water from the plant systems, and some which adhere to the inner surfaces of the equipment of the plant systems, are present in DAW generated when performing maintenance or when replacing equipment components. Such waste may include the component itself or various other materials, such as worker's protective clothing, that become contaminated through contact with the contaminated items.

The SF method is a technique for evaluating the concentration of DTM nuclides, which are typically represented by beta emitting and alpha emitting nuclides, such as ¹⁴C, ⁶³Ni and ²⁴⁰Pu, that exist in these radioactive wastes. In the reactor, key nuclides (gamma emitting nuclides such as ⁶⁰Co and ¹³⁷Cs, which are measurable from outside of the waste package) are generated concurrently with DTM nuclides and transfer within the plant systems. In the SF method, the concentrations of DTM nuclides in radioactive wastes are evaluated by multiplying the concentrations of the key nuclide by the coefficients calculated based upon radio-nuclide data obtained by sampling and radiochemical analysis (i.e. the SFs, which are the ratios of radioactive concentrations between DTM nuclides and key nuclides).

SFs derived from empirical data provide an effective tool for evaluating the activity concentrations of radioactive wastes. Radionuclide analysis data obtained from the study of actual radioactive wastes or other nuclear power plant radiological data are used in considering the various factors affecting the ratios of concentrations between DTM nuclides and key nuclides, which are expected during the course of radionuclide transport from the production stage into radioactive wastes as the final destination. These factors include:

- (a) Similarity in production mechanism;
- (b) Similarity in transport behaviour within plant systems.

The SF method is a very useful radioactivity evaluation technique that has potential for a wide range of application, not only for nuclear power plants, where this method is regularly applied, but also for other nuclear facilities. In applying the empirical SF method, however, it is necessary to judge its applicability through evaluation based on actual analysis of the target radioactive wastes.

SFs can be calculated from measurements of radionuclides obtained through appropriate radiochemical analysis, through modelling code calculation or by a combination of both techniques.

2.1.1. Selection of difficult to measure key nuclide pairs

The DTM nuclides present in nuclear power plant radioactive waste that are typically required to be characterized are listed in Table 1, along with selected gamma emitters typically used as the key nuclide. As shown, the radionuclides differ not only in their means of production but also in their physicochemical characteristics (speciation, solubility, etc.). A DTM nuclide such as ⁶³Ni, which is produced by activation, can be expected to correlate with a key nuclide such as ⁶⁰Co, which is also produced by activation and has physicochemical characteristics similar to it. Similarly, an FP such as ¹²⁹I has a production method and behaviour similar to the ETM nuclide of FP ¹³⁷Cs. Therefore, ⁶³Ni can be expected to correlate with ⁶⁰Co, and similarly ¹²⁹I with ¹³⁷Cs. While this behaviour is generally expected, the underlying basis of the method remains empirical. That is, if a reliable correlation can be developed between a DTM nuclide and a key nuclide, the SF method can be applied with reasonable certainty.

In practice, the available key nuclides, particularly those with sufficiently long half-lives, are limited to a select few, such as ⁶⁰Co, ¹³⁷Cs, ¹⁴⁴Ce and ¹²⁵Sb. Therefore, only a few of the DTM nuclides can be logically paired with the available key nuclides based strictly on the similarity of their production mechanisms and their physic-ochemical characteristics.

For the other DTM nuclides, the SF methodology reverts to purely empirical relationships. For example, ¹⁴C and ⁹⁰Sr may be typically correlated with ⁶⁰Co, even though the paired radionuclides do not have much in common with respect to their production route or chemical similarity. In these instances, as long as a correlation can be demonstrated to exist, the DTM nuclide can still be inferred reliably from the concentration of the key nuclide.

Often, it is convenient to correlate ²³⁹⁺²⁴⁰Pu with ⁶⁰Co or ¹³⁷Cs and mutually correlate the other transuranic nuclides such as ²⁴¹Am and ²⁴⁴Cm with ²³⁹⁺²⁴⁰Pu. In this respect, the ratios ²⁴¹Am/²³⁹⁺²⁴⁰Pu and ²⁴⁴Cm/²³⁹⁺²⁴⁰Pu do not conform to the typical definition of an SF, the latter being defined earlier as the ratio of a DTM nuclide to a key nuclide. Pu-239+240 is used as an auxiliary key nuclide in this case.

Radionuclide	Half-life	Principal radiation emitted	Prevalent means of production	Physical/chemical form
H-3	12.3 a	$\beta^{-} \left(E_{\max} = 18.6 \text{ keV} \right)$	Li-6(n, α) H-2(n, γ) Ternary fission	Tritiated gas and water
C-14	5730 a	$\beta^- (E_{\rm max} = 156 \text{ keV})$	C-13(n, γ) N-14(n,p) O-17(n, α)	Gas and inorganic carbonate/ bicarbonate
Cl-36	3.01×10^5 a	$\beta^{-}(E_{\rm max} = 708.6 {\rm keV})$	Cl-35(n,γ)	Soluble, anionic
Ca-41	1.03×10^5 a	e⁻ (2.97 keV) X (3.31 keV) X (3.59 keV)	Ca-40(n,γ)	Soluble, cationic
Fe-55	2.74 a	X (5.90 keV) X (6.49 keV)	Fe-54(n,γ)	Mostly insoluble
Ni-59	7.6×10^4 a	X (6.93 keV) X (7.65 keV)	Ni-58(n,γ)	Mostly insoluble
Co-60	5.27 a	γ (1173.2 keV) γ (1332.5 keV)	Co-59(n,γ)	Mostly insoluble
Ni-63	100.1 a	$\beta^{-}(E_{\rm max} = 66.9 {\rm keV})$	Ni-62(n, γ)	Mostly insoluble
Se-79	2.95×10^5 a	$\beta^{-}(E_{\rm max} = 151 \text{ keV})$	Fission FP activation: Se-78(n, γ)	Soluble, anionic
Sr-90	28.9 a	$\beta^{-}(E_{\rm max} = 546 \text{ keV})$	Fission	Largely soluble, cationic
Nb-94	2.03×10^4 a	γ (702.6 keV) γ (871.1 keV)	Nb-93(n,γ)	Generally insoluble
Tc-99	2.11×10^5 a	$\beta^{-}(E_{\rm max} = 293.5 {\rm keV})$	Fission Mo-98(n,γ)Mo-99(β⁻)	Soluble (+7), anionic Insoluble (+4)
Ru-106	373.6 d	$\gamma (511.9 \text{ keV})^{a} \ \gamma (621.9 \text{ keV})^{a} \ \gamma (1050.4 \text{ keV})^{a}$	Fission	Soluble, anionic
Sb-125	2.76 a	γ (427.9 keV) γ (600.6 keV) γ (635.9 keV)	Fission Sn-124(n, γ)Sn-125(β^-)	Soluble, anionic
I-129	1.57×10^7 a	X (29.8 keV) X (29.5 keV) X (33.6 keV) γ (39.6 keV) X (34.4 keV)	Fission	Soluble, anionic and gas
Cs-135	2.3×10^6 a	$\beta^{-}(E_{\rm max} = 268.7 {\rm ~keV})$	Fission	Very soluble, cationic
Cs-137	30.08 a	γ (661.6 keV)	Fission	Very soluble, cationic
Ce-144	284.9 d	γ (133.5 keV) γ (696.5 keV) ^a γ (80.1 keV) γ (2185.7 keV) ^a	Fission	Generally insoluble

TABLE 1. PERTINENT DATA FOR VARIOUS RADIONUCLIDES OF INTEREST [12]

Radionuclide	Half-life	Principal radiation emitted	Prevalent means of production	Physical/chemical form
U-235	7.04×10^8 a	α (4.398 MeV) α (4.366 MeV) α (4.215 MeV) γ (185.7 keV) γ (143.8 keV)	Naturally occurring	Generally insoluble
Np-237	2.14 × 10 ⁶ a	α (4.788 MeV) α (4.771 MeV) α (4.767 MeV) γ (29.4 keV) γ (86.5 keV)	U-235(n,γ) U-236(n,γ) U-237(β ⁻)	Generally insoluble
U-238	4.47×10^9 a	α (4.198 MeV) α (4.151 MeV)	Naturally occurring	Generally insoluble
Pu-238	87.7 a	α (5.499 MeV) α (5.456 MeV)	Np-237(n,γ) Np-238(β^{-}) Cm-242(α)	Generally insoluble
Pu-239	24 110 a	α (5.157 MeV) α (5.144 MeV) α (5.106 MeV)	U-238(n,γ) U-239(β ⁻) Np-239(β ⁻)	Generally insoluble
Pu-240	6561 a	α (5.168 MeV) α (5.124 MeV)	Multiple neutron capture	Generally insoluble
Pu-241	14.29 a	$\beta^{-} (E_{\text{max}} = 20.78 \text{ keV})$ $\alpha (4.896 \text{ MeV})$ $\alpha (4.853 \text{ MeV})$	Multiple neutron capture	Generally insoluble
Am-241	432.6 a	α (5.486 MeV) α (5.443 MeV) γ (59.5 keV) γ (26.3 keV)	Pu-241(β ⁻)	Generally insoluble
Pu-242	3.75×10^5 a	α (4.902 MeV) α (4.858 MeV)	Multiple neutron capture	Generally insoluble
Cm-242	162.8 d	α (6.113 MeV) α (6.069 MeV)	Multiple neutron capture followed by $Am-242(\beta^{-})$	Generally insoluble
Cm-243	29.1 a	α (5.785 MeV) α (5.742 MeV) α (5.992 MeV)	Multiple neutron capture	Generally insoluble
Cm-244	18.1 a	α (5.805 MeV) α (5.763 MeV)	Multiple neutron capture	Generally insoluble

^a Decay energy is from the short lived daughter product.

For the purpose of the discussion in this report, any useful relationship between a pair of radionuclides will be deemed to represent an SF.

Owing to the underlying mechanisms for the formation and release of fission and APs, it is feasible to predict values for a number of SFs, recognizing that departure from these values may occur because of plant specific factors. The approaches employed are as follows.

(a) Predictions based on activation models [13, 14].

The relative activity ratio of two radionuclides produced by activation can be simply estimated from:

$$A_2 = \frac{\sigma_2 \lambda_2 M_2}{\sigma_1 \lambda_1 M_1} A_1$$

where:

- λ is the radioactive decay constant (s⁻¹);
- M is the moles of inactive element in-core;
- σ is the thermal absorption cross-section for the parent nuclide (barns).

Note that the above equation is solely based on the production of an activation nuclide by neutron capture and does not address the fate of the radionuclide as a result of processes such as transport, deposition and ion exchange. However, for SFs such as ⁵⁵Fe/⁶⁰Co and ⁶³Ni/⁶⁰Co that involve radionuclides with similar properties, the above equation provides estimates that are in reasonable agreement with measured values.

(b) Predictions based on FP release models.

FP release models [15, 16] are based on a description of the release process for FPs from defective fuel rods and from uranium contamination present on in-core reactor surfaces. The release of any chemical element from defective fuel is dictated by its ability to diffuse through the grain (R_d) and then vaporize from the grain surface (R_v) , where the overall release rate R equals the minimum of R_d and R_v . If $R_v >> R_d$, the rate determining step is diffusion in the fuel matrix. Mobile species, which accumulate in the fuel clad gap, are transported from the gap into the coolant via the defect site. This second transport step has been postulated to be a first order rate process. In contrast to the release processes from defective fuel, FPs are also released directly by recoil from uranium contamination present on core surfaces. Based on the application of such release models, it is possible to estimate ratios such as ¹²⁹I/¹³⁷Cs and ⁹⁹Tc/¹³⁷Cs in reactor coolant, which in turn can be used in conjunction with simple transport models to predict the SFs in various process wastes.

(c) Predictions based on estimated radionuclide inventories in used fuel [13, 14].

Finally, ratios of fuel based radionuclides can be estimated from compilations of specific activity data for used fuel. Such compilations, which are based on ORIGEN or similar code calculations, are generally available. Experimentally determined values for ratios such as ²³⁸Pu/²³⁹⁺²⁴⁰Pu and ²⁴⁴Cm/²³⁹⁺²⁴⁰Pu in the waste are typically in agreement with estimates based on the radionuclide specific activities in used fuel, provided burnup and decay time are appropriately considered. Estimated values for ¹²⁹I/¹³⁷Cs are also found to be in agreement with measured values in instances where tramp uranium is the dominant contributor for both radionuclides. In general, however, departure from the theoretical estimates may be expected if the paired radionuclides manifest dissimilar behaviours during processes such as transport, deposition and ion exchange.

2.1.2. Transport behaviour considerations

Even if there is no similarity in the production mechanism of different radionuclides, the composition ratios between various kinds of radionuclides in the reactor coolant are generally kept in steady state as long as the reactor operating conditions, such as fuel integrity, are stable. Both DTM and ETM nuclides are carried by fluids flowing through the nuclear power plant systems. Therefore, even in the case of a combination of radionuclides whose production mechanisms are different, the SF method is also applicable on the condition that the

transport processes for the radionuclides are either similar or are in equilibrium with each other, resulting in a constant ratio.

For example, the production mechanisms for ²³⁸Pu, as an alpha emitting nuclide, and ⁶⁰Co, as a key nuclide, are quite different. The alpha emitting nuclides such as ²³⁸Pu are produced by neutron multiple captures in uranium fuel, and key nuclides such as ⁶⁰Co are produced from cobalt elements contained in reactor component materials being activated by neutrons. However, neutrons in the reactor constitute a fundamental production source of both radionuclides, and the composition ratio between these radionuclides in the reactor remains constant under stable reactor operation conditions. For this reason, their composition ratio in radioactive waste will show a constant value as long as the transport behaviours during the transit within the plant systems from their origin in the reactor to the final radioactive waste for the two radionuclides are in equilibrium.

In particular, water cooled nuclear plants have adopted a system in which the heat generated by nuclear reactions is conducted by water flowing in the systems. For this reason, solubility in the water is listed as one of the properties of the radionuclides that greatly affect the similarity in the transport behaviour of radionuclides moving within the plant systems.

Different radionuclides behave differently in adsorption and filtration by ion exchange resins and filter elements and adhesion to DAW. This is because the differences in their solubility contribute greatly to the ratio of concentration of radionuclides. For example, highly insoluble radionuclides such as alpha emitting transuranic nuclides have a tendency to adhere to the surfaces of DAW, and ⁶⁰Co, as a transition metal element, also has high insolubility, thus giving it a similar adhesion rate. The composition ratios between ⁶⁰Co and the alpha emitting nuclides will keep constant during the process of transport from the radionuclide production stage in the reactor to the final stage of radioactive waste. (Surface adhesion is more strongly affected by electrochemical properties. Plutonium is generally more electropositive than cobalt and adheres more strongly. The transport of plutonium is delayed by this process.)

The applicability of the SF method is based on the premise that the conditions under which radionuclides are produced is constant. When fuel failure occurs during reactor operation, the activity of alpha emitting nuclides, such as ²³⁸Pu, and volatile FPs, such as iodines, in the reactor water increases as a function of failure defect size, and consequently the composition ratios between these radionuclides and ⁶⁰Co, which is not affected by fuel failure, will show fluctuations. For this reason, when the SF method is applied to a combination of radionuclides having different production mechanisms, it is necessary that considerations such as grouping reactor plants based upon the fuel failure history or other factors should be considered when any fundamental factor of the plant operation has fluctuated.

2.2. BASIS FOR EVALUATION

The concentrations of radionuclides found in nuclear power plant waste streams often show concentration ranges extending over several orders of magnitude. As an example, Fig. 1 shows the ranges of the concentrations of ⁶³Ni and ⁶⁰Co from a Japanese boiling water reactor (BWR). As shown in this figure, both radionuclides show a log-normal distribution.

In addition, DTM nuclides such as ⁶³Ni and key nuclides such as ⁶⁰Co have similar characteristics in terms of the radionuclide production mechanism and transport behaviour; therefore their composition ratio also shows a log-normal distribution, as shown in Fig. 1. From this fact, it is seen that the concentrations of DTM nuclides and key nuclides have the characteristic feature of showing a mutually correlative log-normal distribution [17] (i.e. bivariate log-normal distribution [18]).

In this example the concentrations of radionuclides show a log-normal distribution. To make an appropriate evaluation of this characteristic, it is appropriate that the logarithmic scale should be applied to the axes of scatter diagrams to be used for data analysis and evaluation.

In connection with the axes of these scatter diagrams, Fig. 2 shows the same data plotted on diagrams of two different scales:

- (a) An arithmetic scale;
- (b) A logarithmic scale.



FIG. 1. Distribution characteristic of radionuclide concentrations and composition ratio. (a) Radioactivity concentration distribution of ${}^{63}Ni$; (b) radioactivity concentration distribution of ${}^{60}Co$; (c) composition ratio distribution (${}^{63}Ni$ / ${}^{60}Co$).



FIG. 2. Comparison of arithmetic and logarithmic scale scatter plots. (a) Scatter diagram on an arithmetic scale; (b) scatter diagram on a logarithmic scale.

As can be seen in Fig. 2, it is impossible to grasp the distribution characteristics of the radionuclide from the scatter diagram created on the arithmetic scale, to say nothing of the correlation between the different radionuclides. On the other hand, the scatter diagram on the logarithmic scale makes it possible to clearly grasp the correlation between radionuclides. What is important here is that both the concentration ranges and the SF ratios exhibit log-normal distributions.

3. SCALING FACTOR METHOD APPLICATIONS

3.1. APPLICATION PRINCIPLES

3.1.1. General

Using the SF method, the activity concentrations of DTM nuclides in waste packages are estimated by using a correlation between their activity concentrations and those of ETM key nuclides such as ⁶⁰Co.

One approach to the application of the SF method is to perform an empirical evaluation, in which the combined effects of various factors (e.g. variations in the amounts of radionuclides produced by nuclear reactions, transport behaviour and physicochemical properties of those radionuclides) are accounted for by the use of representative logarithmic mean values. These are derived by combining data from analysis (e.g. radio-chemical) of a series of samples from actual radioactive wastes.

Figure 3 shows a basic flowchart for application of the SF method. The evaluation and application will typically proceed through the steps shown in the figure [8].



FIG. 3. Basic flow for application of the SF method.

3.1.2. Development of a sampling plan (step 1)

For proper use of the SF approach, especially where data from multiple power plants are combined, it is important to evaluate and investigate the characteristic differences between power plants and the properties (e.g. homogeneity) of the radioactive waste subject to sampling. Some plant characteristics important to this process are:

- (a) Reactor type;
- (b) Reactor component materials;
- (c) Fuel performance history;
- (d) Mechanism through which radionuclides are produced;
- (e) Variations in waste treatment;
- (f) Variations in reactor coolant chemistry;
- (g) Plant operational condition and history.

In this step, plant characteristics and other factors such as waste streams that affect the composition ratios between DTM nuclides and key nuclides (i.e. SF) are studied, and SF classifications based on SF variability due to these factors are assessed. In addition, a sampling plan is developed covering the grouping of nuclear power plants, waste streams and range of activity concentration of wastes. This methodology is also relevant in the approach for deriving SFs for the decommissioning stage and for other nuclear facilities.

3.1.3. Sampling and analysis (step 2)

Appropriate sampling is carried out in accordance with the sampling plan developed by the studies shown in step 1. The selected radioactive wastes (e.g. representative samples) are sampled, and radionuclide analysis data are collected through radiochemical analysis of the samples. These data are compiled into a database or spreadsheet for analysis.

3.1.4. Evaluation of applicability (step 3)

Using the radionuclide analysis data, the correlation between DTM nuclides and the key nuclides is observed through the use of log–log scatter diagrams. SF groupings are studied considering the influencing factors studied in step 1.

The applicability of the SF method for a particular grouping is determined based on whether there is an observable correlation between the concentrations of DTM nuclides and selected key nuclides.

If there is no visually apparent correlation in the scatter plots, the data should be segregated by stream and examined in greater detail. It may be necessary in this case to calculate a representative mean value for the SF for each stream. Again, since it is a basic premise that the data have a log-normal distribution, a geometric mean or log-mean value would be most appropriate. A simple arithmetic mean or average would be strongly affected by the highest values and, while it would envelop all of the data, it may be overly conservative (refer to Section 4.2.2).

The possibility of integration or classification of waste streams and power plants is determined based on, for example, observation of data plots on scatter diagrams.

3.1.5. Determining scaling factor and evaluation of radioactivity (step 4)

SFs are determined, for example, from the log-mean average of the ratio of the DTM to the key nuclide, from regression analysis of the paired data or from other calculational methods. This results in a series of radionuclide specific SFs for various waste categories or waste streams. Details of the various calculation methods are given in the country examples in the Annex to this report.

The activity concentrations or total activity of key nuclides in each waste package to be assayed are determined by gamma spectroscopy or by measuring the surface dose rate of the waste package and calculating a key nuclide activity using a dose rate to activity conversion calculation, or by other appropriate measures, such as theoretical calculations. The activity concentrations of DTM nuclides are calculated based on the radionuclide specific SFs and the appropriate key nuclides activity for each waste package.

3.2. SUMMARY OF APPLICATION PRACTICES IN SELECTED MEMBER STATES

The SF development and usage practices in various Member States are summarized in Tables 2–4. Details of the practices in these Member States can be found in the Annex. Where a specific Member State is referred to below, consult the appropriate section of the Annex for details of the practice.

Table 2 summarizes the reactor types, waste streams, disposal programme status and SF methods used in the Member States that participated in the development of this report.

It can be seen that Member States with existing disposal facilities for nuclear power plant low and intermediate level waste (LILW), such as France, Japan, Slovakia, Spain, Sweden, the UK and the USA, all employ some form of the SF process for declaring DTM nuclides in waste packages. In addition, many Member States that are in the planning, approval or construction stages for new disposal facilities, such as Canada, Germany, Republic of Korea, Mexico and Lithuania, are also employing SF methods for waste characterization and reporting purposes. The needs of each Member State's waste characterization and SF programme are driven by the disposal concept they have selected and by the regulatory requirements of the Member State. It can be expected that as the new facilities are licensed and come into service, the waste characterization programmes in these Member States, including development and use of SFs, will be further refined as required to support the facility.

Table 2 also shows that SF methods are being applied to all nuclear power plant types: BWRs, pressurized water reactors (PWRs), GCRs, water cooled, water moderated power reactors (WWERs), high power channel type reactors (RBMKs), pressurized heavy water reactors (PHWRs) and others. They are primarily being applied to operating reactors, but are also being used and/or developed for shutdown reactors (e.g. in Italy), reactors undergoing decommissioning (e.g. in Japan, Spain, Slovakia, USA) and other nuclear facilities (e.g. fuel cycle and industrial facilities in China). As described below, some special considerations may be required for

reactors undergoing decommissioning, as well as those which may have undergone a major fuel failure or contamination incident.

Table 3 summarizes the sampling practices, calculation model and data maintenance practices in various Member States. All Member States employ sampling at some stage in the development process; most Member States sample by waste stream. The details of the waste streams sampled for various Member States are given in the Annex. Additionally, many Member States with multiple nuclear power plants of different reactor types, such as Germany, Japan, Republic of Korea, Slovakia, Spain, Sweden, the UK and the USA, also sample by nuclear power plant. Some Member States, such as Belgium and Canada, also sample from various nuclear power plants and waste streams to confirm predictive models.

Most Member States use some form of mean value to calculate the SFs, either an arithmetical mean (such as Japan, Slovenia and the UK) or a geometric (logarithmic) mean (such as Brazil, Canada, France, Republic of Korea, Italy, Spain, Sweden, Ukraine and the USA). The 'mean value' SF is based on the assumption that the relationship between a key nuclide and a DTM nuclide is linear over the range of activities of interest. The arithmetical mean will tend to produce a conservative (i.e. high) value, while the geometric mean will tend to produce a more representative average value when the data points are spread over several orders of magnitude.

Other Member States, such as Germany, Hungary, Lithuania, Mexico and Slovakia, use a regression of logarithms analysis to calculate SF values. SFs based on this calculation method assume a non-linear relationship between the key nuclide and the DTM nuclide. As such, they can be used to more accurately model complex, non-linear relationships between the radionuclides.

Table 4 summarizes the key nuclides and data grouping practices in Member States. Most Member States presented in the table have standardized on the use of ⁶⁰Co as the key nuclide for scaling activated CP nuclides and ⁶⁰Co or ¹³⁷Cs for scaling FP nuclides. However, there is less standardization with actinides: ⁶⁰Co, ¹³⁷Cs, ¹⁴⁴Ce and ^{239/240}Pu have all been used as key nuclides in different Member States. In all cases, the important point to consider is whether or not the key nuclide and the DTM nuclide show an 'acceptable' correlation for the given circumstances.

In some Member States, such as Canada, Spain and the USA, a two step process is used to determine actinide activity. For example, ^{239/240}Pu or ²⁴¹Pu is scaled to the primary key nuclide (e.g. ⁶⁰Co), then other actinides are scaled to the calculated ^{239/240}Pu or ²⁴¹Pu value based on theoretical ratios, for example those found in spent fuel. In this case, the ^{239/240}Pu or ²⁴¹Pu becomes an 'auxiliary key nuclide' because it is used as an indirect key for calculating other DTM nuclides.

There are some cases where no reliable correlation has been found between a key nuclide and a DTM nuclide. For example, development of SFs for tritium and ¹⁴C has been problematic in several Member States, such as the USA, for some waste streams. In many of these cases, average values, theoretical values or upper bound values have been used instead of SF calculated values.

Some Member State SF programmes, such as those in Belgium and Canada, also include development of theoretical SFs based on physicochemical processes and models for certain radionuclides that are difficult to sample or for activities that are difficult to determine radioanalytically (e.g. below the detection limit in actual samples).

There are two main styles of data grouping. Most Member States, especially those with several reactor types (such as Germany, Japan, Republic of Korea, the UK and the USA), will calculate reactor type and waste stream specific SFs. Other Member States, particularly those with only one type of reactor (such as Canada) or only one reactor (such as Slovenia), will group only by waste stream under the assumption that a given waste stream from similar reactors will have similar characteristics.

The measurement of the key nuclides in waste packages is generally performed by one of two methods:

- (a) Direct gamma assay (at the energy level associated with the key nuclide);
- (b) Dose rate to activity conversion, where a gross gamma dose rate is measured on the waste package and is used to infer the activity of the key nuclide via a calculational model.

Country	Reactors	Relevant waste streams	Disposal programme	Scaling factor methods	Comment
Belgium	PWR	Ion exchange resins, evaporator concentrates, DAW	Planning	Calculation by modelling	Details are shown in Annex A–1
Brazil	PWR	Compacted DAW, non-compactable DAW, evaporator concentrates, ion exchange resin, sludge, cartridge filters	Planning	Planned programme to be based on direct measurements	Detail is shown in Annex A–2
Canada	PHWR	Incinerator ash, ion exchange resins, filters and filter cartridges, sludge, compacted and non-compacted DAW	Deep geologic disposal under licensing for LILW	Waste stream specific, based on measured data, calculated data and modelling for specific radionuclides	Details are shown in Annex A-3
China	Current generation PWR, PHWR and industrial, fuel cycle facilities	Nuclear power plant, decommissioned fuel cycle, retrieval wastes	Operating low level waste (LLW) site not usable for power plant wastes, new sites in planning	Under study	Details are shown in Annex A-4
France	PWR	Ion exchange resin, filters, sludge, DAW, evaporate concentrates, scraps	Operating a surface repository for very low level waste (VLLW) Operating a near surface repository for LILW	Based on direct measurement	Details are shown in Annex A–5
Germany	PWR, BWR	Compacted DAW, DAW (non-compactable), evaporator concentrates, ion exchange resin, sludge, cartridge filters	Previously operated disposal facilities (now shut down) New deep mine disposal facility under construction	Common SFs based on plant type and waste streams	Details are shown in Annex A–6
Hungary	WWER	Compacted DAW, non-compactable DAW, evaporator concentrates, ion exchange resin, sludge	Under construction	Based on direct measurement	Details are shown in Annex A-7
India	BWR, PHWR	Waste streams include resins fixed in polymer matrix, sludge from low level liquid waste treatment fixed in cement, DAW	Permanent near surface disposal facility (NSDF) on each plant site	Under study	Details are shown in Annex A–8

TABLE 2. SUMMARY OF SCALING FACTOR PROGRAMMES IN VARIOUS MEMBER STATES

commissioning plants inter cartridge filters, sludge, ncentrates ncentrates rator concentrates filters, sludge, rator concentrates filters, sludge, oncentrates, oncentrates, evaporator on-combustible AW	Relevant waste streams	Disposal programme	Scaling factor methods	Comment
BWR, PWREvaporator concentrates, ion exchange and GCR in decommissioningEvaporator concentrates, ion exchange resin, filter sludge, filter cartridge decommissioninghic of KoreaPWR, PHWRIon exchange resin, filters, sludge, DAW, evaporate concentrateshis of KoreaPWR, PHWRIon exchange resin, filters, sludge, DAW, evaporator concentratesniaRBMKIon exchange resin, sediments, perlite, DAW, filters, evaporator concentratesoBWRIon exchange resin, filters, sludge, DAW, filters, evaporator concentratesiaWWRIon exchange resin, filters, sludge, DAW, filters, evaporator concentratesoBWRIon exchange resin, filters, sludge, DAW, evaporator concentratesoBWRIon exchange resin, filters, sludge, DAW, evaporator concentratesoBWRIon exchange resin, filters, sludge, DAW, evaporator concentratesoBWRIon exchange resin, evaporatoriaWWER, heavy activated charcoal decommissioningIon exchange resin, evaporatoriaPWRIon exchange resin, evaporatoriaPWRIon exchange resin, evaporator	Operational and decommissioning wastes from power plants	No current disposal site	Based on direct measurement	Details are shown in Annex A-9
of Korea PWR, PHWR Ion exchange resin, filters, sludge, DAW, evaporate concentrates RBMK Ion exchange resin, sediments, perlite, DAW, filters, evaporator concentrates BWR Ion exchange resin, filters, sludge, DAW, evaporator concentrates, activated charcoal WWER, heavy is activated charcoal heavy i	Evaporator concentrates, ion exchreshin, filter sludge, filter cartridge and DAW	nge Near surface LLW disposal facility is operating Planning underground disposal facility for Intermediate level waste ILW	Based on direc measurement	Details are shown in Annex A–10
RBMKIon exchange resin, sediments, perlite, DAW, filters, evaporator concentratesBWRDAW, filters, evaporator concentratesBWRIon exchange resin, filters, sludge, DAW, evaporator concentrates, activated charcoalWWER, heavyIon exchange resin, evaporator activated charcoalWWER, heavyIon exchange resin, evaporator on-combustible compactable DAW, non-combustible non-compactable DAWPWRIon exchange resin, evaporatorPWRIon exchange resin, evaporator	Ion exchange resin, filters, sludge, DAW, evaporate concentrates	Developing underground repository (tunnel and vault type)	Based on direct measurement	Details are shown in Annex A-11
BWRIon exchange resin, filters, sludge, DAW, evaporator concentrates, activated charcoalWWER, heavyDAW, evaporator concentrates, activated charcoalWWER, heavyIon exchange resin, evaporator concentrates, ash, non-combustible gas cooled reactor (HWGCR)PWRIon exchange resin, evaporator non-compactable DAW, non-combustible non-compactable DAW	Ion exchange resin, sediments, per DAW, filters, evaporator concentr	ite, Planning near surface tes repository for LILW Planning long term storage and studying disposal in geological repository for long lived LILW and high level waste (HLW)	Based on direct measurement	Details are shown in Annex A-12
WWER, heavyIon exchange resin, evaporatorwater moderatedconcentrates, ash, non-combustiblegas cooled reactorcompactable DAW, non-combustible(HWGCR)non-compactable DAWdecommissioningnon-compactable DAWPWRIon exchange resin, evaporator	Ion exchange resin, filters, sludge, DAW, evaporator concentrates, activated charcoal	Under study	Based on direct measurement	Details are shown in Annex A-13
PWR Ion exchange resin, evaporator		Operating, near surface repository for LILW le	Based on direct measurement	Details are shown in Annex A–14
trom DAW	Ion exchange resin, concentrates, ashes	Planning long term storage/disposal	Based on direct measurement	Details are shown in Annex A-15

TABLE 2. SUMMARY OF SCALING FACTOR PROGRAMMES IN VARIOUS MEMBER STATES (cont.)

Country	Reactors	Relevant waste streams	Disposal programme	Scaling factor methods	Comment
Spain	BWR, PWR, untreated natural uranium graphite GCR dismantled	Ion exchange resin, filters, sludge, compactable DAW, non-compactable DAW, evaporator concentrates, decommissioned waste	Developing a surface repository for VLLW Operating a near surface repository for LILW Planning central interim storage for HLW	Based on direct measurement	Details are shown in Annex A-16
Sweden	PWR, BWR	Ion exchange resin, filters, sludge, DAW, evaporate concentrates	Operating near surface repository for LILW Planning geological repository for long lived LILW and HLW	Based on direct measurement	Details are shown in Annex A–17
Ukraine	WWER, RBMK	Ion exchange resin, filters, sludge, evaporator concentrates, perlite, compactable DAW, non-compactable DAW, combustible DAW, non-treatable DAW	Planning geological repository for long lived isotopes	Under study	Details are shown in Annex A–18
UK	GCR, PWR	DAW	Operating near surface LLW repository Planning ILW repository	Based on direct measurement	Information provided for the dry solid LLW stream. Details are shown in Annex A–19
USA	PWR, BWR operating and decommissioning	Ion exchange resin, filters, sludge, evaporator concentrates, compactable DAW, non-compactable DAW, combustible DAW, non-treatable DAW	Operating disposal sites	Based on direct measurement	Method dominantly used by nuclear power plant, no systematic application in other types of facility. Details are shown in Annex A-20

TABLE 2. SUMMARY OF SCALING FACTOR PROGRAMMES IN VARIOUS MEMBER STATES (cont.)

Country	Sampling practice	SF determination method	Data maintenance
Belgium	Sampling for validation	Calculation by modelling	Every five years in order to test validity of the model
Brazil	Sampling by nuclear power plant and waste stream	Geometric mean Linear regression of logarithms (correlation function) when no useful SF is obtained by geometric mean (see Annex A–2) Arithmetical mean of data from direct radiochemical measurements when neither useful SF nor correlation function is obtained (see Annex A–2)	By operating cycle
Canada	Sample by waste stream	Geometric mean	Periodic update planned, period not yet defined
China	Sampling practice not yet defined	Final method not selected	Final method not selected
France	Sampling by waste stream and by nuclear power plant	Linear regression when $R \ge 0.7$ Geometric mean when $0.5 \le R < 0.7$ Arithmetical mean when $R < 0.5$ or lack of data (<5)	Periodic sampling to confirm values have not changed and to increase number of data points
Germany	Sampling determined at plant level – no prescriptive direction	Linear regression of logarithms	Annual sample update and review
Hungary	Sample by stream	Linear regression of logarithms	By operating cycle
India	Sampling practice not yet defined	Final method not selected	Final method not selected
Italy	Sample as part of preliminary radiological characterization	Geometric mean	To be defined
Japan	Sampling by waste stream for homogenized waste. Specific sampling (sampling by waste stream and type of contamination) for DAW	Arithmetical mean (applicability of geometric mean had been judged)	Annual sample in each station for validation
Republic of Korea	By waste stream and by plant	Geometric mean	SF revised every two years, or in the event of an incident, or if there is a significant change in plant conditions

TABLE 3. SUMMARY OF SAMPLING PRACTICES AND SCALING FACTOR DETERMINATION METHODS

Country	Sampling practice	SF determination method	Data maintenance
Lithuania	By waste stream Sampling DAW as a function of the origin and type of contamination	Linear regression of logarithms	SF revised three to four years, or when significant change of ⁶⁰ Co/ ¹³⁷ Cs
Mexico	By waste stream	Weighted linear regression of logarithms	Difficult task due to the lack of radiochemical laboratories
Slovakia	By waste stream and by plant	Linear regression of logarithms	SF revised when significant trend change Sampling when tanks are full, sampling for every drum with ash
Slovenia	By waste stream	Arithmetical mean	SF revised every four years, sampling ashes every campaign, low rate of sampling for dried waste due to low production
Spain	By waste stream and by plant	Geometric mean	SF revised every two years, sampling every campaign
Sweden	By waste stream and by plant	Geometric mean	SF revised every ten years with periodic sampling
Ukraine	By waste stream	Geometric mean	Final method not selected
UK	[Method refers to DAW] – for each station by plant areas	Weighted arithmetic mean	Review based on plant conditions and periodic re-appraisal (e.g. sampling, analysis recalculation every five to six years)
USA	Sample by plant and by stream	SFs are calculated as a logarithmic average and the dispersion is the antilog of the variance	SFs are required to be reviewed annually or at the end of each fuel cycle (effectively 12–18 months) or if there is a significant change in plant conditions; samples are usually collected in conjunction with each review

	Key nuclide selection	Data grouping (SF classification)
Brazil	Co-60 and ¹³⁷ Cs are used as major key nuclides, based on radiological relevance and international experience	Nuclear power plant and waste stream specific
Canada	Co-60 and ¹³⁷ Cs isotopes are used as major key nuclides, based on radiological relevance and international experience Actinides are scaled to calculated ^{239/240} Pu Tritium is scaled to calculated ¹⁴ C in DAW streams	By waste stream, not plant specific DAW streams updated by incinerator ash data
China	Selection not defined	Final method not selected
France	Co-60 for CP nuclides Cs-137 for FP homogeneous waste Co-60 or ¹³⁷ Cs for FP heterogeneous waste Co-60 for actinides	Data centrally collected and grouped on basis of plant and stream similarities as well as on observed correlation SFs for homogeneous wastes are based on ion exchange resins, and SFs heterogeneous wastes are based on water filters
Germany	Strong gamma emitter Abundance Long half-life Production link Compatible transport properties	Data centrally collected and grouped on basis of plant and stream similarities as well as on observed correlation
Hungary	Co-60 and 137 Cs isotopes are used as key nuclides, based on radiological relevance and international experience	DAW determined with evaporator concentrates, otherwise plant and stream specific
India	Selection not defined	Final method not selected
Italy	Gamma and alpha emitters; long lived beta emitters to be defined	Classify by type of reactor
Japan	C-60 for CP nuclides Cs-137 for FP and actinides (Applicability of ⁶⁰ Co for actinides had been confirmed)	Classify by type of reactor (difference of reactor component material) for CP nuclides Classify by fuel condition for FP nuclides and actinides
Republic of Korea	C-60 for CP nuclides Cs-137 for FP nuclides Co-60 for Actinides	SF from each stream and each nuclear power plant

TABLE 4. SUMMARY OF KEY NUCLIDE AND DATA GROUPING PRACTICES

Country	Key nuclide selection	Data grouping (SF classification)
Lithuania	Co-60 or ¹³⁷ Cs for CP nuclides Co-60 or ¹³⁷ Cs for FP nuclides Co-60 or ¹³⁷ Cs for actinides Cs-137 is used as a key nuclide for the waste streams in which activity of ⁶⁰ Co is much lower than activity of ¹³⁷ Cs	Waste stream specific
Mexico	Planning ⁶⁰ Co for CP, FP and actinides	Plant and waste stream specific
Slovakia	Co-60 for CP nuclides Cs-137 for FP nuclides Co-60 for actinides	SF from each stream and each nuclear power plant
Slovenia	Mainly ¹³⁷ Cs for alpha emitters; other DTM nuclides not considered	Waste stream specific
Spain	Co-60 or ¹³⁷ Cs for CP nuclides Co-60 or ¹³⁷ Cs for FP nuclides Co-60 or ¹³⁷ Cs for actinides	SF from each stream and each nuclear power plant
Sweden	Co-60 for CP nuclides Cs-137 for FP nuclides Co-60 for actinides	Type of waste and nuclear power plant
Ukraine	Co-60 or ¹³⁷ Cs for CP nuclides Co-60 or ¹³⁷ Cs for FP nuclides Co-60 or ¹³⁷ Cs for actinides	Final method not selected
UK	Co-60 for CP nuclides Cs-137/ ⁶⁰ Co for FP and actinides	[Method refers to DAW] – for each station the waste stream is classified by plant area
USA	Based on compatible transport properties and/or reasonable statistical correlation	Plant and stream specific; grouping of streams may be performed for individual SFs

TABLE 4. SUMMARY OF KEY NUCLIDE AND DATA GROUPING PRACTICES (cont.)

While direct gamma assay can provide a more accurate measurement of the key nuclide, it also requires more sophisticated equipment than the dose rate to activity conversion method. Conversely, while the dose rate to activity conversion method requires simple equipment, it still requires the development of multiple conversion factors for various waste package types and geometries and/or waste types, and the use of well defined measurement geometries in order to ensure reproducibility of results.

The responsibility for developing and implementing SFs varies by Member State. In some Member States, such as Japan, Mexico and the USA, the waste generator (e.g. the nuclear power plant operator) is completely responsible for sampling, SF development, measurement of key nuclides in waste packages and declaring the calculated DTM nuclides. In other Member States, such as Spain or Sweden, the waste generator may only be responsible for supplying samples to the repository operator or some other independent body, who is then responsible for development and application of the SFs; or the waste generator declares the key nuclide value and DTM nuclides are calculated and recorded by the repository operator.

It should also be noted that obtaining samples for analytical purposes usually carries a dose consequence for the operating personnel. In accordance with the as low as reasonably achievable (ALARA) principle, unnecessary doses should be avoided. Therefore, careful planning of which waste streams to sample, how many samples to collect, what size of sample to collect, how the samples are handled, etc., are important factors in designing and optimizing the waste sampling programme. Basic guidance on the number of samples to collect can be found in references such as Ref. [19]. In some cases, another factor that affects the number of samples collected and analysed is the lack of laboratories to perform DTM nuclide measurements.

The tables, along with the details presented in the Annex to this report, demonstrate that good practice in the application of empirical SFs includes the following elements:

- (a) Systematic sampling of waste streams (including determination of which waste streams to sample, how many samples are required, how the samples are collected, etc.);
- (b) Radiochemical analysis for the radionuclides of concern;
- (c) Analysis of data for correlation;
- (d) Assessment of feasibility of combining or grouping data from different waste streams, different reactor types, etc.;
- (e) Selection of ETM key nuclides based on evidence of correlation, modelling or on established practice;
- (f) Selection of an appropriate SF calculation method (e.g. geometric mean, linear regression of logarithms);
- (g) Periodic re-sampling to confirm or re-validate the SFs, especially if plant conditions have changed (e.g. significant changes to major reactor component materials and/or indication of significant fuel failures and/or waste treatment methods).

In addition to the good practice features outlined above, the specific application in any given Member State will be influenced by a number of local factors, such as:

- (i) National policies and regulations (e.g. which may specify the radionuclides to report, the degree of accuracy required, the frequency of sampling, who is responsible for sampling and reporting, the waste classification system).
- (ii) The size and type of nuclear power programme (e.g. a Member State with a single nuclear power plant may not be concerned with combining data from different plants within the country, but may be able to combine data with similar plants in other Member States (see Section 4.2.4). A Member State with many similar nuclear power plants can often take advantage of combining data from the separate plants if analysis shows that there are no significant differences between the plants.).
- (iii) The safety case and/or licence conditions for the disposal facility (e.g. which may determine the important radionuclides, the degree of accuracy required).
- (iv) The availability of analytical resources to provide the specialized radiochemical analyses required to develop the SFs.

In the case of shutdown reactors and those undergoing decommissioning, the ratio of the key nuclide to the DTM nuclide will change over time. The key nuclides (typically ⁶⁰Co or ¹³⁷Cs) will decay faster than the radionuclide of interest (which have typically long half-lives). Decay correction thus needs to be taken into account when developing and using the SFs. This must also be considered for historical wastes (e.g. wastes that have been stored for a long period before characterization).

In the case of those nuclear power plants that have undergone major fuel failure incidents or accidents, differing amounts of various radionuclides may have been transported and deposited to various areas inside and outside of the plant based on the chemical transport properties and the volatility of the elements involved. In this situation, it may be necessary to develop area specific SFs rather than waste stream specific ones.

The basic SF methodology can, in principle, be applied to other situations, such as wastes from research institutes, fuel cycle facilities and other industrial facilities if it can be demonstrated that a suitable relationship exists between the chosen key nuclides and the desired DTM nuclides. For all cases, the specific key nuclides and the numeric values of the SFs need to be developed on a case by case basis, similar to the application for nuclear power plant operational waste.

4. COMMONALITY AND CONSENSUS IN SCALING FACTOR PROGRAMMES

SFs have offered a convenient and economical mechanism for deriving a full radiological profile for a waste stream. As this approach has been taken in many countries over a period of more than 20 years, there has been a steady increase in experience and knowledge in the application of SFs. Much of this experience has resulted in a general consensus among professionals working with SF applications and is reflected in the ISO standard on the SF method. Areas of agreement are discussed below.

The following experience is drawn from power reactors; much of what is discussed can be applied to other utilization facilities, including fuel reprocessing and research reactors, recognizing that the key nuclides, DTM nuclides and statistical relationships may be different in these different applications.

4.1. EVALUATION OF INFLUENCING FACTORS

4.1.1. Plant materials

In most applications of the SF method, ⁶⁰Co is used as the key nuclide for CP nuclides based on the similarities of the production mechanism and physicochemical properties. However, many factors, such as materials used and operating conditions (pH, temperature, etc.), can affect the SF seen on the waste arisings. The example below illustrates how the choice of materials between PWRs and BWRs can affect the SF of ⁶³Ni in resins.

CP compositions change depending on the reactor type. For example, the major sources of CP nuclides from a PWR are nickel alloy and stainless steel, with little use of carbon steel. On the other hand, those of a BWR are carbon steel and stainless steel.

The nickel and niobium contents relative to cobalt in a PWR are higher than those in a BWR. Thus, when the radioactivity of CP nuclides is evaluated, waste can be classified by reactor type because of their varying reactor component materials. This difference is illustrated in Fig. 4 [19].

Figure 5 shows the difference between Spanish PWR and BWR resins for ⁶³Ni/⁶⁰Co SF.



FIG. 4. Comparison of ⁶³Ni/⁶⁰Co ratios for a PWR and a BWR. (a) Scatter diagram of ⁶³Ni/⁶⁰Co; (b) distribution diagram of ⁶³Ni/⁶⁰Co.



FIG. 5. Example of difference of SF between different plant designs.

4.1.2. Fuel

4.1.2.1. Fuel failure

4.1.2.1.1. Experience in Japan

In many applications of the SF method, ¹³⁷Cs is used as the key nuclide for FP and alpha emitting nuclides. In principle, the SFs of FP and alpha emitting nuclides in similar facilities would be expected to be very similar. However, design or operational differences in the two facilities can result in different SFs. For example, changes



FIG. 6. Effect of fuel failure on SF (BWR, total alpha/¹³⁷Cs; homogeneous solidified waste). (a) Scatter diagram; (b) histogram of ratio of DTM nuclide/key nuclide.

in the release mechanisms of FP and alpha emitting nuclides from their original production site, either from tramp uranium or failed fuel in a reactor core, affect the SF.

An example is illustrated in Fig. 6 of how the number and severity of fuel failures affect the SF of ⁹⁰Sr and total alpha in light water reactor homogeneous solidified waste. The assessment of the influence of fuel failures on these SFs found that it would be appropriate for the SF to be classified into three groups based on the fuel failure history [20].

The difference in SF is attributed to the relative release of ¹³⁷Cs and other FP/alpha emitting nuclides dependent on the severity of the fuel failure (i.e. whether the release is from the fuel/clad gap or from the fuel matrix).

4.1.2.1.2. Experience in the USA

In the USA, SFs are required to be reviewed (and updated if necessary) with each reactor fuel cycle or if there is a significant change in plant conditions. The Electric Power Research Institute (EPRI) sponsored a study to identify and evaluate changes to SFs occurring as a result of certain changes in operating conditions [21, 22]. Some of the changing conditions investigated included:

- (a) Changes applicable to all types of plant:
 - (i) Fuel defects, changes in fuel design, extended shutdowns.
- (b) Changes specific to BWRs:
 - (i) Zinc injection, hydrogen water chemistry, iron injection.
- (c) Changes specific to PWRs:
 - (i) Lithium injection, zinc injection.

The study was conducted by identifying plants that had experienced such changes and obtaining their radiochemical sample records for a period of at least three years before and three years after the event. The SF ratios for these periods were evaluated on the basis of geometric means, and then a comparison was conducted to determine if there was a statistically significant difference in the data from before and after the event. The comparison was performed using a pooled variance analysis which would determine at the 95% level if there was a difference in the data. Comparisons were evaluated both graphically and numerically. Figure 7 shows the ⁹⁰Sr/⁶⁰Co SF for the reactor water cleanup (RWCU) system preceding and after a significant fuel failure in a BWR power plant. As can be seen in this case, the change in the condition is readily apparent and easily confirmed with numerical comparisons.





FIG. 7. SF behaviour following a fuel failure event (BWR RWCU resins).

TABLE 5. SCALING FACTOR BEHAVIOUR FOLLOWING A FUEL FAILURE EVENT
(BOILING WATER REACTOR MAJOR WASTE STREAM)

Ratio/stream	RWCU	RW	Condensate	DAW
Sr-90/ ⁶⁰ Co	10	1	3	5
Sr-90/137Cs	10	1	4	10
Pu-239/60Co	2	1	2	1
Pu-239/137Cs	2	1	4	3

The impacts of this event on other streams and other SFs within the same facility are not as apparent and require statistical determinations to verify.

Table 5 provides a summary of significant differences in SFs observed from this event. While the difference was very pronounced in the RWCU, it is much less apparent in some of the other systems. In addition, the ²³⁹Pu ratios were not nearly as strongly affected as the ⁹⁰Sr ratios.

Some of the important findings of the EPRI study are:

- (a) Changes in plant conditions can change SFs.
- (b) Often the normal variability in SF values hides the impact of the change to the SF.
- (c) SF shifts can be less than the dispersion particularly for some of the more problematic SFs.
- (d) SF response to changes in conditions is often not immediate usually there is some delay before the effects are observed, as they become more removed from the primary systems.

4.1.2.2. Fuel type

In order to demonstrate the capabilities of the SF method beyond activity determination, an investigation of the influence of mixed oxide (MOX) fuel on the alpha activity concentrations in waste was performed. MOX fuel elements have been used in routine operation in German nuclear power plants since 1986 [23]. Experience is available mainly from PWRs.



FIG. 8. Effect of MOX fuel [23] (w/o: without).

As the MOX fuel elements contain transuranic elements from the very beginning, an increase might be expected in the radionuclide inventory of higher transuranic elements (e.g. americium, curium) and the heavier plutonium isotopes (e.g. ²⁴²Pu, ²⁴⁴Pu) in waste from nuclear power plants that use MOX fuel. This increase should have an influence on the correlation of the mentioned elements to the key nuclides.

The influence of MOX fuel might be superimposed over other effects such as fuel defects or outer contamination of fuel elements. Unfortunately, little primary information about these parameters is available. Figure 8 shows the effect of MOX fuel elements. Since the difference between operation with and without MOX fuel is in the range of the inherent scatter of data, changes in activity concentration of transuranic elements due to MOX fuel are not readily detectable.

While the activity concentration of 60 Co decreases to nearly the same extent as the activity concentration of the transuranic elements, other factors also influence the ratios (e.g. different period of origin MOX fuel, with the result that the correlations of transuranic elements to 60 Co remain essentially unchanged.

4.1.3. Other factors

The activity composition in various waste streams, such as DAW, resins, filter cartridges, etc., in a facility is usually expected to be different, thus requiring different SFs to characterize them. However, there are occasions when the variation in the SFs is small enough to be able to derive a representative set of common or grouped SFs. This may be able to be justified on the basis of theoretical considerations, or it may result from empirical measurements.

When considering the same waste stream in different facilities of the same type, for example DAW in PWRs or BWRs, the SFs may be expected to show some similarities. This, of course, would depend on factors such as material composition and fuel failures, discussed in the earlier sections, as well as on other parameters such as the operational history of the plant, working practices, etc.

Depending on the use of the SFs and the level of variation that can be considered acceptable, it may be possible to derive SFs that can be applied to a number of waste streams or are common to a group of facilities. This would be particularly applicable where the waste from different areas or facilities is combined, treated, stored or disposed of together, and therefore any individual differences average out.

The following subsections present a number of examples where similarities or differences in SFs occur.
4.1.3.1. Common scaling factor for multiple waste streams

An example in which an SF is applicable to more than one waste stream is illustrated below. It considers the SF of ⁶³Ni to ⁶⁰Co in DAW in BWR plants.

In BWR plants, the reactor materials have a comparatively large influence on the activity concentrations and ratios of the CP nuclides. Thus the ⁶³Ni and ⁶⁰Co source levels and relative ratio depend on the reactor materials. In studying the SF of ⁶³Ni to ⁶⁰Co, the BWR plants were allocated into two groups: the 'plants made of conventional material' group and the 'plants made of low cobalt material' group.

The DAW is categorized into three waste streams: the reactor stream, the turbine stream and the radioactive waste stream. Each stream has three types of DAW: 'direct contamination by liquid', 'direct contamination by gas' and 'indirect contamination'. At first glance it would appear that for each group of BWRs, nine SFs would be needed to characterize the waste.

Figure 9 [24] summarizes the assessment carried out to compare the variation in the ⁶³Ni to ⁶⁰Co SF between the different waste streams and the types of contamination in each waste stream. It shows that there is no appreciable difference in the SFs between either the waste streams or the types of contamination.

Based on this assessment it was concluded that a single SF for ⁶³Ni can be determined and used for all DAW for each plant material group.

4.1.3.2. Solubility of radionuclides

In many practical applications of the SF approach, the FPs are most often correlated to ¹³⁷Cs. The SF of a radionuclide in relation to ¹³⁷Cs may vary depending on the process producing the waste, due to differences in the chemical properties of the radionuclides. In particular, because ¹³⁷Cs is relatively soluble, ¹³⁷Cs does not collect on the surface of heterogeneous waste (solid phase wastes such as DAW) in the same proportion as non-soluble FP nuclides. Therefore, the SF of an FP nuclide for heterogeneous waste would be different from its SF in a homogeneous waste (liquid wastes).

This is illustrated in Fig. 10 [25] by considering the SFs of ⁹⁰Sr to ¹³⁷Cs in homogeneous and heterogeneous waste, and the overall variability between homogeneous and heterogeneous waste in French, German and Japanese PWRs. The SFs for the individual heterogeneous (liquid) waste streams (reactor coolant, spent resin, concentrates) are clearly lower than the SFs of the heterogeneous (solid) waste streams (filters and DAW). This is also reflected in the experience of different groups of plants in different countries.

As a result of this example, it is appropriate to consider the influence of solubility of radionuclides on the SFs for homogeneous and heterogeneous wastes.

4.1.3.3. Volatility of radionuclides

An example where more than one SF is required to characterize DAW associated with a single liquid process stream is illustrated below. It considers the SF of ¹⁴C to ⁶⁰Co in DAW arising from contamination from primary coolant liquid in BWR plants.

In the BWR primary coolant, ¹⁴C exists mainly in the form of CO_2 . The CO_2 and ⁶⁰Co entrained in the steam in the reactor vessel pass into the turbine system. When the steam goes through the turbines and condenser, the concentration ratio of CO_2 and ⁶⁰Co in the turbine system fluid is different from that in the reactor system fluid. Therefore, the concentration ratio of ¹⁴C and ⁶⁰Co differs between the reactor system fluid and the turbine system fluid. In the radioactive waste system, the waste water from the reactor system and the waste water from the turbine system are combined, and the ¹⁴C/⁶⁰Co concentration ratio of this mixture has a wide distribution as a combination of the concentration ratios of both system fluids.

DAW whose contamination sources are the fluid from these systems reflects the variation in the ¹⁴C and ⁶⁰Co ratio in the fluid. Figure 11 [24] shows the variation in the ¹⁴C to ⁶⁰Co ratio (SF), indicating that different SFs are required to characterize the waste from these three different parts of the system.



FIG. 9. Influence of waste streams on the SF for ⁶³Ni in BWRs.



FIG. 10. Example of differences in the ratio of FP ratios in different waste streams. (a) International comparison (ratio of DTM nuclide/key nuclide); (b) international comparison homogeneous and heterogeneous waste (correlation); (c) detailed comparison among waste streams (ratio of DTM nuclide/key nuclide).



FIG. 11. Evaluation of ${}^{14}C/{}^{60}Co$ ratio in BWR plant areas. (a) Migration of ${}^{14}C$ and ${}^{60}Co$ within the plant system; (b) comparison of the distribution of the ${}^{14}C/{}^{60}Co$ ratios of system fluids.

4.2. EVALUATION OF APPLICATION METHODS

4.2.1. Selection of the key nuclides

In theory, key nuclide selection is based on considerations relating to the production mechanism, transport parameters and exposure of the stream to various processes. In practice, the main criterion is the ability to show that a correlation exists between the key nuclide and the DTM nuclide. The ultimate determinant in establishing a viable key nuclide is that it is readily measurable using gamma spectrometry available at the point of generation or packaging for disposal.

Experience has shown both ⁶⁰Co and ¹³⁷Cs provide consistent and reasonable correlations with a range of FPs and transuranic nuclides. Cs-137 is sometimes not viable as a key nuclide because its concentration may not be measurable in waste stream samples. Similarly, ¹⁴⁴Ce, which is often correlated to transuranic nuclides, is generally not a viable key for these radionuclides because it is not readily measured in gamma spectra with a ⁶⁰Co background and its half-life is too short to be used for waste held for extended storage periods. However, ¹⁴⁴Ce may be used as an alternative key nuclide for transuranic nuclides in fresh radioactive waste if it is measured based on the high energy gamma ray of 2.18 MeV. Other radionuclides may provide reasonable surrogate representation for some DTM nuclides based on particular stream characteristics.

The examples below (from various national SF programmes) form a part of a growing knowledge base in the application of SFs.

In general, alpha emitting nuclides (consisting of actinide elements such as plutonium, americium and curium) have an affinity to adhere to surfaces and are most usually insoluble. These radionuclides exhibit similar transport properties to ⁶⁰Co, which is also insoluble in reactor environments. This is also supported by the fact that the ratios of total alpha to ⁶⁰Co show little variability between liquid waste and DAW. This is because in both cases the cobalt and transuranics are transported with particulate matter carried by the flow in the reactor coolant and as surface contaminants on the DAW.

The data in Fig. 12 [26] were obtained by analysis of DAW generated at PWR plants. In both cases, a correlation can be seen between total alpha emitting radionuclides and the key nuclide. It can be concluded that not only ¹³⁷Cs but also ⁶⁰Co can be selected as a key nuclide for total alpha emitting nuclides.

For less soluble FPs (including ⁹⁰Sr and ⁹⁹Tc), ⁶⁰Co often appears as a more stable and reliable key nuclide than ¹³⁷Cs. This could be attributed to two conditions:

- There is little fuel failure, so the production of transuranics and FPs is limited to fuel present as contami-(a) nation on reactor surfaces. The release processes more closely mirror releases of CPs.
- (b) All of the radionuclides are electro-positive and insoluble, therefore tending to transport in the corrosion layers. (This is also true in the relationship between cobalt and transuranic elements.)

As an example from Spain, Fig. 13 shows that ⁶⁰Co is a better key nuclide for ⁹⁰Sr than ¹³⁷Cs, having a lower data dispersion and therefore more reliability in the prediction of ⁹⁰Sr activity in a waste package.

(b)



FIG. 12. Effects of key nuclide selection on CC for total alpha. (a) Key nuclide is 60 Co, CC = 0.72; (b) key nuclide is 137 Cs, CC = 0.78.



FIG. 13. Effects of key nuclide selection for ⁹⁰Sr correlation.



FIG. 14. Spanish example for mean activity concentration of ⁹⁹Tc.

Sometimes it is not possible to obtain any meaningful SF when the DTM nuclide activity concentration does not show any change with the activity concentration of the key nuclide, such as the example shown for ⁹⁹Tc in Fig. 14. In this case, a mean activity concentration of the DTM nuclide is often taken as representative for that waste.

4.2.2. Calculation method

There are two main methods for calculating average (mean) SF values:

- (a) An arithmetical mean;
- (b) A geometric (or log) mean.

The arithmetic mean is calculated by summing up the values and dividing by the number of samples. When there is a large variation in the individual values being averaged, the arithmetic mean will produce a value that is weighted towards the higher values and may be inappropriately skewed.

The geometric or log-mean is calculated by taking the anti-log of the arithmetic mean of the log of the individual values. This method is appropriate for data that can range over several orders of magnitude, since it tends to reduce the effect of extreme values.

The radioactive concentrations of DTM nuclides and key nuclides show a log-normal distribution having a correlation between both radionuclides (i.e. bivariate log-normal distribution).

Using the normalized data for which the CC is 0.7, the distribution of actual radioactivity concentrations was compared with that of radioactivity concentrations determined by each SF calculation method. The results of the comparison are shown in Fig. 15. It is seen from Fig. 15 [26] that the distribution of radioactivity concentrations and the amount of radioactivity are evaluated closer to the true value by the geometrical mean than by the arithmetical mean.

Average SFs calculated from a mean value assume a linear relationship between the key nuclide and the DTM nuclide. More complex mathematical relationships between key nuclides and DTM nuclides can be modelled by methods such as 'regression of logarithms', which can be used to derive non-linear relationships. This practice is used, for example, in Germany, Hungary, Lithuania, Mexico and Slovakia.

4.2.3. Trend evaluation and updates

4.2.3.1. Experience in the United States of America

In many cases where SFs are evaluated, data are collected over extended time periods. These data often show exceptional stability as they are collected. Figure 16 shows the ${}^{63}\text{Ni}/{}^{60}\text{Co}$ ratio for BWR RWCU resins tracked in a single unit plant over a period of more than 20 years. Within the expected variability related to measurement accuracy, the SF has been essentially constant over that entire period. This is even as the plant aimed to reduce overall cobalt and introduced various chemistry programmes to minimize scale buildup. If the data support it, developing averages over long time frames is reasonable and may provide a better statistical basis for the SF than using the most recent or a short term average. Trends can be evaluated by standard least squares methods assuming a log-linear scale. The trend rate is defined by the slope of the regression. A slope = 0 indicates that the value is constant over the time period studied.



FIG. 15. Comparison of calculation methods. (a) Scatter diagram, CC = 0.70; (b) SF calculation by arithmetical mean, radioactivity ratio = 8.2; (c) SF calculation by geometrical mean, radioactivity ratio = 1.4. Note: radioactivity ratio = estimated activity/actual activity.

[Ni-63]/[Co-60] - Norm



FIG. 16. Ni-63/60 Co trend.

4.2.3.2. Experience in Japan

In Japan, commercial nuclear power plants have been in operation since the 1970s, and radioactive wastes generated from reactor operation, such as concentrates, have been stored within the nuclear power plants in the form of waste packages which are prepared by solidifying the wastes in drums using cement or other binders. To evaluate the SFs used to determine the radioactive concentrations of those wastes, waste samples have been taken from the waste packages which have been generated and stored within the stations since the start of commercial operation. The changes in the SFs over a period of about 30 years have been evaluated by analysing the samples.

With respect to the SFs for DTM nuclides originating from the activation of reactor component materials (such as ⁶³Ni and ⁹⁴Nb), there is only a small possibility of the SFs changing, as long as the chemical composition of the reactor material does not change greatly due to a large scale modification of reactor components. Also, concerning the SFs for ⁹⁰Sr and alpha emitting nuclides originating from the activation of nuclear fuels, there is only a small possibility of the SFs changing, as long as there is no change in the occurrence rate and degree of fuel failure.

In Japan, therefore, nuclear plants were classified into groups at the initial stage of SF development based on the effects of reactor materials and fuel failure, and separate SF values have been established for each group of plants.

Figure 17 shows an example of the long term trend in the composition ratio of ⁶³Ni and ⁶⁰Co contained in DAW generated at BWR plants. It can be seen from this figure that the composition ratio has been, for all practical purposes, kept constant over a period of about 30 years.

4.2.3.3. Experience in Spain

Once an SF has been established, future radiochemical data are suitable for both increasing the reliability of the database and for verification of the prior established value. At least three new values are collected to perform the trend test. One group in the test is the additional data, and the other is the original set from which the SF was calculated.



FIG. 17. Japanese trend in the composition ratio of ⁶³Ni and ⁶⁰Co.

If the additional data show values similar to the previous data, the prior SF can remain with the current value, otherwise old measurements are removed from the database and the additional ones included. When the SF remains unchanged after a revision, future collected values will continue to increase the augmented data set. Figure 18 illustrates an example of SF data trend evaluation.

4.2.4. International integration

SFs, although empirical in nature, are an essential prerequisite for the determination of activity concentrations in waste from nuclear power plants. Up to now, each country has independently analysed and assessed SFs, but has in general obtained similar results for similar types of nuclear power plants and wastes. The following sections investigate the consistency of the SF method by comparing evaluation results from different countries for comparable conditions, and check the possibility of enhancing the applicability of the SF method by using a database integrating data from different countries. If consistency has been demonstrated in the first step, the second may improve the quality of the activity determination, increase the number of radionuclides for which the method is applicable, or increase the number of available data points available for the very DTM radionuclides.

4.2.4.1. Activated corrosion product nuclides

One source for the generation of radionuclides in nuclear power plants is the activation of reactor materials and their CPs in the reactor core. Ni-63 is among the activated CP nuclides whose concentrations are limited in most LLW disposal facilities. To determine the activity concentrations of these DTM nuclides, each country usually selects the CP nuclide ⁶⁰Co as the key nuclide and determines the concentration of ⁶⁰Co by direct measurements on waste packages. The activity concentrations of the DTM nuclides are then calculated according to the functional relationship established between the activity concentration of ⁶⁰Co and the DTM nuclides such as ⁶³Ni.

The main reactor materials differ depending on the reactor type: carbon steel or stainless steel are dominant in BWRs and Inconel or Incoloy in PWRs. As the element composition of these reactor materials differs, it was considered reasonable for CP nuclides to be differentiated according to the reactor type.

If international integration of data is a valid concept, the differences in SFs and the calculated activity concentrations for DTM nuclides should be small. Figure 18 [19] shows a comparison of radionuclide data on ⁶³Ni and ⁶⁰Co between different reactor types and between France, Germany, Japan and the USA. As expected, the radionuclide data show an appreciable difference between different reactor types, but show no remarkable difference between different countries for the same reactor type. This demonstrates the consistency of the method applied. Accordingly, it is considered that CP nuclides can be assessed using cumulative data from different countries for each reactor type.





FIG. 19. CP nuclide correlation with regard to reactor type and country (⁶³Ni/⁶⁰Co). (a) Reactor type; (b) country.

4.2.4.2. Fission product and alpha emitting nuclides

Another source for radionuclides in nuclear power plant waste is the FPs from the fission of uranium contamination on the fuel rods and from leaking fuel pins. Sr-90 belongs to the FP nuclides whose activities or activity concentrations are limited for many disposal facilities. APs of uranium, so called transuranics (mostly alpha emitters), are often treated together with the FP, since they originate from the same source.

In assessing these DTM nuclides, most countries select ¹³⁷Cs as a key nuclide and determine the activity concentrations of these DTM nuclides in the same manner as in the case of CP nuclides. Figure 20 shows a comparison of radionuclide data for alpha emitting nuclides, ⁹⁰Sr and ¹³⁷Cs between different reactor types and between France, Germany, Japan and the USA.

For FP nuclides, the effects of the difference in the reactor type on the SFs were in a first approach assumed to be small because the difference in the fission rates is small. However, observations from measurements partially contradict this expectation as appreciable differences between BWR and PWR data for the correlation of ⁹⁰Sr to ¹³⁷Cs exist. This effect is less pronounced for the alpha emitters.

This behaviour is attributed to the fact that different physicochemical conditions in the reactor primary system induce changes in the ratio between ¹³⁷Cs and ⁹⁰Sr. Since ¹³⁷Cs remains soluble in any chemical environment, while ⁹⁰Sr is observed with varying distributions between the solid and the soluble phase for PWRs and BWRs, respectively, a different ratio of these FPs appears in the concentrates. This effect is less pronounced in agreement with the SFs determined for the alpha emitters, as they remain insoluble in both reactor type environments.

In conclusion, the SF behaviour of the FP nuclides cannot be derived from their generation ratio alone, but has to take into account their transport behaviour as well.

While some degree of consistency has been observed in data from various countries, before combining data from different countries additional effort is required in checking comparability of the physicochemical conditions responsible for the radionuclide transport behaviour in the primary cooling system and the adjacent auxiliary systems.



FIG. 20. Alpha emitting and FP nuclide correlation with regard to reactor type and country. (a) Reactor type (total $alpha/^{137}Cs$); (b) country ($^{90}Sr/^{137}Cs$).

4.3. IMPACTS OF WASTE TREATMENT

4.3.1. Incineration

4.3.1.1. Incineration in reducing conditions

Processing of incinerable waste at Ontario Power Generation's (OPG's) waste management facility in Ontario, Canada, results in the production of incinerator bottom ash. While sampling of bulk LLW streams such as incinerable, compactable and non-processed waste presents a significant challenge, sampling of incinerator bottom ash is considerably simpler. Development of SF data would be simplified if data based on bottom ash were applicable to the other waste streams.

Table 6 shows the measured SFs for various types of bulk waste [13, 27]. It can be observed that a number of 60 Co based SFs for incinerator bottom ash, including that for 14 C, appear to be comparable with the corresponding SFs for the other waste streams. This is a consequence of the preferred partitioning of the various non-volatile nuclides into the solid ash phase. In particular, the starved air conditions prevailing in the first stage of incineration result in partial retention of 14 C (which is normally volatile) within the bottom ash. Fortunately, the fraction retained is sufficient for analysis and the 14 C/ 60 Co ash SF is, within the normally accepted uncertainty, also representative of the other bulk waste streams.

Similar to ¹⁴C, tritium (as tritiated water) is also present at elevated levels (relative to light water reactor waste) in CANDU wastes. Quantification of tritium, in general, presents significant challenges because of its mobility. For this reason, attempts to directly correlate tritium with ⁶⁰Co have met with mixed success. In this context, the availability of data for ¹⁴C and tritium levels in the stack emissions from OPG's incinerator has provided an opportunity to correlate ¹⁴C with tritium [13, 27]. In order to develop the correlation, data for total stack emissions of ¹⁴C and tritium for a monitored period were combined with feed burn volume data for the same period, and the residual ¹⁴C inventory in the incinerator ashes, to estimate the concentrations of ¹⁴C and tritium present in the incinerator feed. The results of such an assessment are shown in Table 7. As can be observed, the ¹⁴C/³H ratio was essentially constant (as judged by the low log dispersion (LD) values) over each

monitored period and remained essentially unchanged over a span of three years. (The lower the LD, the better. A value of less than about ten is generally considered to be acceptable.)

The ratio measured above is based on a large volume of waste that was combusted soon after being generated. Owing to limited out-gassing of tritium within the short period of time after generation, the ${}^{14}C/{}^{3}H$ ratio determined was observed to be consistent, with the expected ratio based on the concentration of the two radionuclides in the heavy water reactor coolant. At present, the measured data in Table 7 are used to estimate the inventory of tritium in various bulk LLW packages, recognizing that the tritium inventory during interim storage will diminish in accordance with radioactive decay and as a result of tritium out-gassing. The inventory of tritium is estimated by combining the ${}^{14}C/{}^{3}H$ ratio with the ${}^{14}C$ inventory estimated based on the applicable value for the ${}^{14}C/{}^{60}Co$ SF.

Scaling factor	Statistics	Incinerable	Compactable	Bottom ash	Non-processible
Pu-239+240/144Ce	LM	1.7E-02	1.9E-02	4.0E-02	8.1E-03
	LD	1.4	4.8	2.3	7.1
Pu-239+240/137Cs	LM	2.0E-02	1.2E-02	1.0E-02	1.5E-02
	LD	1.5	8.8	1.4	7.1
Pu-239+240/60Co	LM	8.5E-05	1.4E-03	1.5E-03	1.1E-03
	LD	6.1	7.9	2.7	9.3
Pu-238/ ²³⁹⁺²⁴⁰ Pu	LM	2.1E-01	2.0E-01	2.6E-01	2.0E-01
	LD	1.8	1.8	1.2	1.7
Pu-241/ ²³⁹⁺²⁴⁰ Pu	LM LD		1.6E+01 1.7	1.1E+01	2.1E+01 2.0
Am-241/ ²³⁹⁺²⁴⁰ Pu	LM	8.0E-01	8.7E-01	6.7E-01	1.0E+00
	LD	1.9	2.6	2.6	2.4
Cm-242/ ²³⁹⁺²⁴⁰ Pu	LM	4.6E-01	6.5E-02	1.8E-02	9.0E-02
	LD	3.5	8.9	2.5	7.3
Cm-244/ ²³⁹⁺²⁴⁰ Pu	LM	7.7E-01	4.3E-01	2.0E-01	3.1E-01
	LD	3.1	4.7	2.3	2.5
C-14/ ⁶⁰ Co	LM	1.9E-02	7.9E-02	4.9E-02	5.7E-02
	LD	20.8	9.9	3.0	6.4
C-14/ ¹³⁷ Cs	LM	1.7E+00	4.9E-01	2.2E-01	1.8E+00
	LD	1.8	10.1	3.8	9.9
Fe-55/ ⁶⁰ Co	LM	3.5E+00	2.6E+00	6.1E+00	4.6E+00
	LD	3.9	4.0	4.2	3.3
Ni-63/ ⁶⁰ Co	LM	1.4E-02	9.3E-03	1.5E-01	3.9E-02
	LD	2.9	5.7	10.2	4.6
Sr-90/ ⁶⁰ Co	LM	3.8E-03	1.3E-02	1.4E-01	2.8E-02
	LD	3.3	10.3	5.2	8.7
Sr-90/ ¹³⁷ Cs	LM	6.3E-01	1.4E-01	3.7E-01	2.2E-01
	LD	1.4	7.5	2.8	6.8

TABLE 6. REFERENCE SCALING FACTORS FOR VARIOUS BULK LOW LEVEL WASTES

Note: LM = log-mean, LD = log dispersion.

TABLE 7. ESTIMATED C-14/H-3 ACTIVITY RATIO IN	J
INCINERABLE WASTE	

Data period	Average	LM	LD
2003	5.0E-05	3.6E-05	3.3
2004	8.2E-06	1.8E-05	1.7
2005	1.6E-05	4.4E-05	1.8

Note: LM = log-mean, LD = log dispersion.



FIG. 21. Comparison of SF for ⁶³Ni between DAW and incineration ash. (a) Comparison of DAW and ash by scatter diagram; (b) Comparison of DAW and ash by distribution diagram.

4.3.1.2. Incineration in oxidizing conditions

Incinerators are adopted in many countries, including Japan, as an efficient volume reduction treatment process for combustible waste. In this process, organic waste such as paper is decomposed into carbon dioxide, water, etc., through high temperature combustion in an oxidizing atmosphere at between 800°C and 1000°C or so, whereby combustible waste can be greatly reduced in volume and mass.

In the case of the incineration process, therefore, it is considered that the SF for ash generated in the incinerator will be affected by the volatility of radionuclides as a result of the high temperature process of oxidative decomposition by heat and oxygen.

Concerning such radionuclides as ⁶³Ni and ⁶⁰Co, which are metal elements having very low volatility in the temperature range seen in the operating temperatures of the incinerators used in the treatment of combustible waste, there are no conceivable factors that could cause a deviation from the SFs applied for DAW, which is the source of the incineration ash. As shown in Fig. 21, the composition ratio of ⁶³Ni and ⁶⁰Co shows no appreciable difference between DAW generated at actual BWR plants and its incineration ash. And alpha emitting nuclides also have low volatility under high temperature conditions and come under the category of radionuclides free from the influence of the incineration process. Therefore, no special consideration is required for the SFs applied for the SFs applied to DAW and other combustible wastes are considered to be applicable as they are for these nuclides.

Concerning alkali and alkaline earth metals such as ⁹⁰Sr and ¹³⁷Cs, it is also conceivable that these elements take the chemical forms of metal chlorides or nitrates, due to the chlorine and nitrogen contained in combustible waste. Even in this case, as the boiling points of the metal chlorides and nitrates are above 1000°C, the chlorine



FIG. 22. Comparison of SF for ¹⁴C between DAW and incineration ash. (a) Comparison of DAW and ash by scatter diagram; (b) comparison of DAW and ash by distribution diagram.

and nitrogen contained are considered to have little effect on the SFs for DAW. Therefore, it is considered that the same SFs for DAW are also applicable to these radionuclides.

On the other hand, such radionuclides as ¹⁴C and ¹²⁹I are oxidized into a gaseous form, such as CO_2 , and discharged together with the combustion exhaust gas, when combusted by oxygen under high temperature conditions. Concerning these radionuclides, it is considered that the SF for ¹⁴C in incineration ash can be determined by multiplying that for ¹⁴C in DAW by the coefficient of radionuclide migration into the exhaust gas.

As shown in Fig. 22, the composition ratio of ${}^{14}C$ and ${}^{60}Co$ shows a large difference between DAW generated at actual PWR plants and its incineration ash. If this difference (namely the coefficient of migration for ${}^{14}C$) is taken into account, the SFs for incineration ash can be evaluated based on those for DAW. Note that the reverse is also true. Since it is easier to sample and determine SFs from concentrated and homogeneous ash than from heterogeneous DAW, SFs developed for ash can often be used for DAW.

5. QUALITY MANAGEMENT

5.1. GENERAL

The SF method is used to determine the radioactive concentrations of DTM nuclides in waste packages. SFs are based on radionuclide measurements obtained from radiochemical analyses of representative waste samples taken from actual radioactive wastes or are based on calculation models. The derivation of the empirical SFs and their application process involves the following stages:

- (a) Sampling stage;
- (b) Radiochemical analysis stage;
- (c) SF determination stage;
- (d) SF application stage.

In each stage it is necessary to implement an appropriate quality management programme according to the nature of activities to be carried out at each stage. The value of independent analysis and calculation to verify the work done needs to be appropriately considered in SF programme design.

5.2. SAMPLING STAGE

At the sampling stage, a sampling plan must be developed with consideration given to plant characteristics, waste characteristics and other factors, including access for sampling and operator exposures during the sampling process. Appropriate representative samples must be obtained according to a sampling plan. Also, the samples obtained must be investigated and identified regarding the following data items required for SF calculation and evaluation, and the records of these data on the samples obtained must be managed in an appropriate manner:

- (a) Sample identification number;
- (b) Plant name;
- (c) Reactor identification/building identification;
- (d) Reactor type (e.g. BWR, PWR, heavy water reactor, etc.);
- (e) Reactor condition (e.g. operation or decommissioning);
- (f) Waste stream identification;
- (g) Type of waste (e.g. spent resin, concentrates, metal piece, smear, etc.);
- (h) Date of waste generation;
- (i) Date of waste sampling.

When radiochemical analysis data are studied, it is necessary to evaluate radioactive concentrations under constant conditions. For this reason, the radioactive concentrations of waste samples must be corrected to a standard date, usually the date of waste generation, by making decay backcorrections to the radiochemical analysis data on all the samples. This is usually, but not always, provided by the laboratory.

Additionally, from the standpoint of quality management, the management of waste sampling data such as reactor type, waste stream and type of waste is essential in studying the feasibility of the integration or classification of radiochemical analysis data for various kinds of waste packages.

5.3. RADIOCHEMICAL ANALYSIS STAGE

At the stage of analysing the radioactive concentrations of waste samples, the analysis objects are DTM nuclides such as beta emitting nuclides and alpha emitting nuclides; only a very few radionuclides can be measured directly by simply surveying waste samples with a gamma spectrometer. Furthermore, the analysis of those radionuclides involves complicated chemical analysis operations.

It is therefore necessary to ensure that such operations as pretreatment, extraction/separation and measurement are carried out in an appropriate manner in accordance with the properties of the waste samples and the radionuclides to be analysed. To attain this, it is necessary from the standpoint of quality management in radiochemical analysis to prepare a manual for the radiochemical analysis method that ensures that appropriate analysis techniques are applied in each analysis step, and to properly manage the records of analysis results.

In addition, cross-checking, in which the same specimen is analysed by a number of analysis laboratories, is an effective approach to quality management in radiochemical analysis to verify the adequacy of the radiochemical analysis method and the skills of the analyst [28].

It would be preferable to evaluate outliers in the radiochemical analysis data before including them in the database. When including an outlier, the data should be reviewed for possible transcription errors, laboratory errors and possible reasons for the variation. If the outlier is used incorrectly in the calculated SF it may result in an unwarranted increase in dispersion or skewing of the mean value.

5.4. SCALING FACTOR DETERMINATION STAGE

At the stage of finally determining SFs, SF calculation and evaluation are conducted based on radiochemical analysis data obtained through appropriate selection and sampling of representative waste and appropriate radiochemical analyses. In the case of homogenized sampling, radiochemical analysis data are obtained from representative samples of homogeneously mixed wastes or composite waste samples. As a result, SF evaluation using such data gives accurate SF values.

On the other hand, when it is difficult to obtain samples of homogeneously mixed waste, radiochemical analysis data are collected from an appropriate number of waste samples. When SF values are evaluated using a database created by accumulated sampling, it is necessary to take into account sources of the sample data and relative representation of data of particular origin. It may be necessary to split the data set according to numerical compatibility. In this respect, the ISO standard [8] recommends that the accuracy of SF values be ensured by taking the following three measures to satisfy the accuracy requirement:

- (a) Considering the parameters affecting the uncertainty of SFs (e.g. measurement uncertainty, sample size, sample splitting, etc.);
- (b) Obtaining an appropriate number of analysis data values;
- (c) Considering an appropriate manner of sampling.

The following features are recommended to check for quality control of the SF analysis:

- (i) Accuracy of SF, especially with a limited number of samples:
 - Representative sampling can ensure accuracy even with limited data;
 - Analysis of the uncertainties of the SF;
 - If high data dispersion is present, there could be a need to reduce the uncertainty range by collecting more samples.
- (ii) Trend analysis and data grouping:
 - A good practice is to plot different streams in the same diagram in order to detect similar or opposing trends. A statistical test can provide additional support for the visual confirmation.
- (iii) Analysis of the SF evolution with time:
 - Periodic review is needed in order to detect possible changes in the SF during the time of validity.

5.5. SCALING FACTOR APPLICATION STAGE

At this stage, the activity of the key nuclides in the waste package is calculated from gamma radiation measurements on the actual waste package (gamma spectrometry or dose rate measurements) and knowledge of the physical parameters of the waste package (mass, density, distribution in package, etc.). The SFs are then applied to assess the activity of the DTM nuclides.

Appropriate quality management procedures are required to ensure that the waste package conforms to the package contents and constraints that have been assumed in its modelling.

The process of taking the gamma measurements also needs to be well defined and controlled to ensure that the measurements are accurate and reproducible. For example, the settings of the gamma spectrometry system, the location of the spectrometer in relation to the waste package, the number of measurements, etc., should be defined for each package. Similarly where dose rate measurements are used, parameters such as the number, location, etc., of measurements should be defined.

5.6. UNCERTAINTIES ASSOCIATED WITH THE USE OF SCALING FACTORS

The development and use of SFs depends on a number of stages. Each stage introduces some uncertainty in the end result. These uncertainties are introduced through the following stages:

- (a) Sampling;
- (b) Radiochemical analysis;
- (c) Determination of SF;
- (d) Application of SF.

It is not feasible to define uncertainties that will be applicable to all applications of the SF method. Therefore these uncertainties are discussed in broad terms below and would need to be assessed for each application of the SF method.

In general, while the uncertainty may be relatively large for a specific DTM nuclide in a specific waste package, the aggregate inventory of a repository is generally more certain since it involves the averaging effects of large numbers of packages, some of which will be individually overestimated and some of which will be individually underestimated.

5.6.1. Sampling

The samples taken for analysis need to be representative of the waste. For example, in a resin waste stream, samples taken from a non-homogenized resin tank should come from several levels or areas of the tank rather than just from the top of the tank. The samples also need to be representative of the way the waste is either segregated or mixed.

Ideally there should be a sufficient number of samples collected in order to limit the uncertainty to within an acceptable range, although either for operational or historical reasons this may not always be possible. The acceptable range will vary according to the needs of the waste characterization programme, national regulations, waste acceptance criteria (WAC), etc. A factor of 10 is typically used as the bounds for the 'acceptable range'.

The time period over which the samples are taken is important in ensuring that they are representative of the operating conditions seen by the plant. In some cases, there may be plant changes such as a major component replacement or a change in operating parameters (i.e. primary chemistry, operation with failed fuel) that will influence the SFs to such an extent that a new set would need to be derived.

Furthermore, for radionuclides with shorter half-lives, the radiochemical results need to be corrected for the decay of the radionuclide. Where the sample analysed comes from waste collected at a single time, this decay correction is relatively simple. However, where the sample comes from waste accumulated over time, the decay correction is not straightforward and requires additional knowledge about the waste arisings.

5.6.2. Radiochemical analysis

The radiochemical analysis of the samples is an important step in the process because that is the only place where the DTM nuclides are actually measured. For many of the DTM nuclides the chemical methods needed for separation and the activity measurements are complex processes and need to be able to cope with a wide range of activity concentrations. The accuracy and uncertainty of the results depend on the methods utilized, as well as on providing an adequately sized sample for partitioning by the laboratory.

A further problem is how to treat the measurements of radionuclides that are below the minimum detectable activity (MDA) levels of the analytical process used. In many cases the MDA value is assumed in the analysis, but this may require additional analysis with more elaborate techniques to justify the value selected.

5.6.3. Determination of the scaling factor

The uncertainty in the SF value derived in the determination stage is influenced by the following factors:

- (a) Number of radiochemical analysis data points;
- (b) Dispersion or variance of data (regression is based on normal distribution);
- (c) Determination method (geometrical mean, linear regression of logarithm, etc.).

A limited number of radiochemical analysis data points is the most important contributor to the uncertainty associated with the SF estimate. To reduce the uncertainty, additional radiochemical analysis data points may be necessary. However, it is necessary to consider the following [19]:

"The number of data points is sufficient when investment in additional sampling and measurement produces no appreciable improvement in the statistical uncertainty."

At some point additional samples will not reduce uncertainty and some decision will be required to set an appropriate course of action to meet the characterization objectives.

5.6.4. Application of the scaling factor

During this step, the SFs are combined with a measurement on the actual waste package (gamma spectrometry or dose rate measurements) in order to assess the activity of the DTM nuclides. There are two main sources of uncertainties: the first is linked to defining the physical parameters of the waste package, the second is linked to the measurement of the gamma spectrum or radiation level to determine the key nuclide concentration.

The uncertainties linked to the waste package are associated with the conditioning process and the waste package content. These are uncertainties on all the parameters that appear in the calculation of the ETM nuclides from the gamma radiation measurement (i.e. the assumed values in parameters such as the density of the waste, the geometry and distribution of the waste in the package, the distribution of the activity in the waste, the thickness of the shielding, etc.).

The uncertainties linked to the measurement itself (gamma spectrometry or dose rate) can be established on the basis of the appropriate references or standards.

In the case of the use of dose rate measurement, there are the usual uncertainties associated with the measuring instrument, and in the case of handheld instruments the operator variability in taking the measurement. An additional uncertainty is introduced in the calculation of the key nuclides due to the fact that all the gamma emitters contribute to the dose rate. This is further complicated due to the decay of the shorter lived radionuclides, which needs to be accounted for.

6. CONCLUSIONS

6.1. GENERAL APPROACH TO APPLICATION OF THE SCALING FACTOR

Consistent with the IAEA's published safety requirements for Predisposal Management of Radioactive Waste, Including Decommissioning [29], radioactive waste disposal requires planned and systematic actions to provide confidence that the assessment of the radioactive inventory of the waste will satisfy given requirements for quality.

Where radionuclides in waste streams are being produced in a relatively constant or steady state manner, such as occurs in operating nuclear power plants, the decision relating to when and where to use SFs is relatively straightforward. The general principles covering radiological characterization of materials include four basic approaches to the determination of radionuclide activities in radioactive materials; these are:

- (a) Process knowledge: this applies where certain defined radionuclides are used in the original materials. Such an approach is readily applied with material such as sealed sources, smoke detectors, antistatic devices, and certain industrial as well as institutional wastes.
- (b) Direct sampling: this applies to wastes that appear in unique batches that are generally homogenous and involve small quantities or an infrequent generation rate.

- (c) Numerical analytical methods: a computer program is used to track radionuclide activities in various streams from first principles calculations (e.g. calculating release rates from fuel, calculating activation rates), with further tracking through material balance, and activity distributions in various streams.
- (d) Use of SFs or correlations: this approach is used in conjunction with direct sampling and is applied where there are multiple packages and/or batches of material of similar origin.

All of these methods are inter-related and may be used in support of a radiological characterization effort. Process knowledge is developed through documenting the facility operating history. Once this step is completed, it is necessary to begin an overall inventory, as best as can be achieved, of the sources and volumes involved. Where the origins for the wastes can be determined, probably the best approach is to establish a radioactivity basis from available records. If there are substantial volumes of material in discreet and identifiable forms, adoption of an SF methodology may be appropriate. SFs should be evaluated for each discreet stream or material source.

The first demand for the use of SFs is the availability of radiochemical sample data. Initially, it is unlikely that there will be a substantial pool of data for this purpose. Some new samples will have to be collected and evaluated. In the case of an operating facility, sampling should begin with wastes currently being generated. Since it is fresh material, the activity content of the material can be related directly to the activity spectrum derived from the sample. The sample results should be retained and considered in the context of new sample results as they are collected.

6.2. DECOMMISSIONING WASTE

All levels of radioactive materials, including HLW, LILW, VLLW and materials that can be released from regulatory control (i.e. cleared), have to be managed during decommissioning. The amount of material generally increases with a decreasing level of activity concentration; therefore, the SF method is an important tool for determining the activity of DTM nuclides, especially in larger volume, lower activity wastes, if it is shown to be suitable for the material involved.

Prior to the development of a characterization plan, it is necessary to review the historical data from operation to identify different groups of radioactive material. The radiological data from older nuclear power plants are usually very sparse, and extensive characterization plans may be required for the additional waste streams.

A radiological map of the installation, which can be made by means of mobile instrumentation, would be useful in the selection of samples for radiochemical analysis. A good strategy may be to measure the total alpha, total beta and gamma emitters in a large number of samples before specifying how many samples are needed for radiochemical analysis. Representative SFs are obtained by collecting data from samples covering the highest activity concentration range. The number of samples to be collected is determined as part of the characterization plan.

For power reactors that have already established SFs during operation, some types of waste that were generated while the plant was operating will continue to be produced after shutdown. These include waste from radioactive waste processing, fuel pool cleanup resins and DAW, as well as others that may be specific to the facility. If prior to decommissioning a waste characterization programme had been developed using SFs, it is possible to continue to use the SFs following plant shutdown by taking into account the effect of decay on the SFs.

For activated material, SFs can be determined by calculation methods. These may be extended to similar components where appropriate.

Contamination on other components can often be based on the DAW SFs decayed to a suitable material reference date (such as the reactor shutdown date). This can also apply to material removed from walls, floors, etc., in a contaminated area.

6.3. RESEARCH REACTORS

LLW generation in research reactors is very small compared with power reactors. As such, characterization activities may be relatively infrequent and tend to be directed at a single package or waste management campaign. If in the course of such activities a sufficient database cannot be established to support the development of SFs while the plant is in service, it will be necessary to return to the general approach and assess the most efficient manner to proceed with characterization (i.e. identify the streams and volumes characteristic of the reactor facility and on the basis of economic considerations determine if an SF approach would be beneficial).

6.4. FUEL CYCLE FACILITIES

The SFs in fuel cycle facilities such as fuel reprocessing facilities will be different from operational nuclear power plants. They will tend to be dominated by APs of fuel cladding materials, as well as FPs and actinides. Different waste streams (e.g. fuel hulls) will have distinct nuclide ratios. Other fuel cycle facilities, such as fresh fuel fabrication plants, will not have any APs, FPs or transuranics associated with irradiated fuel. In this case, calculated SFs based on the uranium enrichment profile may be appropriate. MOX fuel fabrication facilities will have yet another distinct set of SFs, depending on the fuels being blended, etc.

6.5. COMMON SCALING FACTORS

The SF methods described in Ref. [8] and supported by this report are empirical in that the determination of the SF is based on statistical evaluation of physical measurements. The success of the method is determined by establishing a correlation between the values included in the analysis. If the correlation meets the criteria set for the SF determination, where the data came from (e.g. from a single waste stream within a single plant, from several waste streams within that plant, from all waste streams, from all waste streams of a type within a fleet of facilities) is immaterial. It can be stated that the SF will determine the concentration in that stream or those streams within some confidence interval and that the result would be acceptable for characterization based on the acceptability of the confidence level implied.

Experimental results from multiple waste streams and nuclear installations can often be combined to produce one or more common SFs that are representative of the overall streams and installations. (The use of common SFs is viewed as a valid basis to reduce sampling and radiochemical analysis requirements. The term 'common' is used in lieu of 'generic' on the premise that generic usually does not indicate a boundary of applicability. The SF methods discussed here are always subject to a boundary condition.)

SF calculations must be supported by specific radiochemical analysis of the waste involved. The usefulness of data from other installations should be evaluated and properly vetted before being applied to specific wastes. Common SFs are applicable for specific wastes of nuclear installations after a validation process has been made. This SF may not be valid for other wastes.

Similar installations often change system components for operational reasons, and these differences could become significant. Therefore, special attention should be paid to verify the applicability of previous common SFs after changes to plant components or operating conditions.

Generic SFs published in the open literature or other sources should be only used for qualitative purposes for similar waste streams if those streams have not been previously sampled and verified/validated for the facility in question.

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Annex

COMPILATION OF INTERNATIONAL EXPERIENCE WITH SCALING FACTOR DEVELOPMENT AND USAGE

The following experience is drawn from nuclear power reactors; much of what is discussed below in the context of methodology can be extended to other nuclear facilities, including fuel reprocessing and research reactors.

A-1. BELGIUM

A-1.1. Introduction

There are seven PWR units in Belgium, all of which are different. Due to this, and to the cost of DTM nuclide measurement, the Belgian operators do not follow the 'empirical' SF technique based on a great number of measurements.

Low level radioactive waste generated from these nuclear power plants is stored on the site of Belgoprocess, an affiliate of the ONDRAF/NIRAS State agency (see Fig. A–1), before disposal in the planned Dessel Disposal Centre.

Acceptable activity or activity concentration limits are regulated for individual radionuclides (²²⁶Ra, ²³²Th) or groups of radionuclides in accordance with the conditions of storage. The main regulation is given for groups of radionuclides in accordance with the conditions of the assumed disposal site (X criteria are linked with the intrusion scenario Y and with the radiological capacity of the repository).

Both criteria are expressed in the form:

$$\sum \frac{a_i}{a_{i,\max}} \le 1$$

The SF method is applied for determining the activity of all DTM nuclides. To evaluate the SFs between DTM nuclides and key nuclides, the activity concentration has been calculated through a radiochemical mass transfer model for all of the nuclear power plants. Each of the individual waste elements (filters, spent resins) or

	Homogeneous solid	Encapsulated solid waste packages			
	waste packages	Untreated product	Compacted product	Caged product	
Conceptual package model (cross-section)		Historic			
Waste to be solidified	Evaporator concentrate, low activity spent resins, etc.	, Metals, concrete, gas filter, etc. (DAW)		(DAW)	

FIG. A-1. Waste packages intended for disposal at the LLW disposal centre.

			Ke	Key nuclide		
Type of nuclide	Evaluated DTM nuclide	Evapo. concentrate, resins, Doel	Resins, Tihange	Filters	Waste conditioned by Belgoprocess	
AP	H-3	None	None	None	Not requested	
СР	C-14, ³⁶ Cl, ⁵⁹ Ni, ⁶³ Ni, ⁹⁴ Nb	Co-60	Gamma emitting (dose rate)	Gamma emitting (dose rate)	Gamma emitting (dose rate)	
FP	Sr-90, ⁹⁹ Tc, ¹²⁹ I	Cs-137	Gamma emitting (dose rate)	Gamma emitting (dose rate)	Gamma emitting (dose rate)	
Transuranic nuclide	Ra-226, ²³⁴ U, ²³⁵ U, ²³⁸ U, ²³⁷ Np, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴¹ Am	Cs-137	Gamma emitting (dose rate)	Gamma emitting (dose rate)	Gamma emitting (dose rate)	

TABLE A-1. SUMMARY OF DIFFICULT TO MEASURE AND KEY NUCLIDES

types of waste (evaporator concentrate, burnable waste, non-burnable waste, oil, etc.) has its own calculated SF, taking into account the plant's fuel failure history, use of MOX, component materials, etc. These SFs are updated each year according to the mean activity of ⁶⁰Co, ¹³¹I, ¹³³I, ¹³⁴I, ¹³⁴Cs and ¹³⁷Cs measured in the primary loop.

Additionally, the gamma emitting nuclides ⁶⁰Co and ¹³⁷Cs were selected as key nuclides because of their detectability when measured by non-destructive radioactivity measuring equipment and the similarity of the production mechanisms with respect to the CP and FP/transuranic nuclide.

Wherever possible, based on qualification of the device, availability, activity concentration and detection limits, a gamma spectrometry device is used. For many waste packages, however, only dose rate measurement is available.

A-1.2. Radionuclides of interest

The SF method has been applied to the DTM nuclides shown in Table A–1. Key nuclides are selected based only on similarity of the production mechanism.

Gamma spectrometry is applied for the conditioned drum of concentrate or resins in the case of Doel and on samples of concentrate in the case of Tihange. It is foreseen to qualify a gamma spectrometry device on conditioned drums on the Tihange site, to avoid the decay effect of short lived nuclides (e.g. ⁵⁸Co). If the ¹³⁷Cs activity is below the detection limit when using a gamma spectrometry device, the declared activity is the calculated activity.

A-1.3. Sampling and radiochemical analysis

Sampling and radiochemical analysis are used only to qualify the calculational model on a five year basis. The focus is on the most critical isotopes for the acceptance criteria (contribution to the X and Y criteria as noted previously): ¹⁴C, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I and ²³⁹⁺²⁴⁰Pu.

A few samples of resins are collected on the Tihange site. (There is no recirculation loop on the Doel site collecting tank to allow proper mixing before sampling.) A few samples of concentrates are collected on both sites. For filters, a special loop was used to collect activity from selected primary loops (Tihange 3 and Doel 3). It is important to obtain radioactive waste having an activity concentration high enough to be measured in the laboratory.

It is also important to know the status of the unit during the production of the sampled waste in order to compare the measured activity with the calculated activity using the most detailed data (all input data of the model must be set to the period of production of the sampled waste).

A-1.4. Validation of the scaling factor method

The measurements are compared with the calculated activities and, in the event of differences, the model is investigated. Usually, differences are the result of modifications on the units, such as:

- (a) Chemistry of the primary loop reducing the corrosion rate;
- (b) Reduction or better knowledge of the amount of impurity in the alloys;
- (c) Evolution of the fuel assembly composition, etc.

The calculated activity is intended to be conservative, or at least a best estimate, in comparison with the measured activity. The lack of coherence between measurements can lead to rejection of one of the measurements.

The LLWAA code, which is used for modelling, has been qualified and accepted by ONDRAF/NIRAS, the Belgian Agency for Radioactive Waste and Enriched Fissile Materials, after an in-depth critical review performed by Association Vinçotte Nucléaire, an organization licensed by the Belgian Government in the field of nuclear safety.

A-1.5. Calculation method

The LLWAA code is site specific, as it takes into account the design characteristics and operating conditions of the different units of a site with a centralized waste treatment facility. The calculation of the activity inventories for each waste package requires the proper modelling of the reactor coolant system (RCS), the nuclear auxiliary systems (NAS) and the liquid waste processing system (LWPS).

Basically, the code parameters are classified into three categories:

- (a) Category 1: containing all parameters related to the operating conditions that can change or be modified from one cycle to another. The values of these parameters are the main input data of the code and are based on the routine reactor coolant measurement results (⁶⁰Co, ¹³¹I, ¹³³I, ¹³⁴I, ¹³⁴Cs and ¹³⁷Cs activities and chlorine concentration). The type of fuel (MOX or ²³⁵U enriched fuel), fuel cycle length or rated plant power are additional input values.
- (b) Category 2: parameters pertaining to the plant basic characteristics and operating procedures; among those we find RCS, NAS and LWPS flow rates, boron content in the RCS and evaporators, etc.
- (c) Category 3: core activity inventories (calculated by means of the ORIGEN2 code), chemistry of the primary coolant, equipment materials and surface areas, equipment corrosion rates, characteristics of the ion exchange resins and filters, physicochemical forms of the activity in the RCS (fraction of activity present in soluble and insoluble forms) during normal operation and refuelling outage and diffusion coefficients through fuel cladding defects.

A-1.6. Data maintenance

The SFs are updated each year according to the mean activity of ⁶⁰Co, ¹³¹I, ¹³³I, ¹³⁴I, ¹³⁴Cs and ¹³⁷Cs measured in the primary loop the year before. In the event of large modification of the input data during the year (fuel incident), the need for a new assessment of the activity of the waste produced must be evaluated. However, due to the delay between the contamination of the waste and the assessment of the activity, the risk of having to do such a new calculation is low. In the event of planned modification of the unit (e.g. for steam generators, fuel assembly type), the model has to be adapted before the modification. Every five years a sampling and measurement campaign takes place in order to verify the validity of the model.

A-2. BRAZIL

A-2.1. Introduction

There are two nuclear power reactors in Brazil, both part of the Central Nuclear Almirante Álvaro Alberto (CNAAA):

(a) Angra 1 (PWR, 657 MW(e), operational since 1982).

(b) Angra 2 (PWR, 1350 MW(e), operational since 2000).

A third unit, Angra 3, very similar to Angra 2, is planned to be operational by 2014. The CNAAA is located at Itaorna Beach, Angra dos Reis, Rio de Janeiro State, and is responsible for 3.3% of the electric power generated in the country today.

Eletronuclear, a federal government owned company, operates both units of CNAAA and a radioactive waste management facility (initial storage), which receives intermediate and low level radioactive waste from CNAAA.

The initial storage facility is located at the CNAAA site. The concept and location for a national final repository are still under study. One of the possibilities is an engineered surface repository adjacent to the initial storage site.

Currently, 5894 radioactive waste packages (2186.3 m³) are stored in the initial storage facility, including drums, boxes, liners and reinforced concrete shields (VBAs). A classification of these packages based on \dot{H}_{M} , the maximum dose equivalent rate on any external surface, is presented in Table A–2.

The regulatory framework in Brazil is established in Acceptance Criteria for Disposal of Low and Medium Level Radwaste (CNEN-NN-6.09, September 2002), where the following statement is found:

"Radionuclides content – Type, composition and content of radionuclides in treated radioactive waste in its final form must be known and documented with enough precision to provide evidence of conformity within the authorized limits."

A-2.2. Radionuclides of interest

Regarding the radionuclides relevant to the characterization of ILLW from CNAAA's PWR reactors, the following steps were taken:

- (a) Elaboration of a catalogue containing pertinent data for radionuclides of interest, including FPs, activated CPs, activated coolant products and isotopes of uranium and transuranic elements, comprising some 71 radionuclides.
- (b) Identification of the DTM nuclides among those included in the catalogue, comprising some 37 radionuclides.
- (c) Selection of the radionuclides for evaluation among the identified DTM radionuclides, comprising some 23 radionuclides.

TABLE A–2. CLASSIFICATION OF RADIOACTIVE WASTE PACKAGES ACCORDING TO THE MAXIMUM DOSE EQUIVALENT RATE

Maximum dose equivalent rate	Number of packages	Fraction of total number
$\dot{H}_{M} \le 2 \text{ mSv/h}$	4950	83.98%
$2 \text{ mSv/h} < \dot{H}_M \le 20 \text{ mSv/h}$	587	9.96%
\dot{H}_{M} > 20 mSv/h	357	6.06%

- (d) Definition of a priority set of radionuclides among the DTM radionuclides selected for evaluation, comprising some 15 radionuclides.
- (e) Choice of key nuclides to be used in the evaluation of selected DTM nuclides, comprising some six radionuclides (three standard and three alternative).

The DTM nuclides selected for evaluation were:

- (i) Priority set: ³H, ¹⁴C, ⁵⁵Fe, ⁵⁹Ni, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ²³⁸Pu, ²³⁹Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴²Cm, ²⁴³Cm.
 (ii) Additional nuclides: ⁹³Zr, ^{108m}Ag, ¹³⁵Cs, ²³⁵U, ²³⁸U, ²⁴⁰Pu, ²⁴²Pu, ²⁴⁴Cm.

Key nuclides to be used in the evaluation of selected DTM nuclides are presented in Table A-3.

Alternative key nuclides that may be used for characterization of fresh radioactive waste are presented in Table A-4.

Once measured, ²³⁸U can be used directly as a key nuclide for ²³⁵U.

Tritium will be evaluated by direct measurement using radiochemical methods or using measured ³H specific activity in the reactor primary coolant applied to the moisture contained in the radioactive waste sample, as the most common chemical form of tritium in PWR reactors is tritiated water.

Use of non-radiochemical methods (mass spectrometry) may be required to measure the specific activity of ⁹⁹Tc, ¹²⁹I and ¹³⁵Cs in radioactive waste samples, since these are present only at very low levels.

The specific activity of gamma emitting nuclides in already stored waste packages will be measured using gamma ray spectroscopy systems that employ a high resolution germanium detector as the main device. Drums will be measured using an SGS system, whereas boxes, liners and VBAs will be measured using an in-situ object counting system (ISOCS).

A computer program is in development to calculate the specific activity of gamma emitting and DTM nuclides in each package. The detailed isotopic inventory of the final repository will be calculated as a function of time and disposal rate by a second computer program, the development of which is also in progress.

TABLE A-3. KEY NUCLIDES TO BE USED IN THE EVALUATION OF SELECTED DIFFICULT TO MEASURE NUCLIDES

DTM nuclide selected for evaluation	Key nuclide
C-14, ⁵⁵ Fe, ⁵⁹ Ni, ⁶³ Ni, ⁹³ Zr, ⁹⁴ Nb, ⁹⁹ Tc, ^{108m} Ag	Co-60
Cs-135	Cs-137
Sr-90, ¹²⁹ I, ²³⁹ Pu	Co-60, Cs-137
U-238, ²³⁸ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴¹ Am, ²⁴² Cm, ²⁴³ Cm, ²⁴⁴ Cm	Pu-239

TABLE A-4. ALTERNATIVE KEY NUCLIDES THAT MAY BE USED FOR FRESH RADIOACTIVE WASTE

DTM nuclide selected for evaluation	Alternative key nuclides
Zr-93	Zr-95
Ag-108m	Ag-110m
Pu-239	Ce-144

A-2.3. Basic sampling plan

Radioactive waste samples for radiochemical analysis are collected considering:

- (a) CNAAA units: Angra 1 and Angra 2.
- (b) Waste stream: primary spent resins, cartridge filter elements, evaporator concentrates, DAW.
- (c) Operation cycle.
- (d) Statistical criteria to achieve representative sampling.

As a consequence, the classification of the obtained results (either SFs or correlation functions) is carried out according to the CNAAA unit and waste stream. Although both units are PWR reactors, Angra 1 is a Westinghouse project whereas Angra 2 is a KWU project.

Checking and updating of the results obtained are planned for every operation cycle of a given unit.

A-2.4. Evaluation methodology

The evaluation of a selected DTM nuclide will be performed using the methodology described below. As a first attempt, search for a useful SF given by:

$$SF = \frac{A_{\rm DTM}}{A_{\rm KN}}$$

where A_{DTM} is the specific activity of a DTM nuclide and A_{KN} is the specific activity of the key nuclide. Determine the geometrical mean following the steps described below:

- (a) Compute the value of the SF obtained from measurements on each sample;
- (b) Compute the ln (log base e) of each value;
- (c) Add all the values together;
- (d) Divide the result by the total number N of values;
- (e) Take the exponential to obtain the average value of the SF;
- (f) Compute the value of 2σ dispersion for the data set;
- (g) Check if the value of 2σ dispersion is lower than or equal to 10.

The average value of the SF is therefore:

$$\overline{\rm SF} = e^{\left(\sum_{i=1}^{N} \ln(\rm SF)_i/N\right)}$$

whereas the value of 2σ dispersion is given by:

$$\mathbf{D}_{2\sigma} = \mathrm{e}^{2 \cdot \left(\sqrt{\sum_{i=1}^{N} \left[\ln(SF)_i - \ln(\overline{SF})\right]^2 / N - 1}\right)}$$

The existence of a useful SF for the data set is defined by the applicability criterion, which requires that 95.5% of the SF values obtained from measurements must be within the range:

$$\frac{\overline{\mathrm{SF}}}{10} \le (\mathrm{SF})_i \le \overline{\mathrm{SF}} \cdot 10 \implies D_{2\sigma} \le 10$$

As a second attempt, if no useful SF is obtained, the search for a correlation function is carried out on the assumption that a non-linear relationship between the specific activities exists:

$$A_{\rm DTM} = a \cdot (A_{\rm KN})^b$$

where a and b are constants. This relation can be written in a more convenient form:

$$\ln(A_{\rm DTM}) = \ln(a) + b \cdot \ln(A_{\rm KN})$$

so that a straight line can be fitted to the ln values of the measured specific activities of the involved radionuclides, using the least square method. This calculation method is also known as linear regression of logarithms, where b is the angular coefficient (slope) and $\ln(a)$ the linear coefficient (intercept) of the fitted straight line.

Using the notation:

$$x_i \equiv \ln(A_{\rm KN})_i$$
 $y_i \equiv \ln(A_{\rm DTM})_i$ $\sigma_i \equiv \text{uncertainty of } \ln(A_{\rm DTM})_i$

the least square method calculations become:

$$\begin{bmatrix} \ln(a) \\ b \end{bmatrix} = \frac{1}{\Delta} \begin{bmatrix} \sum_{i=1}^{N} \frac{x_i^2}{\sigma_i^2} & -\sum_{i=1}^{N} \frac{x_i}{\sigma_i^2} \\ -\sum_{i=1}^{N} \frac{x_i}{\sigma_i^2} & \sum_{i=1}^{N} \frac{1}{\sigma_i^2} \end{bmatrix} \cdot \begin{bmatrix} \sum_{i=1}^{N} \frac{y_i}{\sigma_i^2} \\ \sum_{i=1}^{N} \frac{y_i x_i}{\sigma_i^2} \end{bmatrix}$$

where

$$\Delta = \left[\sum_{i=1}^{N} \frac{1}{\sigma_i^2}\right] \cdot \left[\sum_{i=1}^{N} \frac{x_i^2}{\sigma_i^2}\right] - \left[\sum_{i=1}^{N} \frac{x_i}{\sigma_i^2}\right]^2$$

The uncertainty associated with each one of the fitted coefficients is calculated by means of the following expressions:

$$\sigma_{\ln(a)} = \sqrt{\sum_{i=1}^{N} \frac{x_i^2}{\sigma_i^2} / \Delta} \qquad \sigma_{b} = \sqrt{\sum_{i=1}^{N} \frac{1}{\sigma_i^2} / \Delta}$$

A measure of the degree of correlation between the two specific activities involved is most properly provided by a quantity called the CC:

$$\mathbf{r} = \frac{\displaystyle\sum_{i=1}^{N} x_i y_i - \frac{\left(\sum_{i=1}^{N} x_i\right) \cdot \left(\sum_{i=1}^{N} y_i\right)}{N}}{\sqrt{\left[\left(\sum_{i=1}^{N} x_i^2 - \frac{\left(\sum_{i=1}^{N} x_i\right)^2}{N}\right) \cdot \left(\sum_{i=1}^{N} y_i^2 - \frac{\left(\sum_{i=1}^{N} y_i\right)^2}{N}\right)\right]}}$$

The existence of a correlation function for the data set is defined by the applicability criterion, which requires that the CC for the fit should have a value $r \ge 0.6$ to be considered significant.

Third attempt: if a DTM nuclide cannot be evaluated by means of key nuclides using an SF or a correlation function, its DTM specific activity will be considered equal to the arithmetical mean of data obtained from direct radiochemical measurements on samples of a given radioactive waste stream.

A very important additional issue is the applicability of this methodology in information retrieval from historical radioactive waste.

According to international experience, most operational changes in PWR reactors (small fuel failure, steam generator replacement, extended shutdown, zinc addition in the primary coolant, etc.) have no significant impact on SF values. This opens the possibility for utilization of currently determined SFs and correlation functions in the characterization of already stored radioactive waste, therefore avoiding destructive assay of sealed radioactive waste packages. Studies are in progress in Brazil to consolidate this approach.

A-2.5. Programme organization

The Brazilian characterization programme for low and intermediate level radioactive waste is presented in Fig. A–2.



FIG. A-2. Brazilian characterization programme for low and intermediate level radioactive waste.

Regarding each of the stages shown in Fig. A–2, the present status of the characterization programme can be summarized as follows:

- (a) Sampling: evaporator concentrates samples have been collected periodically since 2005. Some samples of spent primary resins and DAW were collected in 2007. Many issues related to sampling techniques are being studied.
- (b) Radiochemical analysis: preliminary measurements were performed this year for the first time. Studies on analytical procedures for each of the selected DTM nuclides in specific radioactive waste streams are currently in progress.
- (c) Determination of SFs and correlation functions: only the selection of DTM nuclides to be evaluated, choice of key nuclides to be used and study of evaluation methodology to be employed have been concluded.
- (d) Development of computer programs: simulations based on generic data have been carried out in order to test and improve the structure of the programs.
- (e) Other stages of the programme: not implemented.

A-3. CANADA

A-3.1. Introduction

There are 22 commercial sized nuclear power plants in Canada (Table A–5). Ontario Power Generation (OPG) owns 20 of the power reactors, located in the Province of Ontario in Canada, eight of which are leased to Bruce Power for operation. In addition, New Brunswick Power and Hydro-Québec operate one unit each, Point Lepreau and Gentily 2, respectively. There are also several prototype and demonstration reactors, which are now shut down. All the operating commercial reactors (and most of the prototype and demonstration reactors) are of the CANDU type PHWRs, which use unenriched uranium fuel and heavy water in both their primary heat transport and moderator systems.

Both OPG and Bruce Power have started an environmental assessment process to construct new reactors in Ontario, which may not necessarily be CANDU reactors. A number of different reactor designs are under consideration, including CANDU, advanced CANDU, BWR and PWR designs. New Brunswick Power has also launched preliminary studies for new build reactors, as well as a power company in the province of Alberta.

Under Canadian regulations, each waste owner is responsible for the life cycle management of the waste it produces (i.e. there is no national repository in Canada nor a single agency that manages all LILW).

Unlike in other jurisdictions, the regulator in Canada, the Canadian Nuclear Safety Commission (CNSC), is not prescriptive and does not mandate the manner in which waste must be characterized or which radionuclides must be reported. The licensee of the waste management facility must put forward a safety case based on site specific information, including the characteristics of the waste. The CNSC reviews the safety case and, if it finds it to be acceptable, will grant a licence. The licence must be renewed periodically (typically every five to ten years, depending on the facility). Before being granted a licence renewal, the licensee must prove to the satisfaction of the CNSC that continued operation of the facility is still safe.

OPG operates a centralized waste management facility which receives waste from both OPG and Bruce Power plants. Bulk LLW received at the facility is typically segregated into incinerable, compactable and nonprocessible waste categories (Table A-6). These are dispositioned as follows:

- (a) Incinerable waste is fed into an incinerator, resulting in the production of bottom and baghouse ashes as the final products of combustion;
- (b) Compactable waste is compacted into 2.5 m^3 metal boxes and stored;
- (c) Non-processible wastes are packaged and stored as received because they are not amenable to processing via either incineration or compaction.

Name	Reactor type	Location/operator	Date of first operation	Current status
Bruce A	4 × 769 MW(e) (net) CANDU PHWR	Ontario/Bruce Power ^a	1977, 1977, 1978, 1979	Two units operating, two units being refurbished
Bruce B	4 × 860 MW(e) (net) CANDU PHWR	Ontario/Bruce Power ^a	1985, 1984, 1986, 1987	Four units operating
Darlington	4 × 881 MW(e) (net) CANDU PHWR	Ontario/OPG	1992, 1990, 1993, 1993	Four units operating
Gentilly 2	638 MW(e) (net) CANDU PHWR	Quebec/Hydro- Québec	1983	Operating
Pickering A	4 × 515 MW(e) (net) CANDU PHWR	Ontario/OPG	1971, 1971, 1972, 1973	Two units operating, two units shut down in 1998
Pickering B	4 × 516 MW(e) (net) CANDU PHWR	Ontario/OPG	1983, 1984, 1985, 1986	Four units operating
Point Lepreau	635 MW(e) (net) CANDU PHWR	New Brunswick/ New Brunswick Power	1983	Operating

TABLE A-5. SUMMARY OF CANADIAN NUCLEAR POWER PLANTS

^a Bruce Power reactors are leased from OPG.

TABLE A-6. SUMMARY OF TYPICAL WASTE STREAMS

Typical low level waste streams	Typical intermediate level waste streams	
Incinerator bottom ash	Primary coolant purification system IX resins	
Incinerator baghouse ash	Moderator purification system IX resin	
Compacted (bales)	Filters and filter elements	
Compacted (boxes)	Irradiated core components	
Non-processible (boxes)	Fuel channel wastes from reactor refurbishment	
Non-processible (drummed)		
Non-processible (large objects)		
Low level IX resins		
Active Liquid Waste Treatment System sludge		

High activity resins and filter waste are considered to be ILW and are stored in shielded, underground structures at the waste management facility.

At present, OPG is working on a plan to build a deep geological repository (DGR) adjacent to its current LILW management site. The facility, planned for startup by about 2018, will accept both LLW and ILW arising from operational and reactor refurbishment activities of the reactors owned by OPG (including those operated by Bruce Power). Decommissioning wastes are not included in the DGR application. The DGR is based on engineered deep rock caverns approximately 680 m below the surface in a limestone formation overlain by a thick layer of shale. The groundwater movement in the formation is very slow, and contaminant transport is diffusion controlled.

Waste from the Gentilly 2 and Point Lepreau reactors is stored locally in waste management facilities at each of these sites. Some low force compaction in carried out on the wastes prior to storage. Wastes from research facilities and other non-power producing nuclear facilities are generally managed by Atomic Energy of

Canada Limited on a contract basis with various brokers. These waste owners have not yet announced plans for the long term management of their wastes.

Based on the worldwide use of the SF methodology, its application for characterizing the activity of DTM nuclides is generally considered to be acceptable in Canada. In this context, and unlike in other jurisdictions (see Section 3.2), the CNSC has to date not expressed a concern with specific issues such as:

- (a) The appropriateness of 60 Co or 137 Cs as a scaling nuclide; or
- (b) The merits of using an arithmetic versus a geometric mean SF value.

From the CNSC's perspective, the generator is ultimately responsible for the appropriate measurement and accounting of all radionuclides that are of concern both for short term waste handling and long term safety of the planned repository. The CNSC's primary requirement is that the radiological risk to the critical individual from disposal of waste in a planned repository be demonstrated to be acceptably low.

A-3.2. Radionuclides of interest

For OPG's DGR, the main radionuclides of interest for the pre-closure safety assessment are (Table A-7): ³H, ¹⁴C, ⁶⁰Co, ⁹⁰Sr, ¹³⁷Cs, ²⁴¹Am and ²⁴¹Pu. For the post-closure safety assessment, the main radionuclides of interest are: ¹⁴C, ³⁶Cl, ⁹³Zr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ¹³⁵Cs, ²³³U, ²³⁸U, ²³³Pa, ²³⁷Np, ²⁴¹Am and ²⁴¹Pu.

SFs have also been developed for other DTM nuclides, such as 55 Fe, 59 Ni, 63 Ni, 79 Se, 126 Sn, 242 Cm, 244 Cm and 243 Am.

A-3.3. Sampling and radiochemical analysis

OPG and its predecessor companies have been conducting waste characterization research since the 1970s. In support of the planned repository for LILW, a focused programme on waste sampling and analysis was initiated in 1999 and completed in 2006. Between five and ten samples of various wastes were characterized each year. The samples included bulk LLW and ash samples, as well as ILW resins and filters.

Generally, waste streams are randomly sampled. Bulk LLW is sampled at the centralized waste management site. As a result of waste receipts at the site from various plants being processed and stored together, information on the specific origin of the waste is often obscure. ILW such as filters and resins, on the other hand, is more conveniently sampled at the station, and hence the sample origin is readily known.

Based on the gamma spectrometry results for the collected samples, further detailed alpha/beta analysis is carried out on only selected samples. The scope of analysis depends on the type of sample. Generally, only primary coolant system resin samples are analysed for radionuclides such as ³⁶Cl, ¹²⁹I and ⁹⁹Tc because their concentrations are expected to be undetectable in the other wastes.

In addition to the development of radiochemical data based on waste samples, limited in situ gamma spectrometry campaigns have also been carried out on waste containers and the applicability of the dose rate to activity conversion methodology has been investigated.

Key nuclide (marker)	Typical DTM nuclide	Key nuclide (marker)	Typical DTM nuclide
Co-60	H-3 (based on $^{3}H/^{14}C$ ratio)	Cs-137	Sr-90
	C-14		I-129
	Cl-36		Cs-135
	Fe-55		Pu-239+240
	Ni-63		(used for scaling other actinides
	Tc-99		based on calculated ratios in fuel)

TABLE A-7. SUMMARY OF KEY NUCLIDES

Direct measurement of tritium concentrations in bulk LLW presents a significant challenge. Attempts to develop SFs for tritium in bulk LLW have been generally unsuccessful. However, the availability of ¹⁴C and tritium emissions data from a radioactive waste incinerator offered a unique opportunity to correlate these two radionuclides. C-14 can be correlated to ⁶⁰Co, then tritium correlated to ¹⁴C.

A-3.4. Calculation method

Owing to the relatively small size of OPG's current SF database, the present waste stream data are analysed without regard to the sample origin. The available data are considered as a base set, with the size of the database for several streams being generally too small to develop station specific data or to establish a trending pattern.

The method of analysis is similar to that employed in the USA. Based on the well established fact that SF data generally conform to a log-normal distribution, log-mean and LD values are determined after due consideration of outliers.

The key (marker) nuclides used by OPG are ⁶⁰Co and ¹³⁷Cs. SFs are calculated based on:

- (a) Log-mean average of measured samples;
- (b) Theoretical prediction from reactor coolant data and reactor component compositions (confirmed by a few samples);
- (c) Theoretical predictions from reactor fuel burnup calculations (for transuranic and other fuel based nuclides).

A linear relationship is assumed between the DTM nuclide and the key nuclide:

Activity of DTM = activity of key nuclide \times SF

Table A-8 provides a summary of typical scaling factors for CANDU primary heat transport (PHT) and moderator resins, compacted waste and bottom ash from incineration.

A-3.5. Other considerations

In addition to the results for sampled wastes, smear and particulate samples from various system components characterized in support of past and ongoing health physics programmes have also been assimilated into the database. Also included in the database are data obtained from characterization of various reactor components such as steam generator tubes, feeder pipes, pressure tubes and end fittings.

Dose rate to activity conversion is the primary way to measure the key nuclides for application of SFs. Dose rate is measured on a waste package, then a conversion factor is used to derive the package activity for the key nuclides. This is an easy measurement to obtain in the field without complex equipment or procedures.

De die eerdigte weine		Typical scaling factor			
Radionuclide pair	Bottom ash	Compacted waste	PHT [[please define]] IX resins	Moderator IX resins	
C-14/ ⁶⁰ Co	4.9×10^{-2}	8.6×10^{-2}	3.0×10	4.1×10	
Cl-36/60Co	1.0×10^{-6}	1.0×10^{-6}	$2.2 imes 10^{-5}$	1.0×10^{-5}	
Sr-90/ ¹³⁷ Cs	3.7×10^{-1}	1.4×10^{-1}	$1.7 imes 10^{-2}$	n.a.	
I-129/ ¹³⁷ Cs	3.5×10^{-7}	3.5×10^{-7}	1.2×10^{-6}	n.a.	
Am-241/ ¹³⁷ Cs	6.7×10^{-3}	1.2×10^{-2}	$2.8 imes 10^{-5}$	n.a.	

TABLE A-8. SUMMARY OF TYPICAL SCALING FACTORS

n.a.: not available.

The dose rate for every waste package is recorded in the integrated waste tracking system, which also includes the waste characterization information for each waste stream. From this, a complete waste package radionuclide inventory can be calculated.

A-3.6. Data maintenance

Future plans to update the database will be based on the outcome of the DGR safety assessment studies.

A-4. CHINA

A-4.1. Introduction

Nine reactors are currently in operation and two are under construction in China (Table A–9). About 1000 m³ of LILW is produced each year. According to the plans for nuclear energy development of China, the nuclear electricity capacity will be 40 000 MW in 2020, and three nuclear power plants will be built each year. Four thousand cubic metres of LILW will be generated each year by 2020.

Compared with waste from nuclear power plants, radioactive waste from the older nuclear industry, nuclear research centres and nuclear medical systems is much more complicated. These legacy wastes are being treated and will be treated. Some old facilities are waiting for decommissioning, and new waste will be generated during the dismantling and decontamination of the nuclear facilities.

Aqueous legacy wastes are evaporated and the concentrates are solidified in a matrix of cement or bitumen, according to the radioactive level and the salt content of the concentrates. Organic radioactive waste will be incinerated, and the ashes are being studied for a proper method of conditioning. Ion exchange resins are embedding in cement or are studied to discover other good conditioning means.

The situation for DAW is very complicated in China. All kinds of DAW, including metal equipment parts, ventilation filters and other miscellaneous wastes, have been mixed together in a pit without any sorting. Therefore, China is now conducting a research project on demonstration of retrieving, sorting and conditioning of DAW. In this project, the waste will be retrieved from the pit and sorted according to the radioactive level and physical properties. The compactable wastes will be supercompacted and conditioned in cement in a 200 L drum. The incinerable waste will be burnt in an incinerator.

There are two LILW disposal sites in China: Beilong disposal site and Yumen disposal site. Beilong disposal site is situated in Guangdong province in south-east China and has a capacity of 240 000 m³. Beilong disposal site construction was started in 1998 and was finished in 2000. The Beilong disposal site so far not been put into use. The Yumen disposal site, with a capacity of 200 000 m³, is in the Guansu province in north-west

Nuclear power plant	Number	Reactor	Power (MW(e))	Date of construction	Date of operation
Qinshan I	CN-1	PWR	300	1985-03-21	1991-12-15
Daya Bay	CN-2	PWR	984	1987-08-07	1993-08-31
Daya Bay	CN-3	PWR	984	1988-04-07	1994-02-07
Qinshan II	CN-4	PWR	600	1996-06-02	2002-04-15
Qinshan II	CN-5	PWR	600	1996-06-02	2004-05-03
Ling Ao	CN-6	PWR	984	1997-05-15	2002
Ling Ao	CN-7	PWR	984	1987-08-07	2003
Qinshan III	CN-8	PHWR	700	1998-06-08	2002-12-31
Qinshan III	CN-8	PHWR	700	1998-09-25	2003-07-24
Tian Wan	CN-10	WWER	1000	1999-10-20	
Tian Wan	CN-11	WWER	1000	1999-10-20	

TABLE A-9. NUCLEAR POWER PLANTS IN CHINA
China and was prepared for operation in 1999. The two sites cannot accommodate China's LILW, thus China is planning to build another five disposal sites in five districts located in the north-west, south, north, east and north-east regions.

According to China's requirements for the disposal of LILW, waste entering the disposal sites should be analysed for total radioactivity and the main radionuclides. China has been conducting research for the measurement of the total alpha–gamma radioactivity of waste in drums by non-destructive means. Planning for the destructive analysis method has just started. Therefore, means for the measurement of radionuclides and their concentrations in waste are now necessary. The SF method for DTM nuclides in a waste package has not yet been set up. The experience concerning the SF method is currently limited to the measurement of some particular radionuclides.

A-4.2. Radionuclides of concern

According to China's GB913-1995 standard, enforced in 1996, solid radioactive wastes are classified into 11 types according to the radioactive level (A), the half-life $(\tau_{1/2})$ of the contained radionuclides and the heat release rate, as shown in Table A–10.

LILW will be disposed of in near surface disposal sites and high level and alpha bearing waste will be disposed of in a DGR. In 1997, China issued a guideline, GB 16933-1997, to regulate the acceptance of radioactive wastes by disposal sites; this guideline was enforced in 1998. Based on the acceptance guideline, the wastes to be disposed of should be examined for different parameters by random sampling. One of the parameters to be measured is the main radionuclides and their concentrations. In principle, the so called main radionuclides should be the long lived nuclides shown in Table A–11.

Radioactive level	Type of radioactive waste					
	Ι	II	II	IV		
Low	$\begin{array}{l} A \leq 4 \times 10^6 \ \mathrm{Bq/kg} \\ \tau_{1/2} \leq 60 \ \mathrm{d} \end{array}$	$A \le 4 \times 10^6 \text{ Bq/kg}$ 60 d < $\tau_{1/2} \le 5 \text{ a}$	$\begin{array}{l} A \leq 4 \times 10^6 \; \mathrm{Bq/kg} \\ 5 \; \mathrm{a} < \tau_{1/2} \leq 30 \; \mathrm{a} \end{array}$	$A \le 4 \times 10^6 \text{ Bq/kg}$ $ au_{1/2} > 30 \text{ a}$		
Intermediate	$A>4\times 10^6~{\rm Bq/kg}$ $\tau_{\rm 1/2}{\leq}60~{\rm d}$	$A>4\times 10^6~{\rm Bq/kg}$ 60 d $<\tau_{\rm 1/2}$ ≤ 5 a	$\begin{aligned} 4\times 10^6 \ \mathrm{Bq/kg} &< A \leq 4\times 10^{11} \ \mathrm{Bq/kg} \\ 5 \ \mathrm{a} &< \tau_{\mathrm{1/2}} \leq 30 \ \mathrm{a} \\ Q &\leq 2 \ \mathrm{kW/m^3} \end{aligned}$	$A > 4 \times 10^{6} \text{ Bq/kg}$ $\tau_{1/2} > 30 \text{ a}$ $Q \le 2 \text{ kW/m}^{3}$		
High			$A > 4 \times 10^{11} \text{ Bq/kg}$ $Q > 2 \text{ kW/m}^3$	$A > 4 \times 10^{10} \text{ Bq/kg}$ $Q > 2 \text{ kW/m}^3$		
Alpha				$A > 4 \times 10^{6}$ Bq/kg (single package) $A > 4 \times 10^{5}$ Bq/kg (multipackage)		

TABLE A-10. CLASSIFICATION OF CHINA'S SOLID RADIOACTIVE WASTES

TABLE A–11. MAIN RADIONUCLIDES TO BE MEASURED IN LOW AND INTERMEDIATE LEVEL WASTE

Type of radionuclide	Low and intermediate level waste		
CP/AP nuclides	C-14, ⁶³ Ni, ⁹⁴ Nb, ⁹⁹ Tc, ⁶⁰ Co		
FP nuclides	Sr-90, ¹²⁹ I, ¹³⁷ Cs		
Alpha emitting nuclides	Total alpha emitting nuclides		

A-4.3. Scaling factor method in the measurement of radionuclides in different wastes

The SF method has not yet been set up in China to quantify DTM nuclides in waste package to be disposed of at the disposal sites. However, this method has been used for measurement of the total beta activity of high level liquid waste (HLLW).

During the fission process of uranium and plutonium, large amounts of FPs and actinides are generated. Most of these radionuclides are beta emitting nuclides. It is necessary to measure the total beta activity of different feedstocks in the reprocessing process and the waste treatment. However, the number of beta emitting nuclides is large and the beta energy spectrums of the nuclides overlap. Therefore, it is very difficult to measure the radioactivity of each beta emitting nuclide for the purpose of measuring the total beta radioactivity. Hence, the SF method is appropriate.

A-4.3.1. Principle of the scaling factor method

There are about 20 beta emitting nuclides in HLLW. Based on the relationship between the measuring efficiency and the maximum beta energy, the beta emitting nuclides are divided into four groups. Each group has its own efficiency radionuclide and the activity weighting radionuclide as displayed in Table A–12. The total beta radioactivity is calculated by the following equations:

 $A' = N/\kappa$

$$\kappa = \sum \in W_i$$

where

- A' is the total beta activity in the sample in Bq;
- N is the count of the sample in \min^{-1} ;
- κ_i is the measuring efficiency of group i, i = 1-4;
- W_i is the activity weighting factor.

The radioactivity of the activity weighting radionuclides was measured with Ge(Li) gamma spectrometers and liquid scintillation. Based on the fission yield of ²³⁵U and the SF between the radionuclides, the relative radioactivity of the other radionuclides can be calculated. With the relative radioactivity of the other radionuclides in each group, the activity weighting factor of each group will be calculated. Thus the total beta radioactivity of the HLLW is known by measuring the radioactivity of several activity weighting radionuclides, such as 106 Rh, 155 Eu, 137 Cs and 144 Ce.

TABLE A–12. MAJOR BETA NUCLIDES AND THE INDICATING RADIONUCLIDES IN HIGH LEVEL LIQUID WASTE

Group No.	E _β /kev	Main nuclides	Efficiency nuclide	Activity weighting nuclide
1	<100	Ru-106	H-3	Rh-106
2	100-500	Tc-99, ¹²⁵ Sb, ¹²⁹ I, ¹⁴⁴ Ce, ¹⁴⁷ Pm, ¹⁵⁵ Eu	C-14	Eu-155
3	500-1000	Sr-90, ¹³⁴ Cs, ¹³⁷ Cs, ¹⁵⁴ Eu	Sr-90	Cs-137
4	>1000	Y-90, ¹⁰⁶ Rh, ¹⁴⁴ Pr	Y-90	Ce-144

A-5. FRANCE

A-5.1. Introduction

There are 58 PWR units in operation in France: 34×900 MW(e), 20×1300 MW(e) and 4×1500 MW(e). Since 1992, LILW arising during operation of these nuclear power plants has been disposed of at the LILW (short lived) Aube Centre, and since 2003 VLLW (new classification) has been disposed of at the VLLW Morvilliers Centre. A summary of the main waste streams, their treatment methods and disposal routes is shown in Fig. A–3.

In LILW waste disposal, acceptable activity or activity concentration limits are regulated for individual radionuclides and groups of radionuclides according to the conditions of safety assessment scenarios (e.g. intrusion after a survey period of 300 years post-shut down). Particularly, the inventory limits and the maximum concentrations limits of long lived nuclides (¹⁴C, ⁶³Ni, ⁹⁴Nb, ^{108m}Ag, etc., alpha emitters with half-life >30 years) are low. Most of these radionuclides are DTM, and the SF method has been applied since the 1990s.

Firstly, over the period 1992 to 1995, Electricité de France (EDF), with the agreement of ANDRA (the National Agency for Radioactive Waste Management, in charge of the two disposals), implemented the SFs given by the international literature to declare in operational waste six DTM nuclides: ¹⁴C, ⁶³Ni, ⁹⁴Nb, with ⁶⁰Co as the key nuclide, and ⁹⁰Sr, ⁹⁹Tc, ¹²⁹I, with ¹³⁷Cs. The same SFs were applied for all types of waste.

However, following a request by the French safety authorities, EDF in cooperation with the Commissariat à l'énergie atomique (CEA, owner of hot and radiochemistry specific laboratories) launched in 1989 a large campaign of sampling and radiochemical analysis on primary coolants, water filters and active ion exchange resins from ten different reactors.

In 1995, five of the six previous SFs were replaced by those obtained from the statistical treatment of about 500 different results (SFs for 55 Fe, 59 Ni and some alpha emitters were also determined, while the SF for 99 Tc/ 137 Cs was recalculated.

It is interesting to note that the new SFs were not so different, except SF 129 L/ 137 Cs, which moved from 1.5×10^{-4} to 10^{-6} , with a positive consequence on the iodine inventory of packages already disposed of.

A second wave of sampling (during the period 1995–1999) confirmed the SFs (with populations of data increasing) and determined the SF of 99 Tc/ 137 Cs.



FIG. A-3. Summary of waste treatment, conditioning and disposal.

Since 1999, EDF has declared in packages produced during operation 18 pure beta and e emitters, ⁵⁵Fe, ⁹⁰Sr and ²⁴¹Pu (b). However, 12 of these 21 emitters are still declared on the base of SFs from the literature.

The goal for EDF, with the CEA's cooperation, is to develop a complete radiochemical analysis procedures panel to only implement SFs obtained on French reactor waste. Hence the next sampling campaigns concerned essentially ion exchange resins. At present, only the radiochemical determination for ⁴¹Ca and ¹⁵¹Sm are missing.

In parallel, SFs for alpha emitters (²³⁸⁺²³⁹⁺²⁴⁰⁺²⁴¹Pu, ²⁴¹Am, ²⁴³⁺²⁴⁴Cm) were also determined. However, they are not currently used (see below).

In conclusion, in addition to about 300 results obtained on primary coolant samples coming from ten different reactors, more than 1000 different results above the detection limits (see below) obtained on water filters (>400) and on ion exchange resin (>600) samples have been treated.

These results include those obtained during additional destructive control of about 40 packages, managed by ANDRA.

A-5.2. Radionuclides of interest

The SF method has been applied to DTM nuclides, as shown in Table A–13. In the first step, key nuclides were selected based on only similarity of production mechanism, essentially FPs or CPs. In the second step, depending of the strength of correlations with ¹³⁷Cs and ⁶⁰Co, the better correlation of the two could be chosen over the default one. This has been applied only to ⁹⁰Sr (an FP nuclide), which is best correlated to ⁶⁰Co (a CP nuclide) for water filters.

A-5.2.1. Comments about alpha emitters declaration

It has been demonstrated that alpha radioactivity, when present, is due to very tiny particles transported in the primary coolant; because of this, the behaviour of alpha emitters (deposit or trapping on filters or fixing on ion exchange resins) is very similar to that of ⁶⁰Co. This has been confirmed by the strength of correlations obtained for alpha emitters with this key nuclide.

Alpha emitters are declared only in the event of serious fuel failures (gross alpha activity > 4 Bq/L). Despite the existence of SFs for each alpha emitter, the declaration procedure is based on knowledge of the SF gross alpha/⁶⁰Co of the primary coolant obtained by easy and repetitive measurements. This SF (considered as an envelope) is extended to all waste streams of the reactor. Finally, for the declaration of the waste's activity, gross alpha is derived in elementary alpha emitters with predefined conservative spectra depending on the type of fuel (UO₂ or MOX). Normally, with the agreement of ANDRA, alpha emitters are not declared in waste arising during operation.

Type of radionuclide	Homogenous solidified waste (active ion exchanger resins)		Heterogeneous solidified waste (water filter cartridges)		
	Evaluated DTM nuclide	Key nuclide	Evaluated DTM nuclide	Key nuclide	
CP nuclides	Be-10, ¹⁴ C, ¹³⁶ C, ⁴¹ Ca, ⁵⁵ Fe, ⁵⁹ Ni, ⁶³ Ni, ⁹³ Mo, ⁹³ Zr, ⁹⁴ Nb, ^{108m} Ag	Co-60	Be-10, ¹⁴ C, ³⁶ Cl, ⁴¹ Ca, ⁵⁵ Fe, ⁵⁹ Ni, ⁶³ Ni, ⁹³ Mo, ⁹³ Zr, ⁹⁴ Nb, ^{108m} Ag	Co-60	
FP nuclides	Se-79, 90 Sr, 99 Tc, 107 Pd, 121m Sn, 126 Sn, 129 I, 135 Cs, 151 Sm, 241 Pu (β)	Cs-137	Se-79, ⁹⁹ Tc, ¹⁰⁷ Pd, ^{121m} Sn, ¹²⁶ Sn, ¹²⁹ I, ¹³⁵ Cs, ¹⁵¹ Sm, ²⁴¹ Pu (β) Sr-90	Cs-137 Co-60	
Alpha emitting nuclides	Total alpha emitting nuclides and ²³⁸⁺²³⁹⁺²⁴⁰⁺²⁴¹ Pu, ²⁴¹ Am, ²⁴³⁺²⁴⁴ Cm	Co-60	Total alpha emitting nuclides and ²³⁸⁺²³⁹⁺²⁴⁰⁺²⁴¹ Pu, ²⁴¹ Am, ²⁴³⁺²⁴⁴ Cm	Co-60	

TABLE A-13. EVALUATED NUCLIDES AND SELECTION OF THE KEY NUCLIDES

TABLE A-14. TABLE A-14. SAMPLING PLAN

	Homogenous solidified waste (active ion exchanger resins)	Heterogeneous solidified waste (water filter cartridges)
Basic sampling plan	Sampling from the maximum of plants during embedding treatment campaigns involving mobile machines Resins are collected in tanks after homogenization by water recirculation (the same tank collects resins from different reactor auxiliary systems, CVCS, LWT, SFPC, etc.)	Sampling from the maximum of plants and on different reactor auxiliary systems. Cartridges are shipped to the CEA's hot laboratories, then samples are taken on the high, medium and low parts of a cartridge
Number of samples	Several tens with a mass of a few grams	Several tens with a mass of a few grams

A-5.3. Sampling and radiochemical analysis

So far, radiochemical analysis data have been collected from more than 1000 samples of homogeneous solidified waste (ion exchange resins) and heterogeneous waste (water filter cartridges) and of primary coolant samples.

Despite the homogeneity of the EDF network (58 reactors of very similar design with the same materials¹, with the same primary coolant chemistry, etc.²) the goal is to perform sampling campaigns involving all reactors.

The basic sampling plan is summarized in Table A–14. It is important to obtain radioactive wastes having a wide range of activity concentrations in order to ensure effective correlations between DTM nuclides and key nuclides for waste from that stream.

For evaluating the representativeness of collected samples, comparisons of the radioactivity concentration range between those declared in concrete containers (ILW) produced by the 58 reactors during 2006 and those of samples that were analysed are shown in Figs A–4 and A–5.

A-5.4. Calculation method

Linear regression (least squares method) was adopted for calculating SFs. This is the result of initial statistical approaches involving the median test, Student's test and Fischer-Snedecor's test. The relationship considered is a straight line. In the computerized development, lines are forced to pass through the origin, as shown in Fig. A–6.

For the best presentation, a log–log diagram was also adopted; pairs of coordinates and the correlation line are imported, and the correlation factor and R^2 are illustrated in Fig. A–7.

A-5.5. Judgement of applicability of the scaling factor method

Firstly, it is important to reject outliers; Grubbs' test could be adopted. It is also pertinent to take into account only 'clear results' (above the detection limits). Secondly, applicability of the SF method given by regression depends on the number of available data (expected ≥ 5), then of the value of R^2 (expected ≥ 0.7).

¹ The replacement of steam generators with the change of 600 alloy tubing into 690 tubing has no consequence on SFs involving 60 Co as the key nuclide. The replacement of zircaloy fuel rods by those enriched in niobium has required EDF to demonstrate the non-consequence of the waste 94 Nb content. This resulted in the need to develop a new radiochemical analysis procedure to get a very low detection limit of 94 Nb (e.g. 0.1 Bq/L⁻¹ in the primary coolant). In turn, with the new European pressurized water reactor, all SFs involving 60 Co will be revaluated due to the elimination of cobalt hard facing alloys.

 $^{^2}$ A zinc injection in the primary coolants of two reactors to analyse the benefit of a lower level of 58,60 Co in the oxides layers could lead to the modification of some SFs.



FIG. A-4. Comparison of activity concentration range for ion exchange resins.



FIG. A-5. Comparison of activity concentration range for water filters.

Water filters : scaling factor ⁶³Ni/⁶⁰Co



FIG. A-6. Example linear regression analysis.





FIG. A–7. Example of SF correlation on log–log plot.

The standard deviation and confidence interval are also calculated, respectively, where:

$$\text{STDV} = \left(\frac{1}{Nbr}\sum Y/X - \overline{Y/X}\right)^{0.5}$$

and the confidence interval is defined as the:

arithmetical mean
$$\pm 1.96 \left(\frac{STDV}{\sqrt{Nbr}} \right)$$

In the event of non-applicability due to $0.5 \le R^2 < 0.7$, the geometrical mean is adopted and Nbr = number of samples:

$$Gm = \sqrt[n]{(y_1/x_1 \cdot y_2/x_2 \cdot \dots \cdot y_n/x_n)}$$

In the event of non-applicability due to insufficient data (Nbr < 5) or due to $R^2 < 0.5$ (no correlation), an arithmetical mean is adopted.

With the agreement of ANDRA, when for one radionuclide the number of data is less than 10 it is possible to add the results of the packages' destructive super-controls, if available.

A-5.6. Classification of the scaling factors

The same SFs are applied in all French PWRs. It has been decided to extend SFs obtained for water filters to other waste streams: DAW concentrates, sludge, oils, etc. Those obtained on active ion exchange resins are extended to very low level ion exchange resins (10 Bq/g) arising from steam generator blowdown systems.

A-5.7. Continuous use of same scaling factors

As mentioned in the examples given above, continuous use of SFs is approved if any subsequent evolution does not occur. At the least, it is possible by specific sampling to demonstrate that an evolution has no impact on SFs if new results given by radiochemical analysis are among those already obtained.

In the normal course of events, an information package for the safety authorities and ANDRA is required. On the other hand, EDF has to confirm a lot of SFs by increasing the population (sampling implemented on a maximum of different reactors). A third of SFs have been determined with fewer than ten data points.

At present, SFs validated in 1999 are applied. Those obtained in 2004 and after are not yet validated. A summary of the 1999 and 2004 data is given in Table A–15.

A-6. GERMANY

A-6.1. Introduction

At present there are eight PWRs and five BWRs in operation in Germany. Owing to an agreement between the Federal Government and the German nuclear power plant operators, a phase-out of nuclear energy is planned within the next 17 years. This is of importance for waste characterization, since motivation of the nuclear power plant operators for changes or improvements to the plants is correspondingly low.

Ion exchange resins				Water filters and others			
Radionuclides	Key	SF 1999	SF 2004	Radionuclides	Key	SF 1999	SF 2004
Be-10	Co-60	2 E-07	1,8 E-05	Be-10	Co-60	2 E-07	1,1 E-06
C-14	Co-60	1,8 E-02	1,5 E-02	C-14	Co-60	1,1 E-02	3,7 E-02
Cl-36	Co-60	1 E-05	1,8 E-05	Cl-36	Co-60	1 E-06	1 E-08
Ca-41	Co-60	5 E-06	5 E-06	Ca-41	Co-60	5 E-06	5 E-06
Fe-55	Co-60	1,4 E-01	2,6 E-01	Fe-55	Co-60	2,1 E+00	2,1 E+00
Ni-59	Co-60	1,1 E-03	1 E-03	Ni-59	Co-60	5,3 E-04	5,4 E-04
Ni-63	Co-60	1,4 E+00	1,5 E+00	Ni-63	Co-60	2,3 E-01	2,2 E-01
Se-79	Cs-137	4 E-06	9,4 E-08	Se-79	Cs-137	4 E-06	9,4 E-08
Sr-90	Cs-137	2,3 E-03	4,8 E-03	Sr-90	Co-60	2,6 E-02	2 E-02
Mo-93	Co-60	1 E-06	2,3 E-06	Mo-93	Co-60	1 E-06	5,1 E-07
Zr-93	Co-60	5 E-07	5 E-07	Zr-93	Co-60	5 E-05	4,3 E-06
Nb-94	Co-60	1,2 E-04	1,7 E-04	Nb-94	Co-60	1,3 E-04	1,2 E-04
Tc-99	Cs-137	1 E-05	1,1 E-05	Tc-99	Cs-137	4,2 E-04	3,1 E-04
Pd-107	Cs-137	1 E-07	1 E-07	Pd-107	Cs-137	1 E-05	1 E-05
Ag-108m	Co-60	1 E-03	1,4 E-03	Ag-108m	Co-60	1 E-03	6,8 E-04
Sn-121m	Cs-137	2 E-05	2 E-05	Sn-121m	Cs-137	2 E-05	2 E-05
Sn-126	Cs-137	9 E-06	1,3 E-08	Sn-126	Cs-137	9 E-06	1,3 E-08
I-129	Cs-137	1 E-06	1 E-06	I-129	Cs-137	1 E-06	1 E-06
Cs-135	Cs-137	5 E-06	5 E-06	Cs-135	Cs-137	3 E-06	5 E-06
Sm-151	Cs-137	7 E-04	7 E-04	Sm-151	Cs-137	4 E-03	4 E-03

TABLE A-15. SUMMARY OF 1994 AND 2004 SCALING FACTORS

Waste disposal for nuclear power plant waste has taken place in Germany for some ten years in the Asse and Morsleben repositories. Both are located in former salt mines. Waste emplacement was stopped in 1978 in the Asse repository and in 1998 in Morsleben. The WAC for the Morsleben facility had been updated to modern standards and were implemented in 1995, so for the final three years new experience was gained for the practical implications of LLW emplacement. Meanwhile, Asse and Morsleben have both applied for a decommissioning licence, and the licensing procedures are under way.

In 2002, the construction and operation licence for the planned Konrad repository was granted after more than 20 years of licensing procedures. After five more years in court, the licence finally came into force at the beginning of 2007. WAC for Konrad were developed throughout the licensing procedure. They, too, became binding with the licence.

Since 1995, either the Morsleben or the Konrad WAC could be chosen by the waste producers. After closure of the Morsleben repository, mainly the Konrad WAC (preliminary at that time) were applied. Quality control procedures for waste conditioning were adjusted accordingly long before the Konrad licence became valid.

The wastes emplaced in Morsleben are mainly evaporator concentrates and mixed solid wastes. In addition, radiation sources and other types of institutional wastes were disposed of. A special feature in Morsleben was the in situ solidification of liquid concentrates with brown coal fly ash, which was practised until 1989.

Wastes intended for the Konrad repository is called waste with negligible heat production. This category can include all kinds of solid or solidified LLW. For short lived wastes with high portions of strong gamma emitters, or comparable, the term ILW could be applied to this type of Konrad waste.

A-6.2. Radionuclides of interest

A-6.2.1. Radiologically relevant radionuclides

Konrad is prepared for any kind of waste that complies with the WAC and is not designed for any special waste stream. Therefore all radionuclides potentially contained in the waste have to be checked and considered for activity declaration.

Table A–16 gives an overview of the radionuclides that have been evaluated as radiologically relevant in the safety analyses. It can be seen that the number is considerable. Not all of these radionuclides can be determined by SF methods; other methods have to be supplemented. Nonetheless, the SF method has proven applicable and valuable for quite a number of radionuclides. Although in the preference hierarchy for application the SF method comes last after direct measurement results and batchwise activity characterization by radionuclide vectors/distributions, its role is still dominant considering the number of radionuclides that are determined by this method.

A-6.2.2. Selection of the key nuclides

The selection of the key nuclides was performed considering the following criteria:

- (a) Strong gamma emitter;
- (b) Appears sufficiently often at measurable levels in waste samples;
- (c) Sufficiently long half-life;
- (d) Representative for either FPs or activated CPs or APs of fuel (alpha emitters);
- (e) Representative for radionuclide generation and transport conditions.

It was soon clear that ⁶⁰Co was the only radionuclide available for activated CPs. Others, such as ¹⁵²Eu, ⁵⁸Co or ¹²⁴Sb, did not fulfil half the criteria listed. For FPs there was no significant choice except ¹³⁷Cs. Alternatives, such as ⁹⁴Nb, fitted more for CPs, but were less frequently present in the waste samples. A separate key nuclide for alpha emitters was considered at the beginning. However, the occurrence of alpha emitters in waste in measurable quantities was rather low, and later on the similarities of most alpha emitters to ⁶⁰Co led to the decision to omit a separate key nuclide for alpha emitters. Nonetheless, today total alpha activity is sometimes used as an auxiliary key nuclide for checking results of correlation calculations.

-				
Ac-227	Cm-244	Ho-166m	Pu-238	Sn-126
Ac-228	Cm-245	I-125	Pu-239	Sr-90
Ag-108m	Cm-246	I-129	Pu-240	Tc-99
Ag-110m	Cm-247	K-40	Pu-241	Th-228
Am-242m	Cm-248	Mn-54	Pu-242	Th-229
Ar-39	Cm-250	Mo-93	Pu-244	Th-230
Be-10	Co-58	Na-22	Ra-224	Th-232
C-14	Co-60	Nb-94	Ra-226	U-232
Ca-41	Cs-134	Nb-95	Ra-228	U-233
Cd-113m	Cs-135	Ni-59	Rb-87	U-234
Ce-141	Cs-137	Ni-63	Ru-103	U-235
Cl-36	Eu-152	Np-236	Ru-106	U-238
Cf-249	Eu-154	Np-237	Sb-124	Zn-65
Cf-251	Fe-55	Pa-231	Sb-125	Zr-93
Cf-252	Fe-59	Pb-210	Se-79	Zr-95
Cf-254	H-3	Pd-107	Sm-151	

TABLE A-16. RADIOLOGICALLY RELEVANT RADIONUCLIDES

Problems with key nuclides have been observed in the declaration of old waste containers. Owing to the relatively short half-life of ⁶⁰Co, a considerable decay of its activity takes place during interim storage. Thus, in some instances, it was not easy to detect ⁶⁰Co in measurable activity concentrations. This means that in cases where long interim storage is foreseeable, the radionuclide specific waste declaration should be prepared as early as possible.

A similar problem arose in the most recent nuclear power plant wastes; because of very stringent measures to avoid buildup of 60 Co, its concentration in the waste was decreasing accordingly. This affected the 60 Co measurements.

A-6.2.3. Selection of wastes

The type of waste for which activity determination by the SF method might be successful was selected in a pragmatic manner. If the determination of SFs with samples from all wastes provided a correlation of the activity concentrations for a pair of radionuclides, then the SF parameters were accepted. The first differentiation was performed for reactor types, because obvious discrepancies were observed there most often. Further differentiations were not considered necessary and were often not possible because of lack of data.

The selection and differentiation for waste streams today sometimes seems to have another primary aim than activity declaration. It is applied rather for a more detailed understanding of the activity flow in a nuclear power plant than for SF applications. This may be very useful but has to be distinguished strictly from activity declaration, otherwise the danger exists that waste streams have to be organized in a manner that complies with the differentiation in evaluation of selected wastes, rather than the opposite way, which orders the evaluation according to the least efforts for activity determination. In practice this might be converted by the authority to a requirement to adjust waste collection to the calculated SF (e.g. separately for plant area, with or without fuel defects) rather than to adjust the evaluation to the highest possible level of data integration.

In addition, experience shows that differentiation because of the unavoidable decrease of data available for evaluation results in a decrease of quality of the correlation. In Germany, therefore, evaluations have been kept on the highest possible level of data integration.

A-6.2.4. Evaluation method

The evaluation of the correlation between two radionuclides is performed by a linear regression of the logarithms of the measurement results. This reflects the log-normal distribution of the measurement data results and leaves two fitting parameters. Mathematically, the following two functions are determined:

$c(\mathrm{RN}_i) = A \times c(\mathrm{KN})^b$	as the expectation value
$c_{\max}(RN_i) = A_{\max} \times c(KN)^b$	as the upper bound

where:

$c(\mathrm{RN}_i)$	is the activity concentration of radiologically relevant radionuclide <i>i</i> ;
$c_{\max}(RN_i)$	is the maximum activity concentration of radiologically relevant radionuclide;
c(KN)	is the activity concentration of key nuclide;
A	is the proportionality constant;
$A_{\rm max}$	is the maximum proportionality constant;
В	is the regression coefficient (is a measure of non-linearity of the evaluated correlation);
r	is the CC (is a measure of the quality of the evaluated correlation).

Figure A–8 demonstrates the meaning of the parameter values in a log–log diagram of the correlation of ^{14}C to $^{60}Co.$



FIG. A-8. Correlation of ¹⁴C to ⁶⁰Co.

A-6.2.5. Acceptable correlations

The linear regression does not provide a meaningful correlation by itself. The CC, which is calculated in parallel, is a statistical indicator of whether a correlation exists or not. A value of 1 or -1 means that the correlation is ideal, while a value of 0 indicates that no correlation exists. To accept a correlation, a CC of better than 0.7 is required. A CC of less than 0.7 but better than 0.5 will be accepted if it is demonstrated that no systematic difference is responsible for the poor quality.

A second criterion that is applied for selection of an acceptable correlation is the ratio A_{max}/A . If the ratio is higher than 100 it will be assumed that the data scatter is too high to form a representative correlation. In practice, the first criterion in most cases was more selective than the second one.

Both criteria are based on empirical observations rather than statistical evidence; because of the numerous factors besides statistics that influence the quality of a correlation, this approach seems more realistic.

A-6.3. Sampling and radiochemical analysis data

Sampling meanwhile is performed in a regular manner with two additional samples per year for each plant. Table A–17 gives an overview of the available German data sets. Data from other countries are sometimes used for checking results where only a small number of German data are available.

To ensure that radiochemical analyses have the quality that is expected for the establishment of a usable correlation, Germany (by the Jülich Research Centre) took part in an international intercomparison test together with seven other laboratories. Two samples, resins and concentrates, from a German PWR distributed by the Jülich Research Centre were measured by the participants. The analyses to be performed were:

Туре	Concentrates	Filter sludges	Resins	Solids	Total
PWR	485	11	125	243	864
BWR	419	247	104	5	775
Total	904	248	229	248	1.639

TABLE A-17. NUMBER OF SAMPLES COLLECTED BY WASTE TYPE

- (a) α or γ spectrometric measurement directly on the sample upon reception (optional).
- (b) α or γ spectrometric measurement on the solubilization solutions obtained from the mineralizations of the samples.
- (c) Two mineralizations per sample, two analyses per solution.
- (d) Determination of the following radionuclides: gamma emitters, ¹⁴C, ⁵⁹Ni, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am, additional radionuclides optional.

The results were to be reported in Bq/g for resins and Bq/mL for concentrates.

The adjusted results for resins are shown in Table A–18. The results show a remarkable consistency. The scatter of the measurements in relation to the average values is highest for ^{239/240}Pu, owing to a low sensitivity measurement by CEN Cadarache. The rest of the results exhibit a scatter that is well within the range of uncertainty, which is to be expected for this type of measurement.

A-6.4. Establishing confidence

In order to demonstrate the capabilities of the SF method beyond activity determination, an investigation of the influence of MOX fuel on the alpha activity concentrations in waste was performed. MOX fuel elements have been used in routine operation in German nuclear power plants since 1986. Experience is available mainly from PWRs. As the MOX fuel elements contain transuranic elements from the very beginning, an increase might be expected in the radionuclide inventory of higher transuranic elements (e.g. americium, curium) and the heavier plutonium isotopes (e.g. ²⁴²Pu, ²⁴⁴Pu) in waste from nuclear power plants that use MOX fuel. This increase should have an influence on the aforementioned correlation of the elements to the key nuclides.

The influence of MOX fuel might be superimposed by other effects such as fuel defects or outer contamination of fuel elements. Primary information about these parameters is not readily available. However, these parameters may be related to the age of the plant. To avoid age dependent effects in the evaluation, three generations of German nuclear power plants were investigated separately.

Radionuclide	CEN Cadarache	CIEMAT (ENRESA)	Hungarian laboratories	KFA Jülich	SCK/CEN Mol	Average
C-14	3200 ^a	3790	2500	3200	4040	3350
Co-60	340 000	312 000	307 000	293 000	327 000	316 000
Ni-59	45 ^a	<470	84			65
Ni-63	43 000 ^a	37 200		17 000	40 000	34 300
Nb-94	<100	<40.7		<50		<40.7
Tc-99		<76.7	1	<66		1
I-129		<9.1	0.21	< 0.5		0.21
Cs-137	320 000	304 000	322 000	300 000	309 000	311 000
U-234				< 0.2	0.0298	0.0298
U-235				< 0.2	0.000933	0.000933
U-238				< 0.2	0.0984	0.0984
Pu-238	16 ^b	11.7	11.9	13		13.2
Pu-239/240	25	8.92	7.78	9		12.7
Pu-241		373		400		387
Am-241	23 ^b	20	16.8	19		19.7

TABLE A–18. COMPARISON OF ADJUSTED MEASUREMENT RESULTS ON RESINS (IN Bq/g OF WET RESIN)

^a Derived from original data.

^b Distribution between ²³⁸Pu and ²⁴¹Am according to average values.

As Fig. A–9 shows, the activity of transuranic elements is lower in waste from younger nuclear power plants than from the older ones. This holds true irrespective of the type of fuel used. The primary explanation of this effect is an obviously decreasing contamination of the outer surface of the fuel cladding for nuclear power plants of later generations. In some cases, fuel defects in the early years of operation may be an additional reason.

Comparison of Figs A–9 and A–10 shows that this fact superimposes the effect of MOX fuel elements. Since the difference between operation with and without MOX is in the range of the inherent scatter of data, the changes in activity concentration of transuranic elements due to MOX fuel are not significantly above the detection limit.

As the activity concentration of ⁶⁰Co decreases from older to younger nuclear power plants to nearly the same extent as the activity concentration of the transuranics, the correlations of the transuranic elements to ⁶⁰Co remain essentially unchanged.



FIG. A–9. Activity concentrations of some transuranic elements and ⁶⁰Co in waste from German PWRs of different age using MOX fuel.



FIG. A–10. Effect of MOX fuel usage on the activity concentration of transuranics and ⁶⁰Co in waste streams from second generation German PWRs.

A-7. HUNGARY

A-7.1. Introduction

There are four PWR units in the only nuclear power plant in Hungary, at Paks. Different types of LILW are generated during the operation of WWER type reactor units. The main waste streams are as follows:

(a) DAW:

(i) Compacted miscellaneous solid waste;

(ii) Untreated solid waste (non-compactable).

(b) Liquid wastes:

(i) Evaporator concentrates;

(ii) Ion exchange resins.

(c) Sludge.

Conditioning of liquid waste has not yet started. Currently, evaporator concentrates and ion exchange resins are stored on the site of the power plant, and the treatment and conditioning of liquid waste will commence when the LILW repository is operational. Currently, the construction of the repository at Bátaapáti is under way. The first shipment is to be carried out in the middle of 2008.

On the basis of the safety assessment results, the site specific WAC were derived for the Bátaapáti repository. One of the main elements of the WAC is the limit for the (isotope specific) activity content of the waste. There are no prescriptions in the Hungarian regulations either for the list of isotopes whose activity content shall be determined for the waste packages or for the methodology of the determination. However, it has to be mentioned that the defined WAC were officially approved by the competent authority, so indirectly the list of isotopes for which activity content shall be determined for the waste packages was also approved.

For the determination of the isotope content of solid waste packages, segmented gamma spectrometry (SGS) is applied for the gamma emitters, and the SF methodology is used for the DTM nuclides. For the calculation of the SFs, a few hundred samples have so far been taken from the tanks of the evaporator concentrates and the tanks of the ion exchange resins. The radiochemical analysis of the samples is carried out in different institutions.



FIG. A-11. Hungarian system for radioactive waste characterization.

7.2. Radionuclides of interest

Since the Hungarian regulations are not prescriptive, the radionuclides of concern were selected on the basis of international practice and were later confirmed by a safety assessment. The SF method was applied for the determination of the activity of the DTM nuclide part of this list of isotopes (shown in Table A–19).

Generally it can be stated that ⁶⁰Co and ¹³⁷Cs isotopes are used as the key nuclides for the determination of the DTM nuclides' activity content of the package. For the uranium isotopes, ²³⁸Pu is applied as an auxiliary key nuclide. For the calculation of ²³⁸Pu activity, ⁶⁰Co is used as the key nuclide.

The second unit of Paks nuclear power plant suffered a serious incident in 2003, in which 30 fuel assemblies were damaged in a cleaning tank emplaced in an auxiliary vault of the nuclear power plant. (The reactor was not in operation when the incident happened.) Due to this incident, the water system of the second unit became much more contaminated than it used to be. Different SFs have had to be determined for those waste packages involved in this incident ('incidental' waste). For this purpose, sampling of the tanks containing the evaporator concentrates from the second unit — and radiochemical analysis of the samples — was carried out.. For the activity calculation of some DTM nuclides in the incidental waste, 144 Ce is applied as a key nuclide.

A-7.3. Sampling and radiochemical analysis

In Paks nuclear power plant, the evaporator concentrates and ion exchange resins are separately stored in tanks. Approximately ten samples are taken every year from the different layers of the evaporator concentrates' tanks and a few samples from the ion exchange resins' tank. The radiochemical analysis is carried out for each DTM nuclide in each sample and the key nuclide's activity content is also measured. The chemical preparation and the measurement methods are summarized in Table A–20.

DTM	Key nuclides	Key nuclide for 'incidental' waste
H-3	Cs-137	Cs-137
C-14	Co-60	Co-60
C-136	Cs-137	Cs-137
Ca-41	Cs-137	Cs-137
Fe-55	Co-60	Co-60
Ni-59	Co-60	Co-60
Ni-63	Co-60	Co-60
Sr-90	Cs-137	Cs-137
Tc-99	Co-60	Co-60
I-129	Cs-137	Cs-137
U-234	Pu-238	Ce-144
U-235	Pu-238	Pu-238
U-238	Pu-238	Ce-144
Pu-238	Co-60	Ce-144
Pu-239+240	Co-60	Ce-144
Am-241	Co-60	Ce-144
Cm-242	n.d. ^a	Ce-144
Cm-244	Co-60	Ce-144

TABLE A-19. SUMMARY OF DIFFICULT TO MEASURE NUCLIDES AND KEY NUCLIDES

^a n.d.: no data.

Institute Radionuclide		Preparation and measurement
Institute of Nuclear Research of the Hungarian Academy	C-14	Precipitation as BaCO ₃ Proportional gas counting
of Sciences	Н-3	Vacuum distillation LSC
	Tc-99	TEVA SPEC column LSC
	I-129	Precipitation ICP MS or LSC
	Cs-135	Liquid extraction ICP MS
	Cl-36	Precipitation as AgCl LSC
Budapest University of Technology and Economics	Transuranic nuclide (^{238, 239+240, 241} Pu, ²⁴¹ Am, ²⁴² Cm)	Anion exchange, electrodeposition Alpha spectrometry
	Uranium isotopes	Alpha spectrometry
	Sr-90	Sr spec column Low level beta counting
	Fe-55, ⁵⁹ Ni, ⁴¹ Ca	Si(Li) Roentgen spectrometry (XRF)
	⁶³ Ni	XRF corrected beta counting
Paks nuclear power plant	Gamma emitters	SGS

TABLE A-20. SUMMARY OF THE PREPARATION AND MEASUREMENT METHODS

A-7.4. Validation of the scaling factor method

The calculation method applied in Hungary is described in Section A–7.5. For determining the two parameters (m, b) used as SFs, the linear regression of the logarithms method is used. The applicability of the SF method for DTM nuclides and key nuclide pairs is checked by the following ways:

- (a) The linear relationship between the logarithms has been checked by calculating and evaluating the CC and the coefficient of determination;
- (b) A *t* test and *F* test statistical method (at a confidence level of 95%) has been applied for checking the existence of a correlation.

A-7.5. Calculation method

The following relationship has been assumed between the activity content of the DTM nuclide and the activity content of the key nuclide in the waste:

$$c_{\rm dmn} = b'(c_{\rm kn})^m \tag{A-1}$$

where:

 c_{dmn} is the activity concentration of the DTM nuclide; c_{kn} is the activity concentration of the key nuclide; b', m are parameters. After taking the base 10 logarithms of the two sides of Eq. (A-1), we get the following expression:

$$\log(c_{\rm dmn}) = \log(b') + m \log(c_{\rm kn}) \tag{A-2}$$

or:

$$\log(c_{\rm dmn}) = b + m \log(c_{\rm kn}) \tag{A-3}$$

where

 $b = \log(b')$

The linear regression of the logarithms method has been used for determining the two parameters (m, b) used as SFs on the basis of Eq. (A-3).

A-7.6. Data maintenance

A few hundred samples have been evaluated during the operation of the nuclear power plant. The samples are taken each year on the basis of the nuclear power plant's sampling programme; this way the data pairs for the activity content of each DTM nuclide and the corresponding key nuclide have been accumulating. Due to this fact, every year a new linear fitting is performed and a review of SFs is carried out.

Since there was neither significant change in the components of the reactor materials nor fuel failure, the SFs have not changed significantly. Only the incident at the nuclear power plant caused significant change in the SFs for the so called 'incidental' wastes, as described earlier.

A-8. INDIA

A-8.1. Introduction

There are 16 nuclear power reactors currently operating in India, with a combined generating capacity of 3900 MW(e). Six more units are in various stages of construction, building an additional capacity of 2800 MW(e). At present, seven reactor sites are in use; an NSDF is located at each of these reactor sites for the safe and efficient management of radioactive waste arising from the reactors. Waste streams include spent resins fixed in a polymer matrix, sludges from low level liquid waste treatment fixed in cement, and DAW, including high efficiency particulate air filters and other assorted waste.

While contributing towards the social growth and overall progress of human civilization, all industrial activities inevitably generate some kind of undesired waste products, which is also true for the nuclear industry. As waste from nuclear facilities contains small quantities of radioactive material, its safe and effective management has been given utmost importance from the very inception of the nuclear industry, with the primary objective of protecting the human environment in the long term. Safe and effective management of these low level radioactive wastes requires understanding of the nature and the quantities of the radionuclides present in the packaged waste. The risks associated with the transport and disposal of radioactive waste are based on an assessment of the waste packages containing the radionuclides.

Knowledge of radioactivity content in terms of the concentrations of specific radionuclides plays a vital role during disposal of waste packages. Some of the important long lived nuclides present in radioactive waste are DTM nuclides, determined from outside the waste packages using non-intrusive techniques because they are low energy beta or alpha emitting nuclides (i.e. non-gamma emitters).

A-8.2. Radionuclides of interest

Radionuclides important for the safety case for the management and disposal of assorted low level solid radioactive wastes are listed in Table A–21.

Series. No.	Isotope	Half-life (a)	Туре
1	Cs-137	30.2	Beta, gamma
2	Sr-90	28	Beta
3	I-129	1.6×10^6	Beta, gamma
4	Co-60	5.27	Beta, gamma
5	Cs-134	2.06	Beta, gamma
6	Pu-239	24 360	Alpha

TABLE A-21. RADIONUCLIDES IN LOW LEVEL RADIOACTIVE SOLID WASTE^a

^a Preliminary list drawn primarily from international experience. Engineering efforts to develop and expand the list specifically to India's requirements are under way.

TABLE A-22. CATEGORIZATION OF RADIOACTIVE WASTE

<u> </u>	Solid	Liquid	Gaseous
Category	Surface dose rate (mGy/h)	Activity level (Bq/m ³)	Activity level (Bq/m ³)
I	<2	$<3.7 \times 10^{4}$	<3.7
II	2–20	$3.7 \times 10^4 - 3.7 \times 10^7$	$3.7 - 3.7 \times 10^4$
III	>20	$3.7 \times 10^7 - 3.7 \times 10^9$	$>3.7 \times 10^{4}$
IV	Alpha bearing	$3.7 \times 10^9 - 3.7 \times 10^{14}$	_
V		$>3.7 \times 10^{14}$	_

A-8.3. Waste management philosophy

The overall objective of nuclear waste management is to ensure the adequate safety and welfare of the public by effective isolation of radionuclides from the human environment. The Indian philosophy of radioactive waste management embodies three basic principles:

- (a) Delay and decay (for short lived radionuclides);
- (b) Dilute and disperse (for very low active effluents);
- (c) Concentrate and contain (intermediate and high active waste).

A-8.4. Waste classification

The waste classification system depends upon the local disposal conditions and the methodologies followed for handling and storage. The various steps involved in the management of all kinds of waste are:

- (a) Identification and segregation;
- (b) Reduction and recovery;
- (c) Transport and treatment;
- (d) Disposal and surveillance.

The Indian Atomic Energy Regulatory Board has categorized these wastes in accordance with internationally acceptable norms and standards, as shown in Table A–22.

A-8.5. Basic steps in radioactive waste management

The various steps of the management of radioactive waste as adopted in India include:

- (a) Waste characterization;
- (b) Treatment/conditioning;
- (c) Storage;
- (d) Disposal;
- (e) Surveillance/monitoring, etc.

The descriptions presented herein are intended to be general and to apply to the management of radioactive waste arising from mining and milling, fuel fabrication, nuclear power generation, back end activities and medical and industrial applications of radioactive material.

A-8.5.1. Near surface disposal facilities

As a national policy, each nuclear site in India has its own NSDF collocated. There are seven NSDFs currently operational within the country, which have to address widely varied geological and climatological conditions. The various disposal modules currently adopted are listed in Table A–23.

These modules are generally below ground; however, depending upon the local geohydrological conditions, these could be partly or completely above ground. The salient features of the disposal modules are briefly described below.

Stone lined earth trenches: These are employed for potentially active wastes (category I packages having a surface dose rate less than 2 mGy/h). As indicated in Fig. A–12, these are shallow excavations in soil, 1–4 m deep, provided with stone lining for stability and integrity. On completion of the disposal operations, these trenches are backfilled and closed by providing a soil cover of nearly one metre thickness. Vermiculite, bentonite and native soil having good sorption properties are used as backfill materials.

Reinforced concrete trenches: These are employed for category II and III wastes having a surface dose rate of 2–20 mGy/h and 20–500 mGy/h, respectively. These trenches are planned zone-wise and are modular in construction. A typical trench is 4.8 m deep, 2.5 m wide and 15 m long, as depicted in Fig. A–13. The outer containment wall thickness varies from 350 mm at the top to 750 mm at the bottom. On completion of the filling operations, these trenches are closed with pre-cast concrete slabs, which also provide the necessary shielding. Adequate sealing and waterproofing is provided subsequent to closure of the battery.

Category	Surface dose/activity	Disposal options	Nature of waste
Ι	<2 mGy/h	Stone lined earth trenches	Paper trash, concrete chips, cotton mops, rubber items, etc.
ΙΙ	2–20 mGy/h	Reinforced concrete trenches	Contaminated equipment, hardware and filters
III	20-500 mGy/h	Reinforced concrete trenches	Conditioned/processed concentrates, sludges, spent resins
	>500 mGy/h	Tile holes	Hardware from reactors, highly contaminated equipment, conditioned spent resins, etc.
IV	Waste bearing alpha activity (<4000 Bq/g) (>4000 Bq/g)	Reinforced concrete trench and tile holes Tile holes	Solidified alpha waste with beta–gamma activity

TABLE A-23. SOLID WASTE CATEGORIES AND DISPOSAL OPTIONS



FIG. A-12. Stone lined earth trenches.



FIG. A-13. Construction details of reinforced concrete trenches.

Tile holes: Tile holes are used for disposal of category IV waste and also special waste packages of category III that do not qualify for disposal in reinforced concrete trenches. Waste packages in 200 L standard epoxy painted mild steel drums containing conditioned waste with more than 4000 Bq/g of alpha activity and/or surface contact dose rate above 500 mGy/h due to beta–gamma activity are retrievably stored in these tile holes. They are circular vaults nearly 4 m below ground level having an average inside diameter of 710 mm. They are made of a 6 mm thick carbon steel shell with a 25 mm thick concrete lining on both sides and provided with adequate waterproofing. Typical construction details are shown in Fig. A–14.

A-8.5.2. Interim storage of vitrified waste

A storage and surveillance facility collocated with the vitrification facilities is operational at Tarapur. The facility consists of an underground outer vault that houses two inner thermal vaults. Removal of decay heat is achieved by natural convective ventilation induced by a 100 m high stack. The air cooling system has been designed on the basis of storage unit geometry, array design, filling pattern and stack dimension. A schematic of the air cooled solid, storage and surveillance facility is shown in Fig. A–15.



FIG. A-14. Construction details of tile holes.



FIG. A-15. Schematic of air cooled solid, storage and surveillance facility.

A-8.5.3. Assay of the waste package: present status

Primary efforts have been initiated to measure the radioactivity contents in the waste packages. In recent years, these procedures have expanded into the measurement of radionuclides in the waste packages, which need to be quantified before disposal or retrievable storage. The primary problem in accurately assaying gamma emitting nuclides in waste material is to apply a correction factor for the typically unknown waste matrix and source configuration. For small containers with low density waste, these corrections are minor and easily handled. However, as the volumes and types of wastes that must be assayed have increased, the typical container sizes have also accordingly increased. At the same time, regulatory requirements demand more accurate measurements at even lower levels.

A-8.5.3.1. Imaging of waste packages

The imaging technique as shown in Fig. A–16 is useful to segregate compactable and non-compactable waste drums, and is able to differentiate between metal, wood, glass and other objects present. This is a non-destructive/non-intrusive technique involving radiographic inspection using real time radiography by direct measurement. Images can be evaluated and the suitability of the drum under inspection can be decided instantly. Images can be subjected to image enhancement and analysis, for improving the detectability features. An X ray source with a voltage range of 100–200 KV is utilized in this technique. An image intensifier tube and charge



FIG. A-16. Imaging system in operation at the SWMP facility, Trombay.



FIG. A-17. Waste assaying system operational at SWMF, Trombay.

coupled device camera are coupled to the output screen of an image intensifier tube. For ease of operation, an automated PLC based mechanical handling and movement system with safety interlocks and a man-machine interface for displaying the system status and fault messages have been provided.

A-8.5.3.2. Assaying of waste packages

The assaying technique as shown in Fig. A–17 is used for assessment of radioactivity using gamma ray spectrometry in which bulk waste monitors are installed. Spectral data are interpreted qualitatively and quantitatively for determining the activity. Assessment of gamma activity content of ¹³⁷Cs and ⁶⁰Co in 200 L waste drums as a function of weight is carried out. Cs-137 (gamma energy 662 KeV) and ⁶⁰Co (gamma energy 1173, 1332 KeV) isotopes are extensively used as the key nuclides for this purpose.

The assay system comprises a mechanized drum handling facility, where a waste drum is placed on the weighing scale platform, which rotates during monitoring at two to five revolutions per minute. The detector assemblies consist of a multi-channel analyser with an NaI (Tl) detector assembly housed in a lead shield.

A-8.5.3.3. Need for scaling factor method

Estimation of DTM nuclides present in the waste packages necessitates separation of radionuclides by complex chemical analytical techniques. This method is not practicable for large numbers of waste packages containing a wide range of radionuclides. Techniques have been demonstrated for measurement of the ETM

nuclides such as ¹³⁷Cs and ⁶⁰Co in the waste packages. Further study will focus on developing models for correlation of the estimated ETM nuclides and DTM nuclides in terms of SFs.

Measurement/estimation of the concentrations of all relevant radionuclides either directly or indirectly by the application of SFs will be useful in guiding the establishment and deployment of a sound waste disposal strategy.

A-9. ITALY

A-9.1. Introduction

Four nuclear power plants (Garigliano, Latina, Trino and Caorso) were operated until the middle of the 1980s in Italy. At present they are at different stages in the process of being decommissioned, in accordance with a strategy for immediate decommissioning (IAEA level 3) established in the late 1990s. The spent fuel and the largest part of the radioactive waste to be managed in Italy derive from the operation of the above mentioned nuclear power plants and from a few fuel cycle facilities (see Fig. A–18 for the location of nuclear power plants and other facilities).

The legislative and regulatory framework established since the early 1960s envisages a system of licensing of nuclear installations and activities as well as regulatory control. This system fully applies to spent fuel and radioactive waste management activities.

The national operator entitled to perform spent fuel, radioactive waste and decommissioning activities is SOGIN (Società Gestione Impianti Nucleari), a company whose shareholder is the Ministry of Economy and Finance, while the strategic and operational aims are provided by the Ministry of Productive Activities.

As far as the radioactive waste is concerned, almost all the waste generated by the operation of nuclear installations is stored at the sites of origin.



FIG. A-18. Location of nuclear power plants and other facilities in Italy.

A common practice in the nuclear power industry is to use the SFs for estimating the concentrations of DTM nuclides in waste packages being prepared for disposal. Typically, a nuclear power plant periodically samples a variety of LLW types and submits them to analytical laboratories for radionuclide analyses. Then the plant develops activity SFs for the DTM nuclides (pure beta emitters, low energy photon emitters (X rays) and alpha emitters) relative to the most abundant long lived gamma emitting nuclides present in the waste, usually ⁶⁰Co and ¹³⁷Cs, which are easily measured by gamma ray spectrometry. A database of SFs is then generated for the various sample types and used in conjunction with gamma ray spectrometry to quantify the radionuclides and determine their waste classification.

Prominent LLW streams currently included in characterization programmes include process resins from radioactive waste treatment and DAW. Waste streams specific to decommissioning have not been addressed at this time.

A-9.2. Radionuclides of interest

A formal determination of radionuclides of concern has not been completed. Efforts are proceeding to examine alternatives for characterization evaluations.

A number of DTM nuclides lend themselves well to this activity SF methodology, particularly neutron APs (e.g. ⁵⁵Fe, ⁶³Ni, ⁵⁹Ni), which have similar production modes and similar chemical behaviour in the reactor coolant and related waste streams compared with ⁶⁰Co.

A-9.3. Background historical information on the national nuclear programme

Commercial utilization of nuclear power in Italy started in 1964 and by 1981 four nuclear power plants, namely the nuclear power plants of Garigliano (BWR), Latina (gas graphite), Trino (PWR) and Caorso (BWR), and a low enriched uranium fuel fabrication installation (Fabbricazioni Nucleari SpA) had been commissioned.

During that period an extensive research and development programme on the nuclear fuel cycle was developed by the Nuclear Energy Research Agency (CNEN) - now the National Agency for New Technology, Energy and the Environment (ENEA) - with the operation of experimental fuel cycle installations (e.g. ITREC and EUREX).

The three nuclear power plants, Latina, Trino and Caorso, continued to be operated until 1987, when they were definitely shut down based on a governmental decision resulting from a national referendum called after the Chernobyl accident. The Garigliano nuclear power plant was shut down in 1978 for technical reasons.

At the same time that the nuclear programme was closed, the Interministerial Committee for Economical Planning (CIPE) required the National Electricity Company (ENEL SpA) to start the decommissioning of the nuclear power plants, and a safe storage (IAEA level 1/2) option was adopted.

In 1999, all ENEL SpA liabilities and assets connected to nuclear power were assigned to a newly established company, SOGIN SpA, whose shareholder is the Ministry of Economy and Finance, while the strategic and operational aims are provided by the Ministry of Productive Activities. The primary mission of SOGIN SpA is to cover the decommissioning of all Italian nuclear installations and the safe management of the spent fuel and radioactive waste.

The spent fuel and the largest part of the radioactive waste to be managed in Italy derive from the operation of the above mentioned nuclear power plants and fuel cycle facilities. As far as spent fuel is concerned, part of this has already been transferred abroad for reprocessing (namely the Latina fuel and part of the fuel of the Garigliano and Trino nuclear power plants). It is envisaged that treated and conditioned waste resulting from the reprocessing will be returned to Italy.

A-9.4. Italian approach to the scaling factor

Among the several applicable methodologies for the correlation of DTM nuclide activities and reference key radionuclide activities (SF determination), the Italian national regulatory body (APAT) refers to the procedures contained in a National Technical Guide named UNI 11194, Radiological Characterization of Category 2 Packages for the Purpose of Disposal in the Final Repository, which employs the geometric mean

and the dispersion associated with the distribution of the quantity $(A_{\rm HTM}/A_{\rm K})_{\rm I}$ as the most efficient formalism in the statistical analysis of experimental data.

Considering the National Technical Guide UNI 11194, the SF determination is based on empirical considerations. In particular cases, as for example some nuclear power plant components, the SFs can be extracted through computational codes (e.g. SF-STAT, a computer code for deriving SFs for DTM nuclides in LLW streams). For the DTM radionuclides coming from APs, a correlation with ⁶⁰Co is generally used, while for the DTM radionuclides coming from FPs, a correlation with ¹³⁷Cs is used. Regarding the transuranic nuclides, a correlation with ¹⁴⁴Ce can be found (despite its very short half-life).

A–9.4.1. Method of analysis (scaling factor determination)

The procedure provided by the UNI 11194 standard is considered for the determination of the SFs either in the case of radionuclide inventory in radioactive waste or in the case of radionuclide content verification for clearance of solid materials.

The SF determination between a generic DTM radionuclide (x) and a reference key radionuclide (k), representative of a specific group of radiologically homogeneous material, can be articulated in the following steps:

- (a) Subdivision of systems and installation structures into groups (including different components and materials) for which homogeneous conditions are supposed based on installation history, from data extracted during the operative period and specific radiological measurements.
- (b) Sampling an adequate number of N samples representative of the materials constituting each one of the preceding homogeneous groups. Generally $N \ge 20$ is chosen; a number of samples lower than 20 can be accepted exclusively in the case of materials or systems of narrow variability or for which all available information (engineering considerations, historical, experimental measurements) are in agreement with homogeneous radiological conditions.
- (c) N sample radiochemical analysis and measurement, for each of them, of the specific activity associated with the DTM nuclide of interest (a_{xi}) and of the specific activity (a_{ki}) associated with the reference key radionuclide with which it is intended to correlate
- (d) Calculation, for each of the analysed N samples, of the ratio $(FC)_i = a_{xi}/a_{ki}$ for experimental SFs.
- (e) Geometric mean calculation (A_{FC}) of the N experimental correlation factors.
- (f) Dispersion calculation $(D_{\rm FC})$ based on the geometric mean $A_{\rm FC}$, of the *N* SFs; the $D_{\rm FC}$ definition comes from the standard deviation meaning, applied to the logarithmic measurement distribution (FC)_i. Given a value distribution $\ln({\rm FC})_i$, the probability that a new determination for $\ln({\rm FC})$ gives a result belonging to the range $\ln(A_{\rm FC}) - \ln(D_{\rm FC}) \le \ln({\rm FC}) \le \ln(A_{\rm FC}) + \ln(D_{\rm FC})$ is 68.3%. Analogue probability, in the corresponding measurement distribution of (FC)_i, results associated with a new FC measurement whose value is in the range $A_{\rm FC}/D_{\rm FC} \le {\rm FC} \le A_{\rm FC} \cdot D_{\rm FC}$. If $2\sigma = 2 \ln(D_{\rm FC})$, to the dispersion of distribution value $\ln({\rm FC})_i$ is associated, when there will be a new $\ln({\rm FC})_i$ determination, a probability of 95.5% to assume a value in the range $\pm 2 \ln(D_{\rm FC})$ (or in a factor of D_{FC}^2 , for (FC)_i measurements) from the logarithm of the geometric mean $A_{\rm FC}$.
- (g) verify that the value assumed by the 2σ dispersion of the measurement distribution $(FC)_I$ will be lower than the limit of 6 $(D_{FC}^2 \le 6)$. If this criteria is satisfied, it can be said that the number of analysed samples is statistically firm and that the geometric mean A_{FC} constitutes a confidential estimation of the radionuclide x SF, representative of the whole materials constituting the examined homogeneous group. If the 2σ dispersion of the measurement distribution $(FC)_i$ assumes a value greater than 8, a valid SF cannot be defined and the radionuclide x activity will be experimentally set with different methodologies:
 - (i) Evaluations based on the analysis of an adequate number of samples coming from different systems;
 - (ii) Whatever conservative criteria.

If the D_{FC}^2 is in the range between 6 and 8, it is necessary to increase the number of examined samples (up to a maximum of 30) and to make a further analysis, including of data coming from the new samples. If $D_{FC}^2 < 8$, the correlation is established, otherwise the correlation is definitely not accepted.

- (h) If point (g) is positive, the measurements that have not given values in the 2σ distribution limits are rejected.
- (i) $A_{\rm FC}$ assignment for the specified homogeneous group, specifying the SF reference date.

Since the $(FC)_i$ measurement distribution is log-normal, the geometric mean calculation gives the evaluation of the distribution having the maximum confidence.

If the number of measurements with values greater than the MDA for SF determination of a specific radionuclide was insufficient, this lack of data will have to be conservative based on justified and documented considerations.

The following properties are considered in the assessments: the nature of the contaminant fluid (primary cooling, ventilation system particulate, etc.), modes of contamination, relevant operating conditions for the contamination processes (temperature, material typology, present radionuclides) and the operating history. The evaluation of the SF will be done based on measurements of contamination vectors or specific mathematical computer based calculations.

A-9.5. Data maintenance

A-9.5.1. Caorso nuclear power plant example 1: Turbine, annex turbine and off-gas scaling factor determination

As an example, the following explains the technical procedure used in the Caorso nuclear power plant for the turbine, annex turbine and off-gas radiological characterization for decommissioning purposes, and, in our case, for the SF determination.

In this project the SF determination has been used for the clearance of different solid material (metals or bulk insulators).

The main objective of radiological characterization is to give the specific activities, expressed either as surface contamination (Bq/cm^2) or mass contamination (Bq/g), of the DTM radioisotopes present in the components and systems of the building. The preceding procedure permits the collection of the total activity values and their application to the plant components and, mainly, the definition of the SFs, giving the isotopic contribution to the total activity referred to a specific isotope (60 Co, in this case).

Inside the turbine and annex turbine buildings the radioactivity is present mainly as contamination deposited on the surface components, expressed as Bq/cm².

A-9.5.1.1. Measurement typologies

Data are available from the following measurements:

- (a) Direct beta-gamma contamination measurements;
- (b) Radiochemical measurements made on surface oxide samples taken from the inner surface of components;
- (c) Exposure measurements;
- (d) Gamma spectrometry.

During the period 1994–2000, approximately 140 measurements were made.

A-9.5.1.2. Results

The isotopes present in the contamination are ⁶⁰Co, ¹³⁷Cs, ¹²⁵Sb, ⁵⁹Ni, ⁶³Ni and ⁵⁵Fe. No alpha contamination is present (all values are lower than the MDA). The main radioisotope present in the power plant components and systems is ⁶⁰Co, except for the gaseous effluents treatment system, where the main radioisotope is ¹³⁷Cs.

Regarding the ¹²⁵Sb radioisotope, its presence has been revealed just for a few systems, while the presence of ⁵⁹Ni, ⁶³Ni and ⁵⁵Fe radioisotopes has been revealed only in the case of consistently removable contamination.

The ¹³⁷Cs SF changes consistently system by system, while ⁵⁹Ni, ⁶³Ni and ⁵⁵Fe have quite uniform values for the whole power plant installation.

For contaminated systems, Table A-24 shows:

System code	System name	Sb-125/60Co	Fe-55/60Co	Ni-59 + ⁶³ Ni/ ⁶⁰ Co
N11	Main steam system low pressure and high pressure	37	2.5	7
N21	Condensate feedwater system	1.1	2.5	7
N22	Steam relief valves	n.a.	2.5	7
N31	Main turbine	150	2.5	7
N33	Main steam isolation valve	n.a.	2.5	7
N36	Extraction steam	70	2.5	7
N37	Turbine bypass	n.a.	2.5	7
N38	Reheater	n.a.	2.5	7
N61	Condenser and auxiliary	12	2.5	7
N62	Off-gas effluents	n.a.	2.5	7
P61B	Blanketing	3	2.5	7

TABLE A–24. SUMMARY OF SCALING FACTORS FOR CAORSO NUCLEAR POWER PLANT, TURBINE, ANNEX TURBINE AND OFF-GAS

n.a.: Co-60 activity is lower than the MDA.

(a) System code present in the project;

(b) System name;

(c) The SFs for related radioisotopes.

The ⁶⁰Co value of the measured contamination ranges from 0.005 to 29.47 Bq/cm².

A–9.5.2. Caorso nuclear power plant example 2: Radiological characterization of low activity waste packages containing sludge and resins

A sampling campaign on low activity waste packages containing sludge but no conditioned resins was conducted at Caorso nuclear power plant (BWR) during 1999–2000. The sampling campaign was adopted to give the radiological situation of the waste produced by the different water cycle treatment systems and of radioactive waste. [[change okay? or please clarify]]

The main scope of the sampling campaign was to:

- (a) Verify the existence of the packages' homogeneous groups, in terms of isotopic composition of the present radionuclides;
- (b) Define, for each homogeneous group, the SFs;
- (c) Detect or definitely exclude the probable presence of alpha emitting radioisotopes.

A total of 139 samples of resins have been taken and classified in homogeneous groups, based on their origin, while 42 samples of sludge have been obtained from drumsfor the production years 1978–1989).

All samples were extracted through radioactive logging and the obtained material was put in 250 cm³ or 100 cm³ containers. The samples were then dried and submitted to gamma spectroscopy. A limited number of resin samples (31 samples) were dissolved for further gamma spectroscopy and for determining the DTM nuclides: ⁵⁵Fe, ⁵⁹Ni, ⁶³Ni and ⁹⁰Sr. Of the preceding 31 samples, five were analysed, through alpha spectroscopy, for alpha emitting nuclide determination.

The reference chosen gamma emitters were:

- (a) Co-60, to which were correlated 59 Ni, 63 Ni and 55 Fe;
- (b) Cs-137, to which was correlated 90 Sr.

The SFs were calculated based on gamma spectroscopy, and radiochemical measurements were implemented on the 31 resin samples, selected so as to adequately represent the several resin types, differentiated by origin and year of production.

The geometric mean of measurements represents a reliable evaluation of applicable SFs of the presented procedure.

It is interesting to make a comparison among the obtained results and those from two important scientific publications [A–1, A–2] on the radioactive waste SFs extracted from the average of a number of data coming from many BWR nuclear power plants.

The comparison terms are presented in Table A–25, in which all the activities are reported, in the case of Caorso nuclear power plant for 1 January 2006, and at 20 years decay time for literature data. The result obtained is a good agreement between Caorso data and those in the literature, and in all cases the Caorso data are conservative with respect to those in the literature.

Five selected resin samples were subjected to alpha spectroscopy measurements for verifying the alpha emitting nuclides present: ²³⁸Pu, ²⁴⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴²Cm and ²⁴⁴Cm.

For all examined samples, the alpha emitter activity concentration of transuranic radioisotopes was found to be lower than the instrumentation MDA, with MDA values not greater than 6.4×10^{-3} Bq/g (Table A–26). Considering that the resins were used for filtering process fluids, these results confirm the absence of alpha emitters in the power plant contamination.

TABLE A–25. CAORSO NUCLEAR POWER PLANT RESIN SCALING FACTORS AND LITERATURE DATA COMPARISON

Type of scaling factor	EPRI TR-109448 [A-2] BWR industry, 20 years decay time	NUREG/CR-4101 [A–1] BWR, correlation ratio B1, 20 years decay time	Geometric mean, Caorso scaling factors (1 January 2006)
Fe-55/ ⁶⁰ Co	5.14×10^{-2}	4.58×10^{-2}	5.97×10^{-2}
Ni-59/ ⁶⁰ Co	—	5.82×10^{-1}	1.14×10^{-1}
Ni-63/ ⁶⁰ Co	3.02×10^{-1}	$6.88 imes 10^{-1}$	8.59×10^{-1}
Sr-90/ ¹³⁷ Cs	5.84×10^{-3}	5.94×10^{-2}	6.78×10^{-3}

TABLE A-26. SAMPLE SPECIFIC ACTIVITY (ALPHA SPECTROSCOPY)

Sampla ando	Pu-239 (Bq/g)	$Pu-239 + {}^{240}Pu (Bq/g)$	Pu-241 (Bq/g)	Am-241 (Bq/g)	Cm-244 (Bq/g)	Cm-242 (Bq/g)
Sample code	Alpha	Alpha	Beta	Alpha	Alpha	Alpha
MA009225	$< 4.5 \times 10^{-3}$	$< 6.4 \times 10^{-3}$	<3.1	$< 1.3 \times 10^{-3}$	$< 1.0 \times 10^{-3}$	$< 1.3 \times 10^{-3}$
MA015046	$< 4.5 \times 10^{-3}$	$< 6.4 \times 10^{-3}$	<3.1	$< 1.3 \times 10^{-3}$	$< 1.0 \times 10^{-3}$	$< 1.3 \times 10^{-3}$
MA009196	$< 4.5 \times 10^{-3}$	$< 6.4 \times 10^{-3}$	<3.1	$< 1.3 \times 10^{-3}$	$< 1.0 \times 10^{-3}$	$< 1.3 \times 10^{-3}$
MA010220	$< 4.5 \times 10^{-3}$	$< 6.4 \times 10^{-3}$	<3.1	$< 1.3 \times 10^{-3}$	$< 1.0 \times 10^{-3}$	$< 1.3 \times 10^{-3}$
MA010907	$<4.5 \times 10^{-3}$	$< 6.4 \times 10^{-3}$	<3.1	$<1.3 \times 10^{-3}$	${<}1.0\times10^{-3}$	$< 1.3 \times 10^{-3}$

A-10. JAPAN

A-10.1. Introduction

There are 33 BWR units and 22 PWR units in Japan. Low level radioactive waste generated from these nuclear power plants has been disposed of in the Rokkasho LLW disposal centre since 1992 (see Fig. A–19).

In LLW disposal, acceptable activity or activity concentration limits are regulated for individual radionuclides and groups of radionuclides according to the conditions of each disposal site. Examples of regulated radionuclides include ¹⁴C, ⁶³Ni, alpha emitters, which are long lived, and DTM nuclides. The SF method is applied for determining the activity of these radionuclides.

To evaluate SFs between DTM nuclides and key nuclides, activity concentration data of radioactive waste on DTM nuclides have been accumulated through a radiochemical analysis of more than one thousand samples of homogeneous solidified waste and DAW, including:

- (a) Evaporator concentrates;
- (b) Spent resins (low activity);
- (c) DAW (including filter cartridges from PWRs).

SF values are determined based on radiochemical analysis data. Additionally, the gamma emitting nuclides, ⁶⁰Co and ¹³⁷Cs were selected as key nuclides because of their detectability when measured by non-destructive radioactivity measuring equipment. This radioactivity measurement equipment uses a germanium semiconductor detector as the main device.

These key nuclides are selected for the DTM nuclides considering the similarity of the production mechanisms at present. For waste packages, these key nuclides are measured by the radioactivity measurement equipment. When the key nuclide concentration is below the detection limit, the detection limit itself is regarded as the key nuclide concentration.

Additionally, radionuclide data have been statistically analysed considering classification by the physicochemical behaviour of the radionuclides, the plant's fuel failure history and plant component materials. Based on the radiochemical analysis data, generic SFs for DTM nuclides (such as ⁶³Ni, ⁹⁰Sr and total alpha emitting nuclides) have been determined for each plant group.

A-10.2. Radionuclides of interest

The SF method has been applied to the DTM nuclides shown in Table A–27. In the first step, key nuclides are selected based on only similarity of production mechanism.



FIG. A-19. Waste packages intended for disposal at the LLW disposal centre.

Type of radionuclide	Homogenous solidified waste (concentrates and low activity spent resin)		DAW (include filter card	tridges)
	Evaluated DTM nuclide	Key nuclide	Evaluated DTM nuclide	Key nuclide
CP nuclides	C-14 (PWR), ⁶³ Ni, ⁹⁴ Nb	Co-60	C-14, ⁶³ Ni, ⁹⁴ Nb, ⁹⁹ Tc	Co-60
FP nuclides	Sr-90, ¹²⁹ I	Cs-137	Sr-90, ¹²⁹ I	Cs-137
Alpha emitting nuclides	Total alpha emitting nuclides	Cs-137	Total alpha emitting nuclides	Cs-137

TABLE A-27. EVALUATED RADIONUCLIDES AND SELECTION OF THE KEY NUCLIDES

TABLE A-28. SAMPLING PLAN AND RESULT

	Homogeneous solidified waste (concentrates and low activity spent resin)	DAW
Basic sampling plan	Sampling from each plant and each type of solidification system Collecting samples in consideration of the time series after operation of each plant	Sampling from each plant Collecting samples in consideration of the origins of contamination and types of contamination
Number of samples	Several hundred	Several hundred

Two principal points are considered in selecting a key nuclide from ⁶⁰Co or ¹³⁷Cs in Japan:

- (a) Similarity to the DTM nuclide in terms of production source;
- (b) Having observable correlation with the DTM nuclide in radiochemical analysis data.

Cs-137 is selected as a key nuclide for FP nuclides and alpha emitting nuclides, because both originate in the fuel and correlation between them is observed. In the case of solidified DAW packages, however, ¹³⁷Cs is affected by Compton scattering of ⁶⁰Co and there is a limit to the detection of ¹³⁷Cs by the radioactivity measurement unit. This results in overestimating the radioactivity of FP nuclides and total alpha emitting nuclides. With special attention given to total alpha whose radioactivity is most apt to be overestimated, the alternative of using ⁶⁰Co as a key nuclide for total alpha emitting nuclides was recently evaluated from the point of view of the similarity of transport phenomena in the nuclear power plant.

A-10.3. Sampling and radiochemical analysis

Radiochemical analysis data were collected from more than one thousand samples of homogeneous solidified wastes and DAW based on the basic sampling plan shown in Table A–28.

It is important to obtain radioactive waste having a wide range of activity concentrations in order to ensure effective correlations between the DTM nuclides and the key nuclides for waste from that stream.

For evaluating the representativeness of collected samples, a comparison of the radioactivity concentration range between radiochemical analysis data of DTM nuclides and batch sampling data of ⁶⁰Co, which were analysed when concentrates were solidified, is shown in Fig. A–20.

A-10.4. Judgement of applicability of the scaling factor method

The applicability of the SF method for DTM nuclides and key nuclide pairs is checked by both of the following ways:

- (a) Consideration has to be given to their production mechanism, physicochemical behaviour and observation of their correlation diagrams;
- (b) The *t* test statistical method has been applied for checking the existence of a correlation.



FIG. A-20. Comparison of activity concentration range.

$$t_{S} = r \times \sqrt{\frac{n-2}{1-r^{2}}}$$
$$t_{\alpha\%} = t \left(n-2, \frac{\alpha}{100} \right)$$

where:

- *r* is the CC;
- *n* is the number of data;
- α is the error rate.

A-10.5. Calculation method

Based on the requirements of the regulator, the arithmetical mean was adopted for the calculation method of SFs. However, the radioactive concentrations of DTM nuclides and key nuclides show a log-normal distribution having a correlation between both radionuclides (i.e. a bivariate log-normal distribution); because of this, the geometrical mean is considered to be the preferred method of the Japanese nuclear industry for calculation of SFs.

A-10.6. Classification of the scaling factor

Generic SFs are applied, unless there is a significant plant difference, as summarized in Table A-29.

The integration or classification of the plant group or waste streams for SF is checked by both of the following ways:

- (a) Consideration has to be given to their production mechanism, physicochemical behaviour and observation of their correlation and distribution diagrams;
- (b) A statistical method of analysis of variance for average has been applied for checking the significance of difference among averages.

Radionuclide	Homogeneous solidified waste	DAW	
		Generated process (reactor process, turbine system and radioactive waste treatment system)	
Other CP nuclides	Type of reactor (BWR, PWR) Improvements of reactor component materials (BWR: two plant groups)	Type of reactor (BWR, PWR) Improvements of reactor component materials (BWR: two plant groups)	
FP nuclides and alpha emitting nuclides	Fuel failure history (BWR: three plant groups)	Fuel failure history (BWR: three plant groups)	

TABLE A-29. CLASSIFICATION AND INTEGRATION OF SCALING FACTORS

A-10.7. Continuous use of the same scaling factor

To verify continuous usage of the same SFs for newly generated waste, annual sampling has been carried out every year in every plant. Usually, an annual sample is made by compositing from several batches of concentrates. The condition under which continuous use of the same SF can be applied is that the newly measured data does not exceed ten times the value calculated by the previous SF.

However, the ratio of the CP nuclide and key nuclides does not change without a large scale replacement of reactor component materials. The ratio of the FP nuclide and key nuclides does not change without fuel failure. This means that the possibility of change of the SF can be understood by considering these items.

A-11. REPUBLIC OF KOREA

A-11.1. Introduction

There are 16 PWR units and four CANDU units in operation in the Republic of Korea. LILW generated from nuclear power plants is stored at temporary storage facilities at each reactor site (four sites). In November 2005, the Gyeongju site was successfully selected for the final repository for LILW, which will be constructed as an underground repository (tunnel and vault type) with a capacity of 100 000 drums in the first phase. The Gyeongju repository will be put in operation by 2009 or 2010.

For the disposal of LILW, the acceptable activity or activity concentration limits are regulated for individual radionuclides and groups of radionuclides according to the conditions of the disposal site. In order to comply with the disposal limits for the relevant regulatory standard – MOST Notice No. 2005-18 (acceptance criteria for low and intermediate level radioactive waste) – for the disposal of radioactive waste, the licensee has to identify the major radionuclides in solid waste as follows: ³H, ¹⁴C, ⁵⁵Fe, ⁵⁸Co, ⁵⁹Ni, ⁶⁰Co, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ¹³⁴Cs, ¹³⁷Cs, ¹⁴⁴Ce and alpha emitters (gross alpha). Many of these are long lived and DTM nuclides. An indirect method, such as the SF method, can be applied to determine the activity of the above radionuclides for solid waste.

To evaluate the SFs between DTM nuclides and key nuclides, activity concentration data of radioactive waste on DTM nuclides have been accumulated through a radiochemical analysis of several hundred samples collected from DAW, spent resin, spent filters, evaporator concentrates, etc.

SF values are determined by statistical calculation based on the radiochemical analysis data obtained. To determine the SF values, the gamma emitting nuclides, ⁶⁰Co and ¹³⁷Cs were selected as the key nuclides because of easily detectable characteristics when measured by non-destructive radioactivity measuring equipment. HPGe gamma spectrometers (TGS or SGS) were introduced as the main device to measure radioactivity of solid waste drums.

A-11.2. Radionuclides of interest

The SF method has been applied to DTM nuclides, as shown in Table A–30. In the first step, key nuclides are selected based on similarity of production mechanism.

In the Republic of Korea, the following aspects are considered in selecting a key nuclide from ⁶⁰Co or ¹³⁷Cs:

- (a) Similar to the DTM nuclide in terms of production source;
- (b) Having an observable correlation with the DTM nuclide in radiochemical analysis data.

Cs-137 is selected as a key nuclide for FP nuclides, because it originates from fission of nuclear fuel and distinctive correlation between ¹³⁷Cs and other FP nuclides is observed. In the same way, ⁶⁰Co is chosen as a key nuclide for CP nuclides. For alpha emitting nuclides, however, ⁶⁰Co shows a better correlation.

A-11.3. Sampling and radiochemical analysis

The radioactivity concentration data were collected by radiochemical analysis of several hundred samples for each waste stream, such as homogeneous solidified waste and DAW, spent resin, spent filters, sludge and concentrated evaporator bottoms, for reactor types based on the sampling plan shown in Table A–31.

It is important to collect the samples from radioactive waste with a wide range of activity concentration levels to ensure effective correlations between DTM nuclides and key nuclides for each waste stream.

In the Republic of Korea, the licensee suggests the SF depending on each waste stream and reactor type at each licence amendment. Through licence review for SFs, a revision was required for the suggested SF through additional sampling and measurement by lowering the MDA for radioactivity measurement to include additional valid data. By 2008, around 400 additional samples will be collected for radioactivity analysis to establish the SF for each specific reactor, rather than for a generic reactor type.

Two of mulido	Waste (all waste streams, PWRs, CANDUs)		
Type of nuclide	Evaluated DTM nuclides	Key nuclide	
AP/CP nuclides	H-3, ¹⁴ C, ⁵⁵ Fe, ⁵⁹ Ni, ⁶³ Ni, ⁹⁴ Nb	Co-60	
FP nuclides	Sr-90, ⁹⁹ Tc, ¹²⁹ I	Cs-137	
Alpha emitting nuclides	Total alpha emitting nuclides	Co-60	

TABLE 30. SUMMARY OF DIFFICULT TO MEASURE AND KEY NUCLIDES

TABLE A-31. SUMMARY OF SAMPLING PLAN

	Current	Future (~2008)
Basic sampling plan	Sampling from each plant and each type (Determination of SF for reactor type (PWR, CANDU)) Collecting samples in currently occurring waste (not including old waste)	Sampling from each plant and each type. (Determination of SF for each plant) Collecting samples in currently occurring waste (not including old waste)
Number of samples	Several hundred	Several hundred

A-11.4. Validation of the scaling factor method

The applicability of the SF method for DTM nuclides and key nuclide pairs is checked by the following ways.

- (a) Consideration has to be given to their production mechanism, physicochemical behaviour and observation of their correlation diagrams;
- (b) A statistical method has been applied for checking the existence of a correlation.

A-11.5. Calculation method

The radioactive concentrations of DTM nuclides and key nuclides show a log-normal distribution having a correlation between both radionuclides; because of this, the geometrical mean is introduced as the applicable calculation method to identify the SF. Although a specific method to develop SFs is not stated in the regulatory standard, the expected accuracy should be stated, and the value of the SF should be established conservatively (MOST Notice No. 2005-18).

A-11.6. Other considerations

In the Republic of Korea, the licensee developed the SF for each reactor type (PWR, CANDU), since there was not enough time to collect enough samples to identify the SF for each reactor. In addition, some of the collected samples were not used for the development of SFs because they had radioactivities less than the MDA for key nuclides or less than the MDA for target DTM nuclides.

After enough data and related experience are collected and conservativeness is ensured, specific SFs that reflect the grouping concept (reactor type or waste steam) can be considered.

A-11.7. Data maintenance

According to MOST Notice No. 2005-18, the licence needs to verify the adequacy of the SF at least once every two years or in the event of incidents in which significant changes are made to the waste stream contents (e.g. large replacement of reactor component materials, fuel failure) by periodical sampling and radiochemical analysis for each waste stream.

A-12. LITHUANIA

A-12.1. Introduction

Ignalina nuclear power plant is Lithuania's only nuclear power plant. Two Soviet designed RBMK-1500 type reactors (water cooled graphite moderated channel type reactors) are installed at Ignalina nuclear power plant. These reactors are different from those operating in the Russian Federation.

The first reactor (belonging to Unit 1) was commissioned in 1983, the second (belonging to Unit 2) in 1987. Unit 1 was finally shut down on 31 December 2004, and Unit 2 must be shut down by the end of 2009 in accordance with the Lithuania admission protocol to the European Union.

At the moment there are no radioactive waste repositories in Lithuania that could be licensed according to the requirements established by the Lithuanian authorities. The radioactive waste repositories created in previous years are now treated as storage facilities.

It is estimated that Ignalina nuclear power plant operation and decommissioning will generate approximately 5900 m³ of spent resins, perlite and sediments and about 130 000 m³ of DAW. It is planned to dispose of untreated short lived VLLW in a 60 000 m³ capacity landfill facility. Short lived LILW will be cemented and temporarily stored on the site until a near surface repository with a capacity of 100 000 m³ is constructed. Long lived LILW, as well as HLW, will be stored until the construction of a DGR. The volume of bituminized waste following the conditioning will make up 17 000 m³ (this will remain in the existing bituminized waste vaults that were transformed into the repository). After decontamination of the emptied Ignalina nuclear power plant facilities, final dismantling of the building structures will additionally generate some 965 000 m³ of concrete and 190 000 t of steel to be free released.

Activities on radiological characterization of waste for disposal purposes (requiring declaration of specific radionuclides) started in 2003. The Lithuanian Waste Management Agency (RATA) determined the preliminary limits of activity values for cemented LILW to be disposed of in the near surface repository, which were derived on the basis of a safety assessment for generic construction of the repository. The limits will be adjusted after a disposal site is selected. At the moment, there are limits for 20 radionuclides (³H, ¹⁴C, ³⁶Cl, ⁵⁹Ni, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ¹³⁷Cs, ²²⁶Ra, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu and ²⁴¹Am).

Since then, Ignalina nuclear power plant has provided, and the State Nuclear Energy Safety Inspectorate (VATESI) and the Agency of Environment Protection (AAA) have approved, the preliminary lists of the radionuclides to be declared in:

- (a) Ignalina nuclear power plant operational dry active VLLW to be stored in the existing landfill on the power plant site (⁵⁴Mn, ⁵⁵Fe, ⁶⁰Co, ⁶⁵Zn, ⁹⁰Sr, ⁹³Zr, ^{93m}Nb, ⁹⁴Nb, ^{110m}Ag, ¹³⁴Cs, ¹³⁷Cs, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴⁴Cm);
- (b) Cemented LILW to be disposed of in the near surface repository (³H, ¹⁴C, ⁵⁹Ni, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ¹³⁷Cs, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am);
- (c) Bituminized LILW to be stored in the existing vaults of the storage facility on the Ignalina nuclear power plant site (¹⁴C, ⁵⁹Ni, ⁶³Ni, ⁹⁰Sr, ⁹⁴Nb, ⁹⁹Tc, ¹²⁹I, ¹³⁷Cs, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am);
- (d) Ignalina nuclear power plant dry active VLLW and LILW from dismantling of the equipment in the turbine hall of Unit 1 (¹⁴C, ⁵⁴Mn, ⁵⁵Fe, ⁶⁰Co, ⁵⁹Ni, ⁶³Ni, ⁶⁵Zn, ⁹⁰Sr, ⁹³Zr, ^{93m}Nb, ⁹⁴Nb, ⁹⁹Tc, ^{110m}Ag, ¹²⁹I, ¹³⁴Cs, ¹³⁷Cs, ²³⁴U, ²³⁵U, ²³⁸U, ²³⁷Np, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴⁴Cm).

The lists will be finalized after specific WAC are issued in accordance with the conditions of each disposal site.

SFs between DTM nuclides, included in the above mentioned lists, and key nuclides have also been approved.

For the DTM nuclides, ⁶⁰Co and ¹³⁷Cs are selected as the key nuclides because of their detectability by nondestructive radioactivity measuring equipment and because of good correlation of their activity with the activity of radionuclides of corrosion origin as well as with the activity of FPs. It means that the transport mechanism and pathway of the radionuclide have a greater influence on the radionuclide concentration in the waste of the RBMK-1500 reactor than its production mechanisms.

To evaluate SFs between the DTM nuclides and the key nuclides, data on activity concentrations of the DTM nuclides in different waste streams have been accumulated through a radiochemical analysis of samples taken from:

- (i) The reactor coolant;
- (ii) Water in storage pools for spent nuclear fuel;
- (iii) Water in skips for the storage of damaged spent nuclear fuel;
- (iv) Liquid waste in storage tanks;
- (v) Evaporator concentrates;
- (vi) Bituminized waste;
- (vii) Spent resin;
- (viii) DAW.

Owing to the difficulty and expense of measurement of the low activity of some radionuclides (⁵⁹Ni, ^{93m}Nb, ¹²⁹I, individual activity of ²³⁹Pu and ²⁴⁰Pu), evaluation of SFs between DTM nuclides and key nuclides was supported by modelling of nuclear fuel burnup in the RBMK-1500 reactor, evaluation of auxiliary SFs between immeasurable radionuclides and measurable radionuclides having the same or similar chemical properties, and measurement of activity of auxiliary radionuclides.

For example, a ratio of activity of ²³⁹Pu and ²⁴⁰Pu isotopes in spent nuclear fuel with the same burnup as determined from the measurement of activity of the other radionuclides in the waste stream of interest
Type of radionuclide	Homogenous solidified waste (concentrates and low activity spent resins)		DAW (include filter cartridges)	
	Evaluated DTM nuclide	Key nuclide	Evaluated DTM nuclide	Key nuclide
AP/CP nuclides	C-14, ⁵⁹ Ni, ⁶³ Ni, ⁹⁴ Nb	Co-60 or ¹³⁷ Cs ^b	Sr-90, ⁹⁹ Tc, ¹²⁹ I, ¹³⁴ Cs ^a , ¹³⁷ Cs	Co-60 or ¹³⁷ Cs ^b
FP nuclides	Sr-90, ⁹⁹ Tc, ¹²⁹ I, ¹³⁷ Cs	Co-60 or ¹³⁷ Cs ^b	C-14, Mn-54 ^a , ⁵⁵ Fe ^a , ⁵⁹ Ni, ⁶³ Ni, ⁶⁵ Zn ^a , ⁹³ Zr ^a , ⁹³ mNb ^a , ⁹⁴ Nb, ¹¹⁰ Ag ^a	Co-60 or ¹³⁷ Cs ^b
Actinides	U-234, ²³⁵ U, ²³⁸ U, ²³⁷ Np, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴¹ Am, ²⁴⁴ Cm ^a	Co-60 or ¹³⁷ Cs ^b	U-234, ²³⁵ U, ²³⁸ U, ²³⁷ Np, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ²⁴¹ Pu, ²⁴¹ Am, ²⁴⁴ Cm ^a	Co-60 or ¹³⁷ Cs ^b

TABLE A-32. SUMMARY OF THE DIFFICULT TO MEASURE AND KEY NUCLIDES

^a Only in VLLW.

^b Cs-137 is used as a key nuclide for the waste streams in which the activity of ⁶⁰Co is much lower than the activity of ¹³⁷Cs.

calculated by the ORIGIN program from the SCALE 5 package code, taking into account decay of radionuclides after removal of the fuel from the reactor, was used to evaluate individual activities of these plutonium isotopes from the measured total activity of both plutonium isotopes.

The calculated ratio of activity of ¹²⁹I and ¹³¹I isotopes in spent nuclear fuel stored in a skip and the measured activity of the ¹³¹I isotope in the water from this skip enabled the determination of the activity of the ¹²⁹I isotope in the skips.

The ratio of activities of the ⁵⁹Ni and ⁶³Ni isotopes resulting from the modelling of their activation processes in the reactor and the measured activity of the ⁶³Ni isotope in the waste was used to determine the activity of the ⁵⁹Ni isotope in the waste.

In some cases when the radionuclide concentration in the waste was below the detection limit, the detection limit itself was regarded as the radionuclide concentration. Such a conservative approach was used to determine the ⁹⁹Tc activity in VLLW arising from the dismantling of equipment.

A-12.2. Radionuclides of interest

The SF method has been applied to the DTM nuclides shown in Table A–32. The selection of the key nuclide is based on:

- (a) The limit of detection by non-destructive measurement equipment in the waste package;
- (b) The decay half-life;
- (c) The observable correlation with the DTM nuclide in radiochemical analysis data;
- (d) The similarity of its and the DTM nuclide transport phenomena in the nuclear power plant.

There is a good correlation between the activity of ⁶⁰Co and ¹³⁷Cs in most Ignalina nuclear power plant waste streams. As a result, either ⁶⁰Co or ¹³⁷Cs can be chosen as a key nuclide for all DTM nuclides.

Co-60 is chosen as a key nuclide for all waste streams in which its activity is not much lower than the activity of ¹³⁷Cs because of its better detectability in the waste by the gamma spectrometers of non-destructive measurement equipment.

A-12.3. Sampling and radiochemical analysis

Radiochemical analysis data were collected from more than 250 samples of homogeneous solidified waste and DAW based on the sampling plan in Table A–33.

	Homogeneous solidified wastes (concentrates and low activity spent resinS)	DAW
Basic sampling plan	Sampling from each type of solidification system	Collecting samples in consideration of the origins of the contamination and the types of contamination
Number of samples	20–50	50-100

TABLE A-33. SUMMARY OF SAMPLING PLAN

To obtain samples with a wide range of activity concentrations in order to ensure effective correlations between DTM nuclides and key nuclides for DAW, samples were taken after screening of the waste stream by portable gamma spectrometers and by measuring the gamma activity of candidate samples in situ.

For increasing the effectiveness of correlations between some DTM nuclides and key nuclides in low activity homogeneous waste, additional samples were taken from the other more active homogeneous waste streams, taking into account the similarity of the contamination of these streams by the considered radio-nuclides.

A-12.4. Validation of the scaling factor method

The applicability of the SF method for the pair of the DTM nuclide and the key nuclide is checked by analysing their production mechanism and their physicochemical behaviour and by observation of their correlation diagrams. CCs for the ratio of activity of the DTM nuclide and the key nuclide as well as confidence bands (based on a confidence level of 0.95) and prediction bands are calculated.

The value of CC is greater than 0.8 in most cases (in a few cases it is between 0.7 and 0.8).

A-12.5. Calculation method

Accumulated data on the radionuclide specific activity in the waste streams are distributed according to a log-normal distribution; this is the basis for using the linear regression of logarithms method to evaluate SFs between the DTM nuclides and the key nuclide.

In a few cases, when there are not enough data on the activity of some radionuclides because of the impossibility of detecting the radionuclides in many samples, the arithmetical mean method has been adopted for the calculation of SFs.

A-12.6. Other considerations

To improve the quality of evaluated SFs, additional analysis of results is done with the intent of rejecting outliers in the results. If the cause of such outliers is not identified, Grubbs' test, as described in ISO 5725-2:1994, is used to decide how to handle the outlier.

Although specific SFs are used for different waste streams, for the determination of the SF for some DTM nuclides in one waste stream previously collected data from another waste stream can be used if analysis shows that the conditions of transport of this radionuclide and the key nuclide are similar for both streams.

A-12.7. Data maintenance

There are no general requirements for periods of sampling to confirm data for existing waste streams. Requirements are set individually for the SFs of every waste stream under consideration. Permission to use the same SF for the waste stream is granted for three to four years, with some additional requirements as a rule. For example, it is established that the activity ratio of ¹³⁷Cs and ⁶⁰Co isotopes shall be controlled continuously in cemented LILW. If the activity ratio of ¹³⁷Cs and ⁶⁰Co isotopes is out of the range 0.25–2.5, new sampling to confirm data for this waste stream is required.

There are also requirements in some cases to recalculate the SF taking into account decay of radionuclides every year. SFs for new waste streams are established after analysis of samples from these waste streams.

A-13. MEXICO

A-13.1. Introduction

There are two facilities in Mexico that could require development of SFs for waste classification:

- (a) Laguna Verde nuclear power plant;
- (b) National Institute of Nuclear Research.

Data from the Laguna Verde plant are used to describe our current experience with SF determination.

Laguna Verde nuclear power plant comprises two 675 MW(e) BWR-5 units from General Electric, with turbine generators supplied by Mitsubishi. Construction started in 1976 in the Laguna Verde area, about 450 km east of Mexico City on the coast of the Gulf of Mexico. Unit 1 started commercial service in July 1990, while Unit 2 started commercial operation in April 1995. As of this date there are no waste disposal facilities in Mexico for final storage, and the Laguna Verde nuclear power plant has decided to temporarily store waste generated as a result of plant operations.

The waste streams generated by Laguna Verde nuclear power plant include spent resins, precoat sludges, cleanup sludges, concentrates, activated charcoal, DAW and other waste.

A-13.2. Radionuclides of interest

One of the most important Mexican Official Standards is NOM-004-NUCL-1994, Radioactive Waste Classification, which is based on US 10CFR 61, part 55, and contains radionuclides and concentration limits c for waste classification purposes, distributed in two tables (see Table A–34 for long lived radionuclides and Table A–35 for short lived radionuclides).

A-13.2.1. Difficult to measure key nuclides

Our expectation was to use ⁶⁰Co, ¹³⁷Cs and ¹⁴⁴Ce to correlate CPs, FPs and transuranic nuclides, respectively. However, samples analysed showed that ⁶⁰Co is the only constant element in wastes; due this fact, all DTM nuclides have been correlated to ⁶⁰Co.

Radionuclide	Concentration (Ci/m ³)
C-14	8
C-14 in activated metal	80
Ni-59 in activated metal	220
Nb-94 in activated metal	0.2
Tc-99	3
I-129	0.08
Transuranic nuclides with half-life greater than five years	100^{a}
Pu-241	3 500 ^a
Cm-242	$20\ 000^{a}$

TABLE A-34. CONCENTRATION LIMITS FOR LONG LIVED RADIONUCLIDES

^a Units are in nCi/g.

		Concentration (Ci/m	3)
Radionuclide	Col. 1	Col. 2	Col. 3
All radionuclides with half-life longer than five years	700	Not limited	Not limited
Н-3	40	Not limited	Not limited
Co-60	700	Not limited	Not limited
Ni-63	3.5	70	700
Ni-63 in activated metal	35	700	7000
Sr-90	0.04	150	7000
Cs-137	1	44	4600

TABLE A-35. CONCENTRATION LIMITS FOR SHORT LIVED RADIONUCLIDES

A-13.2.2. Dry active waste drums classification

The National Institute of Nuclear Research has a system for measuring total activity in drums containing low level radioactive waste (dose rate to activity conversion method); this system is mounted on a mobile platform that can be moved to different places.

A system called the automated waste assay system (see Fig. A–21) comprises basically a drum elevator with a rotating plate at constant speed that holds 55 gallon drums, a high resolution germanium detector in a horizontal configuration, a multichannel analyser, a detector shielding with 10 cm of lead and a transmission radioactive source (152 Eu). The system performs activity measurements at five elevations (segments) of the drum during the rotation. Additionally, for each surveyed segment, four contact dose rate readings are taken at the drum surface using an ion chamber type survey instrument Eberline (RO-20).

A-13.2.3. Exposure rate vs. total activity curves

Based on the average and maximum contact dose rate values obtained by the surveys performed with an ionization type detector (Eberline RO-20) and the total activity for each drum, experimental contact dose rate versus activity curves (Xc vs. A curves) were generated for the average and maximum dose rates, respectively.

A-13.3. Sampling and radiochemical analysis

A-13.3.1. Laboratories for sample analysis

Correct SF determination methods require costly and sophisticated radiochemical analysis in specialized laboratories. At this time it is not feasible to install such kinds of laboratories in countries such as Mexico, since the need for this type of analysis is very limited.



FIG. A-21. Portable automated waste assay system.

Radionuclide	Number
H-3	25
C-14	20
Fe-55	22
Ni-59	4
Ni-63	33
Sr-90	24
Tc-99	5
Pu-239	2
Am-241	3
Pu-241	3

TABLE A–36. DIFFICULT TO MEASURE NUCLIDES IDENTIFIED IN THE 34 SAMPLES

A-13.3.2. Samples analysed in external laboratories

To date, 34 samples have been analysed in specialized laboratories in the USA (see Table A-36):

- (a) Twenty-four samples from Unit 1;
- (b) Ten samples from Unit 2.

A-13.4. Validation of the scaling factor method

To date, we have not defined a definitive set of SFs to be used for the Laguana Verde waste classification. More data analysis is required for the 34 samples sent to an external laboratory to define their applicability.

It is clear that more samples must be analysed for DTM nuclide determination, and we expect to obtain some correlations between ETM nuclides, as ⁶⁰Co or ¹³⁷Cs, and total beta or alpha concentration in the reactor water coolant. Another option is to use data from other BWR reactors to establish some kind of trend or representativeness when analysing combined data between other BWR reactor data and Laguna Verde data.

A-13.5. Calculation method

Weighted log-mean averages with root mean square fractional deviations (root mean square dispersion) determine the characteristic value for each SF in a sample stream.

$$\overline{R} = \exp\{\sum [w_i x \ln(R_i)] / \sum w_i\}$$
$$w_i = 1/\sigma_i^2 = (R_i / \sigma R_i)^2$$
$$\overline{dR} = \exp\{[\sum |\ln \overline{R} - \ln R_i|^2] / (n-1)]^{1/2}\}$$

where

- *R* is the weighted log-mean average;
- w_i is the weighting factor;
- dR is the root mean square deviation.

An outlier test identifies both anomalous values in historical and current data. Scaling factor selection criteria:

- (a) Use the weighted log-mean value.
- (b) Use the most recent value.
- (c) Use the log-mean value or previously determined real value when the current value is an minimum detectable level value.
- (d) Use a previously determined value and resample.
- (e) Use a previously determined value when the scaling nuclide concentration in the new sample is an MDL value.
- (f) Use an SF value characteristic of another waste stream.
- (g) Use an MDL value.
- (h) Use a generic value.
- (i) Use SFs based on the log-mean values for the identified groupings and revise plant procedures for waste stream identification and sampling.

Weighted least squares technique for generic SFs:

$$b_{1} = \left(\sum w_{i}/DENOM\right)\left(\sum w_{i}x_{i}y_{i}\right) - \left(\sum x_{i}w_{i}/DENOM\right)\left(\sum w_{i}y_{i}\right)$$
$$w_{i} = 1/[(x_{i}^{2} + y_{i}^{2})((\sigma_{xi}/x_{i})^{2} + (\sigma_{yi}/y_{i})^{2})]$$
$$DENOM = \sum w_{i}\sum w_{i}x_{i}^{2} - \left(\sum w_{i}x_{i}\right)^{2}$$
$$\sigma^{2} = \left(\sum w_{i}y_{i} - n\overline{y}^{2}\right)/(n-1)$$
$$\overline{y} = \sum y_{i}w_{i}/\sum w_{i}$$

A-13.6. Other considerations

A-13.6.1. Lower limit of detection

The lower limit of detection of a measurement for direct measurement of a particular radionuclide should be no more than 0.01 times the concentration of that radionuclide listed in Table A–34 or 0.01 times the smallest concentration of that radionuclide listed in Table A–35.

A-13.6.2. Significant radionuclides

A radionuclide shall be determined to be "significant for purposes of classification" if it is contained in waste in concentrations greater than 0.01 times the concentration of that radionuclide listed in Table A–34 or 0.01 times the smallest concentration of that radionuclide listed in Table A–35.

A-13.7. Data maintenance

A-13.7.1. Fuel failure indicator

Special samples should be taken following changes in radioactive waste operations or in fuel performance noted by changes in relative radionuclide activities and total radioactivity in the reactor coolant by factors of four or greater (see Fig. A–22).

It is planned to perform a routine gross alpha analysis, using a scintillation technique in the reactor water coolant, to have an additional indicator in the event of a fuel leak.

A-14. SLOVAKIA

A-14.1. Introduction

There are currently six WWER type nuclear reactors in Slovakia; one of these has been recently shut down and another one has to be shut down by the end of 2008 in accordance with the Slovak admission protocol to the European Union. Decommissioning/dismantling plans are currently being developed. One decommissioning project is in progress, the A1 nuclear power plant, which is a heavy water, CO_2 cooled reactor in Jaslovské Bohunice, Slovakia.

LILW generated from these nuclear power plants and from decommissioning has been disposed of in the national radioactive waste repository in Mochovce.

A-14.1.1. Waste management at nuclear power plants

Historically, the radioactive waste arising and treated inside Slovakian nuclear power plants (PWR/WWER type 440 MW(e) reactors with a common waste management system) has been classified according to the design approaches. Active nuclear power plant waste waters are collected and treated in evaporators. Liquid waste concentrates from this process are stored under controlled conditions (pH, temperature, salt content) in storage tanks. In spite of the controlled storage conditions, there is a small volume of bottom sediments in the tanks. Another liquid waste stream is represented by spent ion exchange resins stored in special tanks, segregated by lower activity from the water purification systems and higher activity (i.e. intermediate level) from purification of the primary circuit. There is also a very small volume of active oils.

Solid waste is traditionally divided into:

- (a) Combustible;
- (b) Non-combustible compactable;
- (c) Non-combustible and non-compactable.



FIG. A-22. Radiochemical analysis of reactor coolant.

It is also relevant to mention the waste at the A1 nuclear power plant (HWGCR, 150 MW(e), which was shut down after accidents in 1978). This reactor is currently in the first stage of decommissioning, which involves, among other activities, the solidification of all the liquid waste, which has been stored for tens of years on the site. There are miscellaneous liquid waste streams, some of which have abnormal contents of radionuclides, so it has been difficult to establish a waste categorization system here. Nevertheless, for the purposes of the Mochovce repository safety assessment, the liquid wastes were divided onto two general groups: those suitable for near surface disposal and those with too great a concentration of long lived radionuclides for such a practice.

A-14.2. Radionuclides of interest

The SF method has been applied to DTM nuclides identified from near surface vault type repository WAC (Table A–37).

A-14.3. Sampling and radiochemical analysis

A-14.3.1. Characterization before conditioning

Before conditioning, the radioactive wastes are controlled as below.

Concentrates from the V1 and V2 nuclear power plants (WWER plants in Jaslovské Bohunice). The producer provides the results from one complete analysis (i.e. for all radionuclides) of each type of waste, as a minimum. Each storage tank has to be analysed once every three years, after its filling and directly before the treatment of waste. The representative sample has to be taken from the tank after the maximum possible homogenization of its contents (including slurries), without further filling. The producer determines the volumetric activities of ¹⁴C, ⁶⁰Co, ⁶³Ni, ⁹⁰Sr, ¹²⁹I, ¹³⁷Cs and ²³⁸Pu. Am-241, ⁹⁹Tc, ⁹⁴Nb and ^{239/240}Pu also need to be determined in cases where the activity of ¹³⁷Cs is higher than 10⁹ Bq/dm³. Before filling of the transport container (for transport to the place of conditioning), the waste producer shall homogenize the tank in the same way as before sampling. SE-VYZ monitors the radionuclide characteristics of the waste after each filling of the collecting reception tank, after its homogenization, by gamma spectrometric measurement of ¹³⁷Cs. Determination of volumetric activities of the above mentioned radionuclides are undertaken by their direct measurement or by measurement of easily measurable nuclides and application of SFs. The waste producer determines the SFs according to a sufficiently representative database of real results. They must be approved by all concerned parties (i.e. the waste producer, the operator of the conditioning facility and the operator of the repository).

Concentrates from the A1 nuclear power plant. The same radionuclide characterization is applied here. The only difference is that SE-VYZ is, at the same time, the waste producer and the operator of the conditioning facilities. All 11 radionuclides have to be determined where necessary.

Ion exchange resins and slurries. The same approach is applied as described above. Am-241, 99 Tc, 94 Nb and $^{239/240}$ Pu must be determined in cases where the activity of 137 Cs is greater than 2×10^{11} Bq/dm³.

DTM nuclide	Key nuclide	DTM nuclide	Key nuclide
C-14	Co-60	Pd-107	Cs-137
Ca-41	Co-60	Sn-126	Cs-137
Ni-59	Co-60	I-129	Cs-137
Ni-63	Co-60	Sm-151	Cs-137
Se-79	Cs-137	Pu-238	Co-60
Sr-90	Cs-137	Pu-239	Co-60
Mo-93	Co-60	Am-241	Co-60
Zr-93	Cs-137	Cs-135	Cs-137
Nb-94	Co-60		

TABLE A-37. DIFFICULT TO MEASURE NUCLIDES AND KEY NUCLIDES

Solid combustible waste. The operator of the repository determines the mass activity of ¹³⁷Cs and ⁶⁰Co by measurement with a gamma scanning device. According to the waste origin and the mass activity of ¹³⁷Cs and ⁶⁰Co, radionuclides ¹⁴C, ⁶³Ni, ⁹⁰Sr, ¹²⁹I and ²³⁸Pu are calculated using the SFs. Am-241, ⁹⁹Tc, ⁹⁴Nb and ^{238/240}Pu must be determined in cases where the activity of ¹³⁷Cs is higher than 2×10^8 Bq/kg.

Incinerator ash. Representative sampling has to be performed from the ash tank directly before its next treatment, after maximum possible homogenization of the tank contents. The same radionuclides (241 Am, 99 Tc, 94 Nb and $^{239/240}$ Pu) have to be determined in cases where the activity of 137 Cs is higher than 2 × 10¹⁰ Bq/kg. Three statistically relevant results are required for each radionuclide. The same approach to direct measurement and the use of CCs is applied as described above.

Solid compactable waste. The same approach as in the case of combustible waste. The decisive activity of ¹³⁷Cs in this waste stream is 2×10^8 Bq/kg. In this case, only ¹³⁷Cs and ⁶⁰Co are used in developing CCs.

Solid non-compactable wastes (concrete debris, contaminated soils). The same approach as above. The list of radionuclides is amended with the inclusion of ⁴¹Ca. The decisive activity of ¹³⁷Cs is 2×10^{11} Bq/kg.

A-14.3.2. Characterization after conditioning

Determination of the radionuclide content of filled fibre reinforced containers is performed by gamma spectrometric measurement of ¹³⁷Cs and ⁶⁰Co in representative samples of cement mortar and subsequent calculation according to the CCs and the technological processes used in the conditioning. At the same time, detailed radionuclide analyses will be performed two to three times per year for the verification of consensus between calculated and measured values. Such analyses are performed by authorized independent organizations, according to the requirement of the regulatory authority.

Cemented concentrate from the V1 and V2 nuclear power plants. Representative samples are taken from each batch of cement mortar. Cs-137 and ⁶⁰Co are measured by gamma spectrometry. Sr-90, ¹⁴C, ⁶³Ni, ²³⁸Pu and ¹²⁹I are determined according to known characteristics of the concentrates before conditioning, knowledge of the conditioning process, gamma spectrometry of ¹³⁷Cs and ⁶⁰Co and known CCs. The activity of ¹³⁷Cs triggering the need to determine also ⁹⁴Nb, ⁹⁹Tc, ²⁴¹Am and ^{239/240}Pu is 10¹¹ Bq/m³. In the case of heterogeneous waste packages (i.e. if the fibre reinforced container is filled with drums containing bituminized concentrates or supercompaction products), the radionuclide content of these inhomogenities is also required. In cases where the drums are filled with concrete debris from decommissioning and inserted into fibre reinforced containers, the list of radionuclides must be amended with ⁴¹Ca.

Cemented concentrates of the A1 nuclear power plant. The same radionuclide characterization is applied here. All 11 radionuclides (plus ⁴¹Ca if needed) have to be determined where necessary.

Cemented ion exchange resins, slurries and ash. The same approach is applied. The decisive activity of ^{137}Cs is $2\times10^{11}\,Bq/m^3$.

In conclusion, the activity or mass activity of 17 radionuclides is limited as the WAC for the Mochovce repository. According to the operational procedure of the waste package producer and, at the same time, the operator of the disposal facility, the concentration of ten radionuclides from this set is routinely controlled; that is: ¹³⁷Cs (by gamma spectrometry); ⁹⁰Sr, ¹⁴C, ⁶³Ni, ²³⁸Pu and ¹²⁹I (according to the known characteristics of the waste before conditioning, knowledge of the conditioning process, gamma spectrometry of ¹³⁷Cs and ⁶⁰Co and known SFs); ⁹⁴Nb, ⁹⁹Tc, ²⁴¹Am and ^{239/240}Pu are controlled by measurements and established SFs. Additionally, ⁶⁰Co is determined as a key nuclide by gamma spectrometry, plus, in some cases, ⁴¹Ca.

A-14.4. Validation of the scaling factor method

SFs are calculated as a linear regression of measured values, excluding low detection limit values. The only requirement to be met after identification of the outliers is that the regression coefficient should be greater than 0.8.

A-14.5. Calculation method

A linear regression model of calculation of SFs was adopted after approval by all concerned parties (i.e. the waste producer, the operator of the conditioning facility and the operator of the repository). However, at the

present time in the case of poor correlation, linear regression of logarithms supported by statistical tests is used as the applicable calculation method.

A-14.6. Other considerations

For the Slovakian waste management system, each waste stream is characterized separately. At this time there are some studies under way to find a more robust method involving several waste streams taken together to provide improved statistical correlations.

A-14.7. Data maintenance

To use SFs for newly generated waste, periodical sampling is carried out as discussed in Section A–14.3; if there are differences in regression analysis, new SFs are calculated.

A-15. SLOVENIA

A-15.1. Introduction

The nuclear programme in Slovenia consists of one commercial power plant, Krsko nuclear power plant, a Westinghouse PWR plant (two loop, 700 MW(e)), which has been in commercial operation since 1982.

All LILW from Krsko nuclear power plant operation is stored in on-site interim storage, operated by the plant. This represents about 95% of radioactive waste in Slovenia. SFs are applied on spent resin, dried concentrate and DAW to determine the alpha content in the storage inventory.

Long term storage/disposal solutions are not available at the moment. A site selection process for a final repository for LILW is in progress. The limiting concentrations of particular radionuclides are not yet regulated.

A-15.2. Radionuclides of interest

The SF method has been applied to determine alpha emitters, which include total alpha, as well as individual alpha emitting nuclides: ²³⁸Pu, ²³⁹Pu, ²⁴¹Am, ²⁴²Cm and ²⁴⁴Cm. Correlation factors are derived based on ¹³⁷Cs as a key nuclide. Co-60 was also considered as a key nuclide during the development phase, and the results were compared. Other DTM nuclides are currently not included in the scope for determination in LILW stored at the Krsko nuclear power plant. The applied SF methodology is approved by the regulator.

A-15.3. Sampling and radiochemical analysis

Radiochemical analysis data are collected from samples of in-drum drying products (spent resin and dried concentrate) and from ashes/filter dust from the incineration of DAW. The number of samples of in-drum drying products is rather low — a few ten of samples per year — due to the low generation rate of waste packages in the process. Samples of ashes/filter dust are collected and analysed during incineration campaigns (by a contractor). Total alpha is analysed from all the above mentioned samples. Particular alpha emitting nuclides are analysed in dried spent resin samples. The percentage of particular alpha emitters in the total alpha is then applied to all waste streams. Values of SFs are monitored periodically to detect significant changes and to update values.

A-15.4. Validation of the scaling factor method

The minimum frequency to recalculate SFs is once per four years. Samples are collected regularly from the process to be analysed.

A-15.5. Calculation method

The arithmetical mean is used to calculate SFs. All together, the number of samples is insufficient for good statistics, due to the low generation rate of waste packages in the process.

A-15.6. Other considerations

In the event that ¹³⁷Cs is not found in the gamma analyses report, the MDA for ¹³⁷Cs is used to determine the alpha content of the package.

A-15.7. Data maintenance

Samples are collected from newly generated drums of spent resin and dried concentrate. Ashes/filter dust is analysed from each incineration campaign. Data are collected in the plant radioactive waste database and reported to the regulator. The minimum frequency to recalculate SFs is once per four years.

A-16. SPAIN

A-16.1. Introduction

A generic description of the Spanish nuclear industry can be summarized as follows:

- (a) Currently eight nuclear reactors operate in Spain (six PWRs and two BWRs).
- (b) Two decommission projects have been developed:
 - (i) Vandellós 1 nuclear power plant (graphite GCR);
 - (ii) Experimental Material Testing Reactor (MTR), and attached facilities, which has just begun dismantling.

The Jose Cabrera nuclear power plant (a PWR) has been recently shut down, and decommission/ dismantling plans are being developed.

A-16.2. Radionuclides of interest

The SF methodology is applied to the following isotopes that are usually present in LILW from the nuclear power plants: ³H, ¹⁴C, ⁵⁹Ni, ⁶³Ni, ⁵⁵Fe, ⁹⁴Nb, ⁹⁰Sr, ⁹⁹Tc, ¹²⁹I, ²³⁸Pu, ^{239/240}Pu, ²⁴¹Pu, ²⁴¹Am, ²⁴²Cm, ^{243/244}Cm.

Some others, such as ⁹³Mo, ⁹³Zr and ³⁶Cl (for specific wastes), are currently under study, all of which could be significant for the long term of the safety assessment of the repository [A–3].

A-16.3. Sampling and radiochemical analysis

LILW produced in the Spanish nuclear power plants (PWR, BWR) is shown below:

- (a) Homogeneous waste:
 - (i) Spent resins (beads or powder);
 - (ii) Evaporator concentrates;
 - (iii) Sludges;
 - (iv) Dried sludges.
- (b) Heterogeneous waste:
 - (i) Cartridge filters;
 - (ii) Compactable waste;
 - (iii) Non-compactable waste.

Every time a nuclear power plant produces homogeneous waste, a sample is taken for radiochemical analysis, for both SF determination and verification purposes (when there is a suitable number of data for a specific waste stream).

A suitable number of data points are necessary for developing a consistent SF based on an accumulated sample. While collecting such a number of samples, an SF is calculated based on:

- (a) Grouping waste streams from the same nuclear power plant;
- (b) If not, grouping waste streams from similar nuclear power plants in the same country;
- (c) Finally, if none of the mentioned options are possible, grouping waste streams from similar foreign nuclear power plants (industrial databank).

The grouping process is supported by statistical tools. This means that the grouped streams are, from the statistical point of view, the same. The test statistic is defined by:

$$t_{n_1+n_2-2} = \frac{|\mu_1 - \mu_2| + Ln(2)}{sb\sqrt{\frac{1}{n_1} + \frac{1}{n_2}}}$$

where μ_1 and μ_2 are the population means for the two data groupings and S_b is the composite variance.

$$s_b^2 = \left(\frac{(n_1 - 1)s_1^2 + (n_2 - 1)s_2^2}{(n_1 + n_2 - 2)}\right)$$

If the test value is less than the Student's *t* value for the given degrees of freedom, defined by $n_1 + n_2 - 2$, it cannot be concluded that the means are different. The higher the dispersion of data, the less likely it is that there will be a distinguishable difference.

Several options can be chosen for determining the suitable data number for accumulated samples; one can be based on the data variance, for example (see Fig. A–23). The uncertainty of SF (red lower curve) has to be less or equal to one half of the data uncertainty (blue upper curve). It is reached with $n \approx 15$. This is a useful tool in the event of lack of correlation, and a mean activity concentration is taken instead of the SF.

For decommissioning purposes, an extended characterization process should be performed in order to identify different streams. In addition, the intrinsic heterogeneity of the material to be sampled makes it necessary to design a scheme or strategy of sampling for radiochemical analysis.



FIG. A-23. Uncertainty of data number of samples.

The main topic to take into account in a decommissioning characterization plan is to collect samples with a large activity range to get representative SFs.

A-16.4. Judgement of applicability of the scaling factor

The starting point for the applicability of the SF is the production mechanism (CP and FP) and the physicochemical behaviour of the radionuclides. This means that the first step is to correlate CP with a CP key nuclide, and FP with the FP. The second aspect to take into account is that the physicochemical behaviour of both ETM and DTM nuclides should be the same.

Finally, many times two different radionuclides present a very good correlation, supported by statistical tools, but it is difficult to justify theoretically by the production mechanism and the physicochemical behaviour, as mentioned above.

Useful information about the SF applicability, from a statistical point of view, is a comparison between the variances of the mean and the regression analysis $(F_{n-1,n-2})$. If the variances are similar, the geometric mean is used as the SF. This is an empirical result when there is correlation. If the variances are different, there is no correlation and a null slope test is usually present.

A-16.5. Selection of the key nuclides

In addition to the topics mentioned above (production mechanism and the physicochemical behaviour), a key nuclide should have the following basic features:

- (a) Gamma emitting and usually present in the waste, therefore easy to detect from outside the waste package;
- (b) Relatively long lived;
- (c) Good correlations with the DTM nuclide.

Many times a group of DTM nuclides (transuranic nuclides, uranium isotopes, nickels) present a very good correlation between them, and it is useful to first correlate them with respect to one of them, and finally correlate the latter with an ETM nuclide. This way of proceeding reduces the overall uncertainty.

Co-60 is generally a very good key nuclide for all the DTM isotopes, if there is no significant fuel failure at the nuclear power plant. When this occurs, ⁶⁰Co and ¹³⁷Cs show a good correlation between them.

Using a radionuclide with a shorter half-live than ⁶⁰Co may give rise to large errors when decay correction has to be applied, especially in relatively old waste.

For dismantling old facilities, the only option for the key nuclide is generally ¹³⁷Cs, and in some cases ^{152/154}Eu (graphite) (Table A–38).

A-16.6. Calculation method

The geometric mean as the SF has shown the best behaviour, better even than regression analysis. Mainly three reasons support this fact:

Type of radionuclide	Evaluated DTM nuclide	Key nuclide
Activated coolant products	H-3, ¹⁴ C	Co-60
CP nuclides	Ni-59, ⁶³ Ni, ⁹⁴ Nb	Co-60
FP nuclides	Sr-90, ¹²⁹ I	Co-60, ¹³⁷ Cs
Alpha emitting nuclides	Total alpha emitting nuclides	Co-60, ¹³⁷ Cs

TABLE A-38. EVALUATED RADIONUCLIDES AND SELECTED KEY NUCLIDES

- (a) When correlation exists, there are no differences in the variance of the two models. Therefore, it is better to chose a one parameter model rather than a two parameter model..
- (b) The geometric mean better predicts activity values outside of the data range from which the geometric mean has been calculated. The regression analysis does not work well outside of the data range.
- (c) The geometric mean is the best estimator of the median, and it is robust even when outliers exist. Both the arithmetical mean and the regression analysis are affected to a great degree by outliers.

The ETM nuclide should be first estimated for every waste package, based on the following possibilities:

- (i) Gamma spectrometry analysis;
- (ii) Sample gamma spectrometry analysis, for homogeneous waste;
- (iii) Theoretical dose rate calculation.

Once the ETM nuclides have been quantified, the following step is the application of the SF in order to estimate the activity of the DTM nuclides.

Currently, some DTM nuclides do not show correlation with any key nuclide, but show a stable temporal trend, which indicates that the mean activity concentration could be taken as the representative activity of the

waste. It does not mean that the uncertainty of the mean activity concentration is higher than the SF uncertainty. The assurance of the accuracy of the SF will be determined essentially by the following:

- Sampling in an appropriate manner;
- The different samples covering the widest possible range of activities, similar to the actual range.

Consequently, this method is valid for both homogeneous and heterogeneous waste.

The SF is used to determine both the activity in a waste package and the activity stored in a radioactive waste storage facility. In the first case the aim is to assess the uncertainty of an element belonging to a population, while in the second it is to assess the uncertainty of the expected value from that population.

To assess the uncertainty [A-4] of the SF, the most appropriate method involves using the data logarithms, used to establish the existence of a correlation, where the geometric mean is converted into the arithmetic mean of the normal distribution.

The standard deviation shown by the data is generally the main element contributing to the uncertainty of the SF, and it may be appreciated that the better the correlation between the isotopes, the smaller the standard deviation of the SF, and that when the correlation is close to a value of 1.0, the uncertainty of the measurement processes becomes of some importance as regards the final uncertainty. Normally, the uncertainties of the measurement processes are included in the standard deviation of the data, depending on the measurement method used. As a result of this an interval of confidence of 95% for the SF, calculated using n samples, would be as follows:

$$\frac{SF_E}{e^{t_{n-1}\frac{S_{SF_j^{\,\cdot}}}{\sqrt{n}}}} \leq SF_T \leq SF_E e^{t_{n-1}\frac{S_{SF_j^{\,\cdot}}}{\sqrt{n}}}$$

where

 t_{n-1} is the value of the quintile $1 - \alpha/2$ (=0.975 if $\alpha = 0.05$) of the *t* of Student of n - 1 degrees of liberty; SF_E is the sample SF (i.e. the one estimated using the data);

 SF_T is the true SF.

As a result of this, DTM activities in a single waste package could have a relatively high uncertainty, but the final uncertainty in DTMs stored in a repository is decreased by a considerable amount, due to the large number of waste packages disposed of.

A-16.7. Classification of the scaling factor

The perfect scenario is to have available one SF for each waste stream of a nuclear power plant, but measurements have a real cost and the very similar behaviour of data from a similar nuclear power plant makes it possible to have a unique SF for several nuclear power plants (Table A–39) and several waste streams. Grouping tests are performed in order to prove such behaviour.

A-16.8. Continuous using of the same scaling factor

If accumulated samples have been collected during a long period of time, and the values have not shown a trend change, the same SF could be used for that period of time. For old waste, the prior statement can be used supported by some radiochemical measurements over that waste and by statistical tools.

A-16.9. Data verification

Two verification processes should be made:

- (a) Data set verification and outliers. If outliers are identified, care should be taken to identify the source of that value (transcription errors, laboratory process, etc.).
 - (i) One outlier identification: Grubb's test [A–5].

(ii) Either one or more outliers (statistical heterogeneity): normalized 4th central moment, to be compared with the 95% tabulated data.

$$m_4 N = \frac{\sum_{i=1}^{i=n} (x_i - \overline{x})^4}{ns^4}; \ s = \sqrt{\frac{\sum_{i=1}^{i=n} (x_i - \overline{x})^2}{n-1}}$$

(b) SF verification with new data. Once the SF has been established, future data serves to both feed the database and verify the prior established value. At least three new values have to be collected to perform the grouping test defined above; one group is the additional data and the other is the original set from which the SF was calculated. If the additional data have a good behaviour, the prior SF can remain at the current value, otherwise old measurements are removed from the database and the additional ones included, in order to have a current SF.

Radionuclides	Features affecting the SF
C-14, ³ H	Type of reactor (PWR, BWR)
Ni-59, ⁶³ Ni, ⁹⁴ Nb	Type of reactor (BWR, PWR) Improvements of reactor component materials
FP and alpha emitting nuclides	Fuel failure history Type of waste Key nuclide

TABLE A-39. CLASSIFICATION AND INTEGRATION OF THE SCALING FACTOR

A-17. SWEDEN

A-17.1. Introduction

The nuclear power plants in Sweden are all light water reactors (seven BWRs and three PWRs). Two BWR reactors have been shut down during recent years and are now under care and maintenance and are foreseen to be dismantled, starting in 2020. The plants still in operation have an estimated operational lifetime of 50–60 years. Since all reactors were brought into operation between 1972 and 1985, operation is planned to continue for several decades. The operational short lived waste is disposed of in a common repository (SFR, shown in Fig. A–24) that has been in operation since 1988. Long lived waste is stored waiting for a final repository to be built around 2045.

As shown in Fig. A–25, waste packages disposed of in the repository are mainly drums and boxes filled with cement and bitumen solidified ion exchange resins from water cleanup systems. Scrap and trash are collected in boxes or sea containers.

Short lived waste contains small quantities of long lived radionuclides, and these small quantities have a significant impact on the long term safety of the repository. This section focuses on the estimation of long lived radionuclides in the short lived waste.

The safety analyses of the repository are based on a source term derived from the prognosis of the waste to be disposed in different parts of the repository until the final closure and sealing. In the operational licence the estimated amount of radionuclides in the repository were given as radiation protection limits. The quantities of gamma emitting nuclides were based on measurements on the waste and the DTM nuclides were estimated with the use of SFs to key nuclides, the ETM nuclides ⁶⁰Co for CPs and ¹³⁷Cs for FPs. Alpha emitters were analysed in the reactor coolant and assumed to be transferred to waste packages.



FIG. A-24. Final repository for LILW (SFR).



FIG. A-25. Waste packaging for short lived LILW.

TABLE A-40. EXAMPLES OF RADIONUCLIDES CLASSIFIED ACCORDING TO FORMATION PATHWAYS

Activation products	Fission products	Transuranics
H-3, ¹⁰ Be, ¹⁴ C, ³⁶ Cl, ⁵⁵ Fe, ⁶⁰ Co, ⁵⁹ Ni, ⁶³ Ni, ⁹³ Zr, ⁹³ Nb, ⁹³ Mo, ⁹⁹ Tc	Se-79, ⁹⁰ Sr, ⁹³ Nb, ⁹⁹ Tc, ¹⁰⁷ Pd, ¹⁰⁸ Ag, ¹¹³ Cd, ¹²⁶ Sn, ¹²⁵ Sb, ¹²⁹ I, ¹³⁴ Cs, ¹³⁵ Cs,	U-238, ²³⁷ Np, ²³⁹ Np, ²³⁸ Pu, ²³⁹ Pu, ²⁴⁹ Pu, ²⁴¹ Pu, ²⁴² Pu, ²⁴⁴ Pu, ²⁴² Am,
	¹³⁷ Cs	²⁴³ Am, ²⁴¹ Am, ²⁴² Cm, ²⁴³ Cm, ²⁴⁴ Cm

Note: key nuclides are marked in bold.

A-17.2. Radionuclides of interest

Methods for correlating DTM nuclides to ETM nuclides have been developed over the years and new knowledge on radionuclide behaviour has been gained. The focus is on improving the knowledge of radionuclides important to safety of the repository, as summarized in Table A–40. Special attention is given ¹⁴C, ⁵⁹Ni, ⁶³Ni, ³⁶Cl, ⁹³Mo, ⁹⁹Tc, ¹²⁹I and ¹³⁵Cs.

For APs and FPs a set of correlation factors based on the radionuclide contents in surface contamination has been applied to safety analyses. Co-60 is the key nuclide for APs and ¹³⁷Cs is used for FPs. Alpha emitters (transuranic nuclides) are correlated to ²³⁹⁺²⁴⁰Pu, based on analyses of reactor water samples.

To evaluate the uncertainty of correlation factors, both a statistical method and a qualitative approach have been used. For the statistical method, the geometric mean value from different sources is used (measurements on different waste in different plants and also from different countries) and the standard deviation was calculated. The statistical evaluation has to be modified by qualitative arguments. Such arguments typically include:

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

A safety assessment of a future deep repository for long lived LILW was conducted during the second half of the 1990s. Within that project, correlation factors for estimating the inventory of radionuclides that are DTM experimentally were developed (Table A–41). These factors have also partly been used to estimate a reference inventory for SFR.

Radionuclide	Correlation factor	Source for selected correlation factor
H-3 ^a	1×10^{-4}	Data on induced activity in metals
Be-10 ^a	6×10^{-10}	Data on BWR spent fuel assembly
C-14 ^a (BWR)	3×10^{-3}	Measurements of ¹⁴ C and ⁶⁰ Co in Swedish ion exchange resins
C-14 ^a (PWR)	8×10^{-2}	Measurements of ¹⁴ C and ⁶⁰ Co in Swedish ion exchange resins
Cl-36 ^a	6×10^{-7}	Activity ratio: modelled activity of ³⁶ Cl in resins from Swedish nuclear power plant and measured activity of the key nuclide in operational waste
Fe-55 ^a	1	Data on Swedish operational waste
Ni-59 ^a (BWR)	1×10^{-3}	Measurements of ⁵⁹ Ni and ⁶⁰ Co in Swedish ion exchange resins
Ni-59 ^a (PWR)	3×10^{-2}	Measurements of ⁵⁹ Ni and ⁶⁰ Co in Swedish ion exchange resins
Ni-63 ^a (BWR)	8×10^{-2}	Measurements of ⁶³ Ni and ⁶⁰ Co in Swedish ion exchange resins
Ni-63 ^a (PWR)	4	Measurements of ⁶³ Ni and ⁶⁰ Co in Swedish ion exchange resins
Se-79 ^b	4×10^{-6}	Data on Swedish BWR spent fuel assembly
Mo-93 ^a	1×10^{-6}	Activity ratio: modelled activity of ⁹³ Mo in resins from Swedish nuclear power plant and measured activity ⁶⁰ Co in operational waste
Zr-93 ^a	1×10^{-6}	Data on US PWR spent fuel assembly
Nb-93m ^a	1×10^{-3}	Correlation to ⁹⁴ Nb
Nb-94 ^a	1×10^{-5}	Data on spent fuel assembly
Tc-99 ^b	9×10^{-4}	All Swedish data
Pd-107 ^b	1×10^{-6}	Data on spent fuel assembly
Ag-108m ^a	6×10^{-5}	Data on Swedish BWR spent fuel assembly
Cd-113m ^b	6×10^{-4}	Data on Swedish BWR spent fuel assembly
Sb-125 ^a	1×10^{-1}	Mainly on measured Swedish data
Sn-126 ^b	5×10^{-7}	Data on Swiss model inventory, reactor internals
I-129 ^b	3×10^{-6}	Activity ratio: modelled activity of ¹²⁹ I in resins from Swedish nuclear power plant and measured activity of ¹³⁷ Cs in operational waste
Ba-133 ^a	1×10^{-5}	Data on Swiss model inventory, reactor internals
Cs-134 ^b	1	Mainly on measured Swedish data
Cs-135 ^b	1×10^{-5}	Activity ratio: modelled activity of ¹³⁵ Cs in resins from Swedish nuclear power plant and measured activity of ¹³⁷ Cs in operational waste
Pm-147 ^b	9×10^{-1}	Data on Swedish BWR spent fuel assembly
Sm-151 ^b	3×10^{-3}	Data on Swedish BWR spent fuel assembly
Eu-152 ^b	7×10^{-5}	Data on Swedish BWR spent fuel assembly
Eu-154 ^b	1×10^{-1}	Data on Swedish BWR spent fuel assembly
Eu-155 ^b	7×10^{-2}	Data on Swedish BWR spent fuel assembly
Ho-166m ^a	4×10^{-6}	Data on Swedish BWR spent fuel assembly

Radionuclide	Correlation factor	Source for selected correlation factor
U-232 ^c	3×10^{-5}	Data on Swedish BWR spent fuel assembly
U-233 ^c	2×10^{-8}	Data on Swedish BWR spent fuel assembly
U-234 ^c	1×10^{-3}	Data on Swedish BWR spent fuel assembly
U-235 ^c	2×10^{-5}	Data on Swedish BWR spent fuel assembly
U-236 ^c	3×10^{-4}	Data on Swedish BWR spent fuel assembly
U-238 ^c	4×10^{-4}	Data on Swedish BWR spent fuel assembly

TABLE A-41. CORRELATION FACTORS IN SAFETY ASSESSMENTS OF THE SFR REPOSITORY (cont.)

^a Correlates to ⁶⁰Co.

^b Correlates to ¹³⁷Cs.

^c Correlates to 239 Pu + 240 Pu.

Note: The correlation factors are aimed to be representative for waste one year after discharge from the nuclear facility.

A-17.2.1. Selection of correlation factors

New data from estimations and measurements of the radionuclide content and correlation factors in different types of radioactive waste have been collected. The data are evaluated and updated correlation factors representative for fresh waste for eight radionuclides (¹⁴C, ³⁶Cl, ⁵⁹Ni, ⁶³Ni, ⁹³Mo, ⁹⁹Tc, ¹²⁹I and ¹³⁵Cs) are suggested. New information from measurements in Sweden have made it possible to define correlation factors that are BWR and PWR specific for ¹⁴C, ⁵⁹Ni and ⁶³Ni. Previous reactor specific data were few and therefore the correlation factors were made generic.

A-17.2.2. Uncertainties

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

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

In some cases there is a significant difference between the statistical approach and the selected values. This has called for special considerations, and for some DTM nuclides special projects have been performed in recent years and new sampling programmes have been introduced.

A-17.2.3. Considerations concerning C-14

In safety assessments of the SFR repository, the highest contribution to long term dose consequences is from ¹⁴C. It is therefore desirable to reduce the uncertainty in the estimated ¹⁴C contents in the repository.

The common correlation of ¹⁴C to ⁶⁰Co has been proven not to be optimal. A correlation only exists momentarily, which requires more or less continuously sampling and analyses to reduce uncertainty to an acceptable level. This is both time and dose consuming. An SF approach needs reactor specific as well as waste stream specific SFs rather than a generic factor (Table A–42). A recent research programme [A–6] has suggested that correlation should be based on energy production rather than by correlation to ⁶⁰Co. The annual and prognosticated amounts of ¹⁴C are calculated reactor specific, owing to the production of thermal energy. Specific analysis has been performed to evaluate the degree of the uptake of ¹⁴C in different cleanup systems in the different types of reactors. The ¹⁴C content is also divided into inorganic and organic phases.

	Correlation factor	Uncertainty	
Radionuclide		Minimum	Maximum
C-14, BWR	3.10-3	3.10-6	1.10-1
C-14, PWR	8.10-2	2.10-2	9.10-1
Cl-36/60Co	6.10-7	_	_
Cl-36/ ¹³⁷ Cs	3.10-6	_	_
Ni-59, BWR	1.10-3	7.10-5	6.10-3
Ni-59, PWR	3.10-2	2.10-2	6.10-2
Ni-63, BWR	8.10-2	9.10-3	5.10-1
Ni-63, PWR	4	3	6
Mo-93	1.10-6	_	_
Tc-99	9.10-4	8.10-7	1
I-129	3.10-6	_	_
Cs-135	1.10-5	_	_

TABLE A-42. SCALING FACTOR

Since the amount of ¹⁴C produced in the coolant of a specific reactor directly corresponds to the energy production, the total accumulation of ¹⁴C in the ion exchange resins can be estimated if the following factors are known:

- (a) The generated electricity of the reactor (MW·h(e));
- (b) The thermal efficiency of the unit $(MW \cdot h(e)/MW \cdot h(th))$;
- (c) The normalized production rate of 14 C in the coolant water (Bq/MW·h(th));
- (d) The total accumulated fraction of 14 C in the cleanup systems (per cent of production in the coolant).

The methodology to estimate organic fractions of ¹⁴C has been further developed, and it has been possible to derive different ratios for BWRs and PWRs.

A-18. UKRAINE

A-18.1. Introduction

There are 15 WWER units currently operating in Ukraine: two 440 MW(e) units (at the Rovno nuclear power plant site) and thirteen 1000 MW(e) units (two at the Rovno nuclear power plant site, two at the Khmelnitsky nuclear power plant site, three at the Uzhnoukraiskaya nuclear power plant site and six at the Zaporozhskaya nuclear power plant site). There are also three RBMK type reactors at the Chernobyl nuclear power plant site at the stage of operation termination (preparation to decommissioning). The accidentally destroyed Unit 4 is under transformation to an ecologically safe system (the 'shelter'). Low and intermediate level short lived radioactive waste generated by these nuclear power plants will be disposed of in the near surface disposal facilities of the Vektor complex site (see Table A–43). The first stage of the Vektor complex is planned to be commissioned in 2008.

According to the recommendations on establishing acceptance criteria for conditioned radioactive waste for disposal in near surface repositories, the radionuclide composition of radioactive waste in a package should be identified with adequate accuracy. In establishing the radioactive waste acceptance criteria, the disposal facility operator should require from the waste supplier information on the radionuclide composition of the radioactive waste in a package (a list of radionuclides and their content in the package) for the following radio-nuclides: ³H, ¹⁰Be, ¹⁴C, ³⁶Cl, ⁴¹Ca, ⁵⁴Mn, ⁵⁵Fe, ⁵⁹Ni, ⁶³Ni, ⁶⁰Co, ⁹³Zr, ⁹⁴Nb, ⁹⁰Sr, ⁹⁹Tc, ¹¹⁰Ag, ¹²⁹I, ¹³⁴Cs, ¹³⁵Cs, ¹³⁷Cs,

TABLE A-43. WASTE PACKAGES INTENDED FOR DISPOSAL AT THE VEKTOR COMPLEX



²²⁶Ra, ²³²Th, ²³⁵U, ²³⁶U, ²³⁸U, ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Pu, ²⁴¹Am and other transuranic elements if present, which are mostly long lived and DTM nuclides. Acceptable activity or activity concentration limits are defined for individual radionuclides and groups of radionuclides based on safety analysis results (according to the operational safety issues, scenarios of potential exposure, conditions of the disposal site, facility designed engineered barriers, etc.).

To evaluate SFs between DTM nuclides and key nuclides for the Chernobyl nuclear power plant site waste, activity concentration data on radioactive waste for DTM nuclides have been accumulated through a radiochemical analysis of more than one thousand samples:

- (a) Evaporator concentrate;
- (b) Spent resin;
- (c) Spent perlite;
- (d) Samples of contaminated equipment, pipes, wires, walls, etc. (samples from Units 1–3).

Thousands of spectrometric measurements, including X ray spectrometry, in the frame of a complex engineering and radiological survey were performed. The information database for decommissioning purposes (Infodec) stores the complex engineering and radiological survey results. Also, results of activation and FP inventory calculations for the Unit 2 reactor are included.

The gamma emitting nuclides, ⁶⁰Co and ¹³⁷Cs were selected as the key nuclides because of their detectability when measured by non-destructive radioactivity measurement equipment (gamma spectrometers with a germanium semiconductor detector). These key nuclides are selected for DTM nuclides, considering the similarity of the production mechanisms and/or the physicochemical behaviour of the radionuclides.

Based on the radiochemical analysis data, generic SFs for DTM nuclides were determined for most streams of waste. The geometric mean is used for the calculation method of the SFs.

A-19. UNITED KINGDOM

A-19.1. Introduction

The operation of nuclear power plants gives rise to large volumes of dry solid LLW. In the UK this LLW is disposed of in a LLW repository at Drigg in Cumbria. In accordance with the policy for waste minimization and early disposal of LLW, the strategy adopted is to process the waste using a combination of sorting, incineration and compaction to reduce the volume where appropriate, followed by packaging and disposal. The waste is either put in drums or wrapped and placed in larger containers.

The conditions for disposal require information on specific radionuclide levels. There is, therefore, a need for detailed data concerning the radionuclide content of the LLW arising at the plants. Any approach used to obtain this information has to take into account the nature of the LLW arisings, the volumes involved and the difficulty of direct analysis of each item of LLW for low energy X ray, pure beta and alpha emitting nuclides (i.e. DTM nuclides).

The overall approach adopted was to establish a method whereby the LLW is collected, processed and stored in drums or packages. The inventory of gamma emitters (ETM nuclides) is then established from radiation measurements, such as gamma spectrometry or dose rate measurements on the outside of the drum or package. The activity of the DTM nuclides is then derived using knowledge of their ratios (SFs) to choose the key gamma emitting nuclides.

The approach used is based on the fact that the activity on the dry solid LLW comes from the surface contamination of the plant areas and components where work is being carried out and where the item was used, and on the premise that the transfer process of contamination, the transfer coefficient, from the area to items which become LLW is the same for all radionuclides. Therefore, while only a fraction of activity is transferred to the waste during the contamination process, the isotopic ratios (SFs) of radionuclides in the waste will remain constant.

A-19.2. Waste stream fingerprints (scaling factors)

Areas where the contamination is similar are grouped for LLW processing purposes. The set of isotopic ratios for a given area activity contamination is called the area 'fingerprint'. In order to manage the large number of plant areas where LLW is produced, areas are combined into waste streams based on the similarity of their fingerprint and by taking into account the volumes and, more importantly, the activities of the waste produced. The waste stream fingerprints are presented as ratios to either ⁶⁰Co (AP dominated) or ¹³⁷Cs (FP/actinide dominated). The ⁶⁰Co to ¹³⁷Cs ratio is also determined for each waste stream for use in determining its inventory where dose rate measurements are used.

Waste stream fingerprints are produced for each plant. Typically, each plant has four to seven waste streams. The waste streams are defined based on the likely sources of activity, the type of work that is being carried out and operational considerations in managing the LLW produced.

Typical waste streams for a gas reactor plant are as follows (the detailed areas may vary from plant to plant):

- (a) A dry fuel route stream: this would include the pile cap and areas associated with handling fuel assembly components and spent fuel prior to storage in the spent fuel pond.
- (b) A wet fuel route: this would include the spent fuel storage ponds area, spent fuel flask handling and washing area, and active effluent treatment plant areas.

A-19.3. Radionuclides of interest

The radionuclides that need to be evaluated are defined by the requirements of the repository. Typical radionuclides evaluated by the fingerprint approach are shown in Table A–44.

The area fingerprints are produced from plant measurements and are therefore empirically derived. Co-60 and ¹³⁷Cs were selected as the key nuclides.

TABLE A-44.	EVALUATED	RADIONUCLIDES	AND SELECTION	OF THE KEY NUCLIDES

Type of radionuclide	Evaluated radionuclides	Key nuclide
Activation products	H-3, ¹⁴ C, ³⁵ S, ³⁶ Cl, ⁴⁵ Ca, ⁵¹ Cr, ⁶³ Ni, ⁵⁴ Mn, ⁵⁵ Fe, etc.	Co-60
Fission products	Sr-90, ¹⁴⁴ Ce, ¹⁴⁷ Pm, etc.	Cs-137
Actinides	Uranium, plutonium, curium isotopes, ²⁴¹ Am, etc.	Cs-137

A-19.4. Sampling and radiochemical analysis

The fingerprint for each waste stream is derived from radiochemical analysis of a large number of 'swabs' from the areas and components in the waste stream. The areas from which the swabs are taken are identified in conjunction with the plant operators. The number of swabs taken in each area depends on the size of the area, its complexity and type of work carried out and the volume of LLW produced in the area. Typically 50–250 filter paper swabs are taken for each waste stream. In addition, samples of actual waste are obtained for analysis where appropriate. When establishing or revising the waste stream fingerprint, the sampling takes place in a single campaign over a period of a few days.

It is impractical to carry out radiochemical analysis on all the swab samples. Therefore, bulk samples are produced for each area. A representative subsample is taken from the bulk sample for radiochemical analysis. Prior to the analysis being performed, the representative subsample is checked against the bulk sample by gamma spectrometry. Following the radiochemical analysis and the determination of the concentrations of radionuclides as Bq/g sample, the area fingerprint ratios are calculated. Radionuclides that fall below the minimum detectable limits in the analysis are assessed on a case by case basis using a variety of methods in comparison with the results that have been obtained.

The waste stream fingerprint (F_{stream}) is then derived from the area fingerprints (F_i) using the weighted mean. In deriving the waste stream fingerprint, the area fingerprints are weighted based on the relative contribution (X_i) of each area to the activity of waste arising from the waste stream:

 $F_{\text{stream}} = \text{Sum}(F_i \times X_i)$

A-19.5. Judgement of applicability of the scaling factor

The ISO 5725-2:1994 [A–5] use and accuracy of the fingerprint/SF approach depends on a number of factors. These are:

- (a) The variation in area contamination: the impact of this on the SFs is smoothed out because the activity on LLW arises from many places in an area where work is carried out, and the various LLW items are collected and put together. The collection of a large number of swabs ensures that the fingerprint developed is representative of the average activity in the area.
- (b) The assumption that the transfer coefficient, whereby contamination is transferred from the area to the items that become LLW, is the same for all radionuclides: for radionuclides that the SFs (ratio to the key nuclide) are determined from the radiochemical analysis of the measurements, as the measurement process (swabs) replicate the transfer of activity that occurs in operation, the validity of this assumption is not strictly necessary. However, for radionuclides that are below the minimum detectable levels in the measurements, this assumption is used in their assessment methodology.
- (c) The quality and consistency of the sampling programme: this is controlled by using dedicated teams for carrying out the sampling (swabbing) process.
- (d) The precision of the analytical radiochemical analysis: the analytical work is carried out using specified work instructions with appropriate quality assurance procedures.
- (e) Operational conditions: where the LLW arisings from an area depend on operational conditions and vary between normal operation and outages, swab sampling is carried out during both normal operation and outages to allow for this variation.
- (f) The calculation of the key nuclides from measurements: the key nuclides (⁶⁰Co and ¹³⁷Cs) are derived from radiation measurements such as gamma spectrometry or dose rate measurements. The relationship of the key nuclide activity to these measurements is derived from theoretical modelling, taking into account the random distribution of activity in the waste and the nature of the waste (plastics, concrete, cloth, etc.). Usually, a conservative approach is used in deriving the key nuclide and measurement relationship.
- (g) Area weighting factor: used in combining area fingerprints to give waste stream fingerprints, these factors are based on historical information. As significant changes in waste production patterns could have an impact on the accuracy of the waste stream fingerprint, periodic review of waste arisings is essential.

A-19.6. Selection of the key nuclides

As stated above, the area fingerprints are produced from plant measurements and are therefore empirically derived. Co-60 and ¹³⁷Cs were selected as the key nuclides for the following reasons:

- (a) They are gamma emitting and tend to be present in the waste and are therefore easy to detect from the outside of the package;
- (b) They are relatively long lived;
- (c) They correlate with the DTM nuclides that they are linked to in terms of production and/or behaviour.

Although it may be desirable to have a link between a radionuclide and its key nuclide based on the similarities in their production mechanisms and on their physical and chemical properties, very often such an approach is extremely complex and can only provide an indication of the likely magnitude of the SF. In practice, the SFs are derived empirically.

A-19.7. Calculation method

A waste stream is composed of a number of areas. A fingerprint is produced for each area based on a representative sample of a large number of swabs. Where the activity of LLW produced in each area is the same, the waste stream fingerprint is the arithmetic mean of the area fingerprints. In practice, the activity of LLW produced in each area is different. As the LLW from all the areas in a waste stream is collected together, the waste stream fingerprint is derived using a weighted average of the area fingerprints:

$$F_{\text{stream}} = \text{Sum}\left(F_i \times X_i\right)$$

where

 F_{stream} is the waste stream fingerprint; F_i is the area fingerprints;

 X_i is the relative contribution of area to the LLW volume (sum $X_i = 1$).

Approximate activity information is provided by the plant, generally in the form of contact dose rates on items, bags or drums of waste, collected from each area over a period of time.

The LLW from a waste stream is collected separately, and when there is a sufficient volume it is processed and stored for disposal. On the basis that a large volume of LLW is processed each time for a waste stream, it is considered that, on average, the fingerprint is representative of the activity on the LLW.

A-19.8. Continuous use of the same scaling factor

Plant waste stream fingerprints have been developed following a large programme of sampling and analysis. A waste stream or area fingerprint can be re-assessed as follows:

- (a) When plant operation is likely to result in a significant change in a waste stream or area fingerprint, then a specific re-assessment of that waste stream and/or area fingerprint is carried out;
- (b) Periodic reviews of plant operation and of the performance of the LLW management process may identify reasons for partial or total fingerprint re-assessment or justify the continued use of the established fingerprint;
- (c) At periodic intervals a complete re-assessment of the waste stream fingerprint is initiated.

A-20. UNITED STATES OF AMERICA

A-20.1. Introduction

The first nuclear power plants were placed into service in the USA in the late 1950s. Currently there are 100 operating nuclear power reactors, including 67 PWRs and 33 BWRs. While none of the earliest plants are currently operating, some of the current group have more than 35 years of operation. Prior to separating the responsibilities of the former Atomic Energy Commission in 1973, low level radioactive waste was disposed of in government operated disposal sites. The principal limitation imposed on disposal was the demonstration that the waste contained less than 10 nCi/g of transuranic nuclides. This requirement was carried over to the first commercial low waste disposal sites. Utility waste generators had sampling programme in place during the 1970s to determine the transuranic content. In extending the applicability of radiochemical sample data, scaling ratios were developed to determine the transuranic nuclides from ¹⁴⁴Ce, which could often be observed in gamma spectrum data. The idea of SFs was endorsed in subsequent criteria developed by the Nuclear Regulatory Commission in support of new regulations issued in 1983 covering the licensing of new radioactive waste disposal sites.

Title 10 of the US Code of Federal Regulations, Part 61 (10CFR61) [A–7], imposes a system of classification for disposal on the generators of radioactive waste. The classification system is based on the concentrations of certain radionuclides found in the waste streams. Many of these radionuclides are non-gamma emitting and cannot be measured using the conventional equipment typically found in a nuclear power plant. Measurement is accomplished by collecting samples from the various waste streams and sending the samples off-site for analysis at a specially equipped radiochemistry laboratory. Results from the laboratory analysis are generalized by defining ratios between the DTM nuclides and commonly found gamma emitting nuclides, particularly cobalt and caesium. These ratios, which are referred to as SFs, can then be used in the characterization of other batches of material collected from the applicable streams. The US Nuclear Regulatory Commission recognized and advocated such an approach in 1983 in the Low Level Waste Branch Technical Position on Classification issued concurrent with 10CFR61.

Most of the currently operating US nuclear power plants have been collecting samples and purchasing radiochemical analyses since the beginning of their operation. Some of these plants have undertaken a systematic evaluation of their sample data to check for long term trends or commonality of values between waste streams. Typical PWR and BWR waste streams are shown in Tables A–45 and A–46, respectively. Such an analysis can provide support for the use of constant SFs for some ratios, combining some waste streams for sample collection and reducing radiochemistry requirements. In addition to the potential reduction in sampling costs, another benefit would be more consistent and predictable waste disposal classification.

A-20.2. Sample database

Depending on the particular plant type and specific processes, usually sampling is limited to about six or seven samples per year, which are subjected to full radiochemical analysis. These may serve as the basis for defining SFs or they may be combined with prior year samples to build statistical confidence.

*
Reactor coolant filter
Primary/fuel pool high activity resins
Boron recovery system cartridge filters
Spent fuel pool filters
Radioactive waste processing system charcoal
DAW

TABLE A-45. TYPICAL PRESSURIZED WATER REACTOR WASTE STREAMS

Description	
RWCU powdered resins	
Fuel pool/radioactive waste resins	
Condensate cleanup system resins	
Filter sludge	
DAW	

TABLE A-46. TYPICAL BOILING WATER REACTOR WASTE STREAMS

TABLE A-47. EVALUATED RADIONUCLIDES

Radionuclide group	Examples
Fission products	Cs-137, ⁹⁰ Sr, ⁹⁹ Tc, ¹²⁹ I
Metal activation products	Co-60, ⁵⁹ Ni, ⁶³ Ni, ⁹⁴ Nb, ¹⁴ C
Transuranic	Isotopes of plutonium, curium and americium
Activated coolant products	H-3, ¹⁴ C

A-20.3. Selection of the key nuclides

Radionuclides subject to SF evaluation are specifically identified in US regulations. As summarized in Table A–47, the radionuclides fall roughly into four groups: FPs, activated CPs, transuranic nuclides and activated coolant products.

Ideally, a key nuclide would have very comparable production characteristics and transport properties as the scaled radionuclide. For example, ⁶³Ni and ⁶⁰Co are produced by the same process and in the same material. The two corresponding elements, nickel and cobalt, are both transition metals in group VIIA in the periodic table. As such, their oxidation potentials are very similar and are both expected to behave in the same way when transported through the reactor system. In fact, ⁶³Ni and ⁶⁰Co make very reliable and predicable SFs.

In contrast, ⁹⁰Sr and ¹³⁷Cs are both FPs with very close production rates and half-lives. Both are alkaline metals from groups IIA and IA, respectively, in the periodic table. However, caesium, in contrast to strontium, is readily soluble in water and mobile in reactor fuel. Strontium is soluble under varying conditions and effectively immobile in reactor fuel. The ⁹⁰Sr/¹³⁷Cs ratio is not as predictable. It can be quite stable under conditions of high fuel integrity but can vary widely under other conditions.

The determination of which SFs are evaluated is made on the basis of: (a) a reasonable empirical relationship between the scaled radionuclide and the key nuclide and (b) consistent observation of the key in local gamma measurements. With improved fuel performance in US power plants, there has been increasing dependence on 60 Co as the key for a number of FPs and transuranics.

For less soluble FPs, including ⁹⁰Sr and ⁹⁹Tc, and for transuranic nuclides, ⁶⁰Co often appears as a more stable and reliable key than ¹³⁷Cs [A–8]. This could be attributed to two conditions. One, there is little fuel failure, so the production of transuranics and FPs is limited to fuel present as contamination on reactor surfaces; the release processes more closely mirror releases of CPs. Two, all of the radionuclides are electropositive and tend to adhere to reactor surfaces and transport in the corrosion layers. This is especially true in the relationship between cobalt and transuranic elements.

A-20.4. Scaling factor analysis

SFs may be determined on the basis of an individual stream or, for specific ratios where the data support it, by various combinations of waste streams. Statistical evaluation of the sample data is performed using logarithmic values. Studies performed in the USA [A–9] early in the implementation of the SF method

demonstrated using probability plots that radiochemical sample data followed a log-normal distribution. Using the sample database, SFs are determined as geometric mean ratios. The determination of the ratio can include data from various streams and over varying time spans. The geometric mean ratio is reported along with the geometric dispersion and median to assist the users in the evaluation of the results. The geometric mean and dispersion are determined by transforming the data to logarithms and by calculating the average and variance. The variance is the square root of the standard deviation. The geometric mean is the antilog of the logarithmic average and the dispersion is the antilog of the variance.

Geometric Mean = antilog
$$\left(\frac{\sum_{i=1}^{n} \log \left(\frac{y_i}{x_i}\right)}{n}\right)$$

Geometric Dispersion = antilog ($\sqrt{s^2}$)

where:

$$s^{2} = \frac{\left(\sum \log \left(\frac{y_{i}}{x_{i}}\right)\right)^{2} - \sum \log \left(\frac{y_{i}}{x_{i}}\right)^{2}/n}{(n-1)}$$

where

- y_i is the scaled radionuclide concentration;
- x_i is the key nuclide concentration;
- *n* is the number of samples;
- s^2 is the standard deviation;

s is the variance.

While not universally applied, statistical tests can be performed to determine whether different data populations are statistically the same (i.e. given the mean and variance, can the data be treated as one population for determining the mean?). The pooled variance [A-10] tests the data for differences at the 95% confidence level based on a *t* test. The statistical report includes a comparison of each stream with the combined set of streams and a comparison between individual streams. The pooled variance tests whether or not the data from two or more streams can be statistically the same. That is, given the number of samples, the mean and the variance, can one conclude that the results are different at the 95% level? The comparison incorporates a factor of difference, where if the factor is 1, it implies that the two data sets are statistically the same. A factor of 2 would imply that the two data sets are different by a factor of 2 at the 95% level.

The pooled variance is calculated from:

$$S_b^2 = \frac{(n_1 - 1)S_1^2 + (n_2 - 1)S_2^2}{n_1 + n_2 - 2}$$

where

 S_1^2 is the variance squared of waste stream 1;

 S_2^2 is the variance squared of waste stream 2.

The test statistic is then defined using the pooled variance:

$$t = \frac{|\mu_1 - \mu_2| - \log(D)}{S_b \sqrt{\frac{1}{n_1} + \frac{1}{n_2}}}$$

where

 μ_1 is the average log value of waste stream 1;

 μ_2 is the average log value of waste stream 2;

 n_1 is the sample count for waste stream 1;

 n_2 is the sample count for waste stream 2;

 S_b is the pooled variance;

log(D) is the test difference in means (factor of D).

The test statistic is compared with the Student *t* distribution value at the 95% confidence level. A value of D = 1 (log(d) = 0) implies that there is no significant difference between the data sets being compared. A value of 2 would imply a factor of two differences, and so on.

An additional test that can be important in planning future sampling is a trend test. Using data accumulated over some time period, the trend test determines whether the SF is changing, and if it is changing it determines the rate of change. Depending on the units selected, the change rate divided into the allowed variability will determine the maximum time before sampling must take place.

To test for a trend, SF values are regressed according to date. A linear regression analysis is performed along with a slope = 0 test to see if the SF is varying with time. If the null hypothesis (H_0 : $\beta_0 = 0$) is true, this means statistically that the slope cannot be determined to be different from 0 at the 95% level and, effectively, that the SF is constant with time.

Given the true slope β_0 (assumed to be equal to 0), the calculated slope *b* and the variables *x* and *y*, the test statistic is defined by:

$$t = \frac{b - \beta_0}{s \sqrt{s_{xx}}}$$

where

$$s^2 = \frac{s_{yy} - bs_{xy}}{n-1}$$

and

$$s_{xx}^{2} = \sum_{i=1}^{n} (x_{i} - \overline{x})^{2}$$

$$s_{yy}^{2} = \sum_{i=1}^{n} (y_{i} - \overline{y})^{2}$$

$$s_{xy}^{2} = \sum_{i=1}^{n} (x_{i} - \overline{x})(y_{i} - \overline{y})$$

In this case, the variable x refers to the time in days from the earliest sample date and y is the SF logarithmic value. Since this test is performed on log values, the calculated trend slope represents a change in the

log value per day. To convert to per cent change per year, the slope (m) is multiplied by the number of days in a year and applied as a power to the log base of 10 (i.e. $10^{mt} \times 100$).

A-20.5. Analysis standard

From the US Nuclear Regulatory Commission Branch Technical Position on Waste Classification [A-11]:

"The staff considers a reasonable target for determining measured or inferred radionuclide concentrations is that the concentrations are accurate to within a factor of 10."

Since the analysis deals generally with two sided variations, differences should be limited to a factor of 5. In fact, across the board differences can be limited to a factor of 3 or less when waste streams are judiciously combined.

A-20.6. Updating of scaling factors

US regulatory guidance suggests updating SFs every year, or with each operating cycle of the reactor for higher activity waste, and every other year for lower activity waste. As US plants have moved to longer operating cycles, the intervals between SF updates have on average increased. In most cases these are running at between 1.5-2 years. Usually when updates are performed, a complete set of samples is sent to a radiochemistry laboratory for analysis. When results are returned and accepted by the plant operations staff, these are used either to directly replace the current set of SFs, to verify that the new sample results are consistent with the SFs being used — invoking no change to the existing SFs — or folded into a sample database to generate a new three or five year running average that will replace the existing SFs.

With the 25 years of experience in the US in collecting radiochemistry sample data, some SF ratios have been shown to be stable over the entire period of operation, even with relatively dramatic changes in plant conditions and operating practices. Ratios between transuranic nuclides adapt to signature values and remain almost constant both across waste streams and over extended time periods.

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ABBREVIATIONS

ALARA	as low as reasonably achievable
ANDRA	National Agency for Radioactive Waste Management
AP	activation product
CC	correlation coefficient
CEA	Commissariat à l'énergie atomique
CNSC	Canadian Nuclear Safety Commission
СР	corrosion product
DAW	dry active waste
DGR	deep geological repository
DTM	difficult to measure
EDF	Electricité de France
EPRI	Electric Power Research Institute
ETM	easy to measure
FP	fission product
HLLW	high level liquid waste
HLW	high level waste
HWGCR	heavy water moderated gas cooled reactor
ILW	intermediate level waste
ISO	International Organization for Standardization
LD	log dispersion
LILW	low and intermediate level waste
LLW	low level waste
MDA	minimum detectable activity
MOX	mixed oxide
NSDF	near surface disposal facility
OPG	Ontario Power Generation
RWCU	reactor water cleanup
SGS	segmented gamma spectrometry
VLLW	very low level waste

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