

Activated Waste Activity Assessment Based on Radioactive and Non-Radioactive
Measurements: Generalised Data Assimilation Method – 11135

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ABSTRACT

Any type of waste repository, of interim storage or even any type of facility devoted to treatment of radioactive materials, needs to know the radioactivity of the nuclides. The reason is to prove the compliance with limits and criteria derived from its safety assessment.

When the radionuclides (RN) are coming from a contamination process, which represents the most part of waste generated during operation of nuclear power plants, numerous methods have been developed. These methods are empirical because no general and suitable contamination model could possibly match with all the possible contamination configurations. When the RN have been produced directly by activation by a neutron flux without any migration phenomenon occurring between its formation and its measurement in the solid waste, we can talk about "activated waste". Starting from knowledge of neutron flux cartography, physics allows to solve the identification problem of activation model. On basis of a limited number of measurements of radioactivity and/or of chemical composition, the resulting calculation with "Data Assimilation" or "Melding" is optimal with respect to measures available

"Data Assimilation" or "Melding" is an method of identification which gives a general tool to assess Scaling Factor (SF) values and uncertainties. This tool allows to solve every activation problem including the problems of nuclear dismantling. The reason is that, unlike the domain of activated waste generated repetitively by the operation process, no empirical method for activated waste in dismantling is completely general and suitable. By example, for graphite bricks or for metallic structures as a vessel, one can use neither correlation factor nor index of burnup in order to get any appropriate activity assessment. Anyway, a scaling factor may be calculated for each corresponding object, but as the most robust result of the general "Data Assimilation" or "Melding" tool.

This paper gives guidelines for the general basic tool to calculate the scaling factor (SF) of any RN generated directly by a known neutron flux. Including a known history of that flux and excluding any RN migration between its formation and its measurement in the solid, this activity is from simple activation. It gives guidelines to calculate on the basis of a limited number of measurements of radioactivity and of chemical composition, the neutron flux and its history being known. It consists in using physics laws and experimental measurement simultaneously and is also relevant to compute a confidence interval as a simple byproduct.

INTRODUCTION

"Data Assimilation" method itself is explained in "restricted" and "general" versions to show how explanatory compositions and their uncertainties are estimated. Then, the use of these explanatory compositions allows the global calculation of inventories for each RN by incorporating the calculation of uncertainties and the detailed calculation of SF in relation to RN tracers (or key RN).

In order to treat radioactive waste, we must be able to quantify both the impact on health due immediately to the treatment (incineration, melting, recycling, etc.) and differed impact (shallow or deep geological storage of waste concerned as ultimate).

There are two categories of treatment:

- treatment of radioactive waste preliminary to the direct shallow or deep geological storage of residues
- direct shallow or deep geological storage which requires an assessment of the immediate impact on health during the operation of the storage facility and an assessment of the differed impact on health and environment after closure, which is due to the maximum limits of specific activity and/or total activity of each RN according to the possible return to man and environment scenarios.

The safety analysis of the treatment of radioactive waste leads to the obligation of quantifying the health impact for man and environment. For this, knowledge of RN is essential and this paper covers the ways and means to access knowledge on RN produced by neutron activation and which are not subject to migration by diffusion in a nonporous solid or transported by a fluid (liquid or gaseous). In this paper, the term "activation" covers all the transformations in a neutron flux of the nuclei of the initial chemical compositions, including the formation of fission products and heavy nuclei from the fissile impurities of materials, remembering that their role as neutron sources is negligible.

On the basis of the non-destructive measurement of a RN tracer (or key RN), the application of a scaling factor (SF) may be an effective strategy. This paper gives later the means for determining this SF and its confidence interval. Indeed, we shall see that, by construction of the SF, the fact of multiplying the SF by the overall activity of the key RN which is easy to measure by a non-destructive method as a gamma emitter, gives precisely the activity of the RN which cannot be measured by a non-destructive method, but only for its overall inventory.

CHARACTERIZATION OF RN DUE TO ACTIVATION

Characterisation is expressed according to three conventional approaches: (1) an exclusively calculation mode (theoretical estimation), (2) an exclusively experimental mode (measurements in situ or on samples), (3) a mode combining the two approaches by optimising with calculation-measurement adjustment (data assimilation).

Set of hypotheses and use of the laws of physics (calculations)

The determination of the activity of RN is based on the calculation carried out on the basis of a given set of hypotheses, the quality of which is mainly based upon the user's expertise in the method of calculation used and the sensitivity of the parameters. These two aspects justify that, during a later phase, we are often asked by the regulator to validate these hypotheses by comparing the results obtained with the measurements made in situ or on samples.

The quality of the results is therefore based on the set of hypotheses used for the input data during the digital simulations and therefore, the simplifications that had to be made, whether deliberately or not (lack of data, finesse of calculations, etc.). The main limit to this characterisation is our lack of knowledge concerning impurities; that is, compositions in the form of uncontrolled traces, but the activation of which is often a determining factor in the radioactivity of waste, due to a very large cross section and/or significant neutron flux.

Approach limited only to the experimental phase (measurements)

Determination is based only on measurements. Depending on the nature of the measurements (and therefore of the radiations detected), we are limited to the RN that can be measured. In addition, the detection limits which may be poor with regards to the expectations of the disposal. It is also clear that the purely experimental approach may only concern a limited number of sample. The question of the representativeness of the measurements is therefore recurring, just like the extrapolation of several points to all waste or of several RN to all RN required.

The last point concerning RN difficult to measure or even impossible to measure (under current radiochemical knowledge) shows the limits of this approach. Indeed, in this case, the use of scaling factor is meaningless, since the purpose of this approach is to calculate such SF and cannot therefore use it as a hypothesis ...

Interpretation by identification of calculations and measurements (Data Assimilation or Melding)

Given the respective shortcomings of the two previous methods, the Data Assimilation (or Melding) illustrated in the diagram below enables us to exploit the measurements and calculations to the full. When the determination combines the calculation with the results of the measurements, the final quality of the results is in this case, directly associated with the process of adjustment which guarantees the performance and the number of points on which the calculation-measurement adjustment is carried out. In principle, this approach combines the advantages of the two previous approaches and may be seen as the optimum of the three. We shall also see that it provides, as a by-product and with no specific effort, the means of quantifying uncertainty.

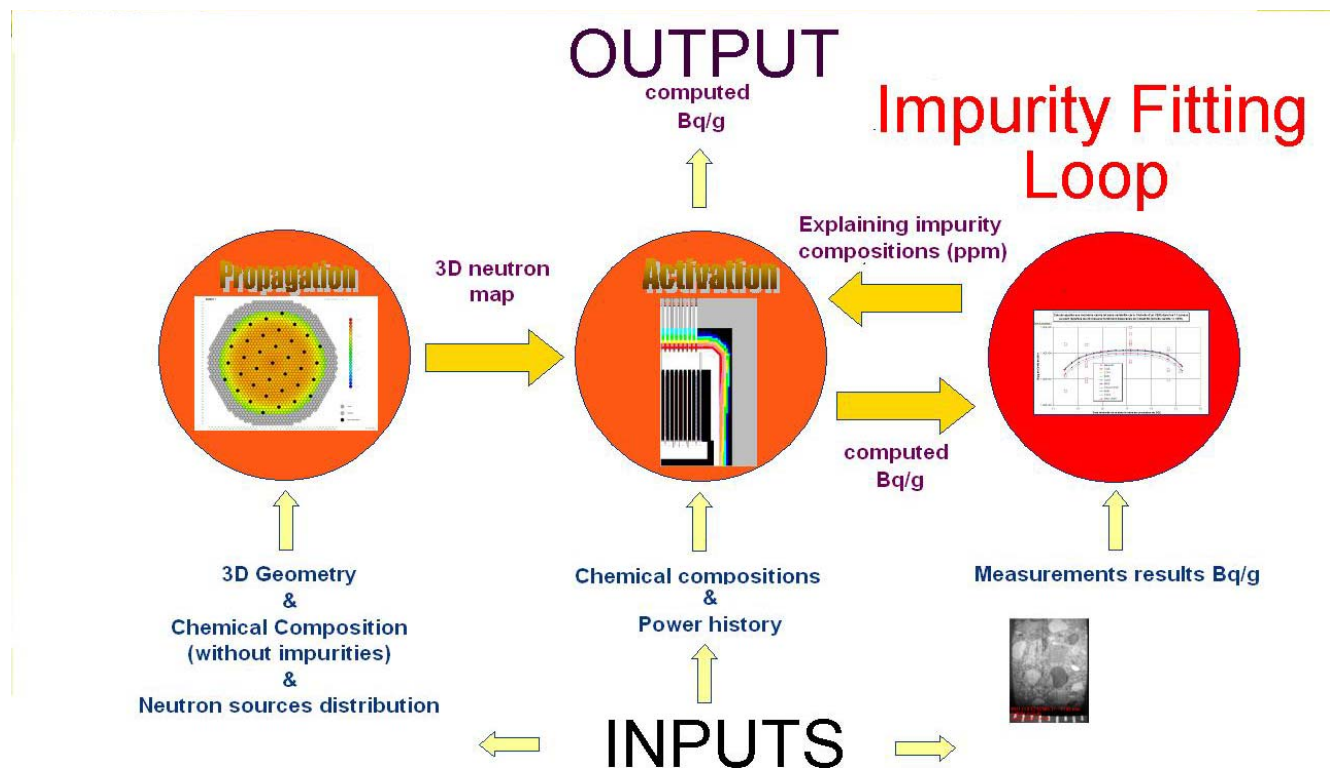


Figure 1 — Data Assimilation ("restricted" version): Adjustment of impurities by identification of measurements and calculations of RN

STOCHASTIC REPRESENTATION OF THE IMPURITIES

In order to take uncertainties into account conservatively, the only final result possible corresponds to the upper boundary of the confidence interval of the average specific activity for a given risk of underestimation. We shall see that heterogeneity may exist on a global scale for locally homogeneous materials (such as metal alloys for example) and locally for inherently heterogeneous materials (such as graphite and/or concrete). Therefore, given the random nature of the heterogeneity of real materials which make them inaccessible for deterministic prediction, modelling can only be stochastic.

Central Limit Theorem

The principle of the statistical processing of measurements consists in applying the Central Limit Theorem (CLT) and in deducing average values by giving them a confidence interval. It should be remembered that this theorem guarantees the convergence of the average of N random variables (independent and according to the same law) with a normal law (i.e. Gaussian). Conventionally, the littlest minimum population for application of the CLT corresponds to $N_{\min} = 6$ but we will see that the more N is low, the more the confidence interval is broad. Since N is equal to or greater than N_{\min} and since a value for the risk of underestimation α has been adopted (for instance $\alpha = 2,5\%$), a practical corollary of this theorem is used to generate a confidence interval (at $1 - 2\alpha = 95\%$ of confidence in this case) of this average in the following form:

- a value which we shall call "best estimated", \bar{X} which is the average of N applications X_n of these variables
- an absolute uncertainty calculated as the product of Student's variable with N-1 degrees of freedom and which corresponds to risk α selected, with the standard deviation of this mean $\Delta_\alpha X$

$$\Delta_\alpha X = \lambda_{(\alpha, N-1)} \sqrt{\frac{\sum_{n=1}^{n=N} (X_n - \bar{X})^2}{N(N-1)}} \quad (\text{Eq. 1})$$

$$\sigma_X = \sqrt{\frac{\sum_{n=1}^{n=N} (X_n - \bar{X})^2}{N-1}} \quad (\text{Eq. 2})$$

$$\sigma_{\bar{X}} = \frac{\sigma_X}{\sqrt{N}} \quad (\text{Eq. 3})$$

The graph below illustrates the diminishing return of measurements. We note that the gain in precision is significant between $N = N_{\min}$ and $N = 20$ but becomes very low beyond $N = 30$. Indeed, Student's variable with 95% of confidence ($\alpha = 2.5\%$) is close to 2 around 29 degrees of freedom (whilst its asymptotic value is 1.96) and the gain in terms of precision provided by \sqrt{N} is approximately 1.6% for taking $N = 30$ to $N = 31$; that is, slightly significant, not more.

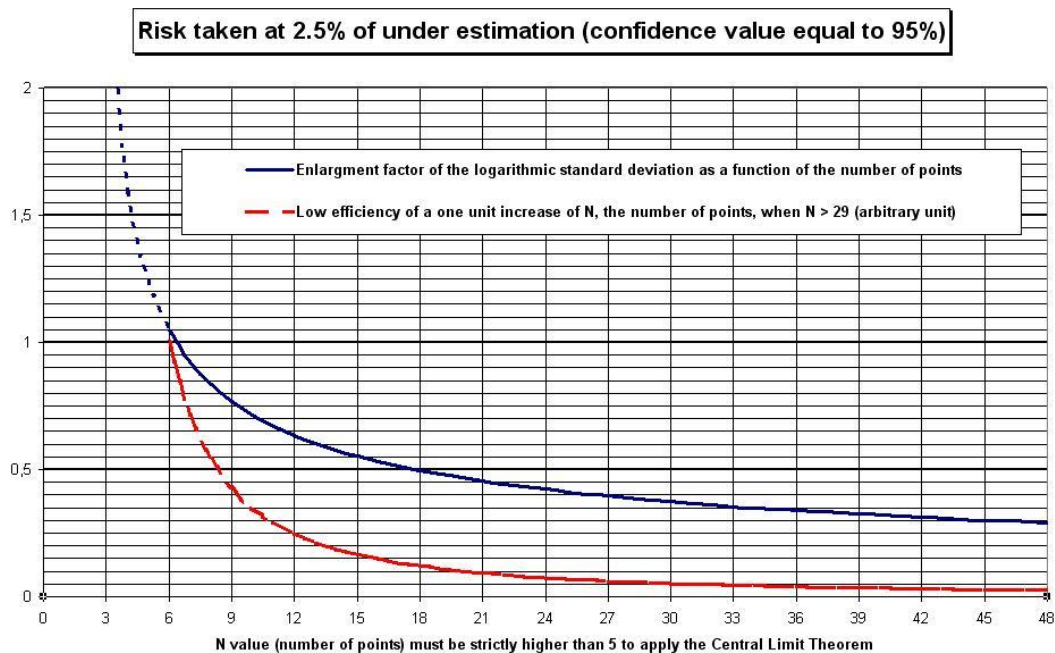


Figure 2— N being the number of points of the calculated average, the graphic display the factor of uncertainty for a 2.5% risk

$$\text{Factor of uncertainty} = \frac{\lambda_{(\alpha, N-1)}}{\sqrt{N}} \quad (\alpha = 2.5\%) \quad (\text{Eq. 4})$$

It should be noted that below $N = 6$, the uncertainty factor is too high to be of any meaning, since the dependency of the choice of one point rather than another is too great. We note after $N = 30$ that the gain provided by an additional point is negligible and that the fact of removing a point has no effect on the result, which is the best proof of robustness.

Adaptation to fundamentally positive data

Given the nature of normal distribution, the stochastic modelling of the absolute uncertainty of the average in the form of normal distribution leads to granting positive probability to negative values of the average since the Gaussian law extends from $-\infty$ to $+\infty$ (this is paradoxical but assumes all its meaning if the standard deviation is not negligible in front of the average). This modelling is of no use for concentrations of chemical elements or specific activity.

This simply means that, if the standard deviation of the population of measurements is not small compared to the average, the estimate of "absolute" uncertainty is meaningless and only an estimate of "relative" uncertainty is meaningful. Furthermore, switching to relative uncertainties enables us to compare the results for various RN and the various impurities which produced them. Also, the fact of giving the same weight to high and low levels of activation is a considerable advantage.

To obtain all the results of these findings, just **change the scale** (note, this is not a change of variable) by switching to the logarithms of the values of the measurements. With these new random variables, the Central Limit Theorem may be applied in the following form of a value which we shall call "centred logarithm", average value of the N logarithms of the instantiations of these random variables. This corresponds to a geometrical average, the result of the change of logarithmic scale:

- absolute uncertainty of the logarithms calculated as the product of Student's variable with N-1 degrees of freedom (and which corresponds to the risk α selected) with the standard deviation of this average.
- the standard deviation of this average is calculated as an unbiased estimate of the standard deviation of the population of the N logarithms of the values divided by \sqrt{N} .

Since the first order development shows that the "absolute" uncertainty of the logarithm of a positive measurement coincides with the "relative" uncertainty of this measurement, we note that we recover the first order meaningful Gaussian distribution of real positive and negative values, i.e. for "minor" uncertainties and we limit the probability distribution to positive values, which makes the approach using CLT in a positive context, applicable.

In this case, the "best estimated" value is replaced by the geometrical mean average of the population of measurements. The upper boundary of the confidence interval $B_{\text{sup},\alpha}$ corresponds, for a given risk of underestimation α , to the multiplication of the geometrical mean average by a factor F_α ; this factor is the exponential function of the absolute uncertainty of the logarithms.

$$B_{\text{sup},\alpha} = \left(\prod_{n=1}^{n=N} X_n \right)^{\frac{1}{N}} \times F_\alpha \quad (\text{Eq. 5})$$

The lower boundary of the confidence interval $B_{\text{inf},\alpha}$ corresponds, for the same risk α of over estimation, to the division of the geometrical mean by the same factor F_α .

$$B_{\text{inf},\alpha} = \frac{\left(\prod_{n=1}^{n=N} X_n \right)^{\frac{1}{N}}}{F_\alpha} \quad (\text{Eq. 6})$$

For the sake of thoroughness, we must also take into account the uncertainty on the logarithm of the measurement which is due to the uncertainty of the very process of measuring chemical concentration or radioactivity. The uncertainty of logarithm Δx_n when it is calculated with a coefficient of 1.96 which corresponds to the choice of a level of confidence, namely $1 - 2\alpha = 95\%$ in the most common case of chemical or radiochemical measurements, is added "quadratically" to that due to the dispersion of the population of measurements.

$$\Delta_\alpha x = \frac{\lambda_{(\alpha, N-1)}}{\sqrt{N}} \sqrt{\frac{\sum_{n=1}^{n=N} (x_n - \bar{x})^2}{N-1} + \sum_{n=1}^{n=N} \left(\frac{\Delta x_n}{1.96} \right)^2} \quad (\text{Eq. 7})$$

Chemical elements of metal alloys

Because it goes through a liquid phase, the Representative Elementary Volume (REV) of a metal alloy is particularly small. The chemical analyses of the metal alloys are not affected by variability due to inherent heterogeneity, but only to variability due to the uncertainty of measurement, on the one hand, and on the other, variability due to a change of ingot or to a change of the casting phase of the metal alloy used.

In alloys, there are two categories of chemical elements: (1) Those the dosage of which is controlled by the manufacturer in accordance with specifications of chemical and/or mechanical properties. Nevertheless, the mean or maximum mass concentration may not be known if the manufacturing record has been lost; (2) Those the dosage of which is not controlled and the value of which remains random according to the ingot or casting phase. For such impurities, even if the manufacturer guarantees that their total sum remains below a given mass concentration limit, their individual, even mean, quantification must involve a stochastic approach. It may be noted that concentrations of certain impurities (uncontrolled) may be due to certain controlled elements and that correlations (in the strict, that is stochastic, meaning of the term) may be found between controlled elements and uncontrolled impurities.

Example of cobalt in steels

Cobalt knowing is important in a context where Co-60 is used as an RN tracer (key RN), since its uncertainty will directly affect that of the scaling factor (see below). It should therefore be noted that to increase the chance of exploring different ingots or casting phases, a given part must only be sampled once. Given the homogenisation obtained by going through the liquid phase (during the manufacture of the ingot for example) and, therefore, that the sample always exceeds the REV, the origin of any variation in the two samples taken from the same part of reasonable size, could only be due to the uncertainty of the measurement. The only opportunity of collecting independent random variables requires samples from different parts or, for very large waste, parts of components from different casting phases.

Example of chlorine in steels

Chlorine (Cl), as an impurity in the form of traces would appear to be never due to deliberate control. It would therefore appear to be difficult to make a statistical study of it whilst remaining within the scope of a single grade and/or supplier of the alloy. Another justification lies in the fact that this measurement is difficult and rare. In this case, we simply consider steels in general since, for example, the stainless nature of steel or not would not appear to have any reason to affect the presence of chlorine. Whilst several hundred ppm are negligible with regards to the concerns of metallurgists, the opposite is true for RN produced by activation since very small quantities of chlorine (1 or 2 ppm) are enough to produce Cl-36 (from the stable isotope Cl-35) in significant quantities in terms of the regulatory limits of a shallow or deep geological storage facility (high cross section and/or neutron flux).

Elements of inherently heterogeneous materials

These materials are concrete or graphite for example. Their method of manufacture using respectively, a known formulation or a given manufacturing process, on the basis of a given petroleum coke, a given type of purification and impregnation of tar, enables us to consider only one category of concrete or graphite for a given facility.

By nature, the Representative Elementary Volume (REV) of an inherently heterogeneous material is large. To avoid being affected by variability due to inherent heterogeneity, chemical analyses should be carried out on samples larger than the REV. If the size required is too large for the analysis, a sufficient number of samples smaller than the REV should be processed to obtain significant average values; that is, values which could be obtained with a volume greater than the REV.

RN MEASURED TO DETERMINE THE EXPLANATORY COMPOSITIONS

RN of metal alloys

Like chemical analyses, the radiochemical analyses of metal alloys are not affected by variability due to inherent heterogeneity, but to the uncertainty of measurement and/or to variability due to a change of ingot or casting phase of the metal alloy. It should be pointed out that for certain parts (i.e. activated waste generated during operation) the irradiation history (and particularly the various positions in the pile) is a parameter in its own right which must be known specifically. Finally, processing a sufficient number of samples as independent as possible (at least on separate metal parts) is unavoidable.

RN of inherently heterogeneous materials

These materials may be concrete or graphite. Their method of manufacture using a known formulation or a given manufacturing process respectively, on the basis of a given petroleum coke, a given type of purification and impregnation of tar, enables us to consider only one category of concrete or graphite for a given facility. Like chemical analyses, to avoid being affected by variability due to inherent heterogeneity, radiochemical analyses should be carried out on samples larger than the REV. It should be pointed out that for certain elements (i.e. fuel graphite sleeves), irradiation history (and particularly the various positions in the pile) is a parameter in its own right which must be known specifically. With respect to an irradiated material, such as graphite or concrete, for almost all measurable RN, it is not possible to perform radiochemical analyses on samples of the size of the REV and in this case, a statistical approach is the only possible one.

CHEMICAL COMPOSITIONS MEASURED DIRECTLY

Compositions based on the processing of available statistics

When determination via the activation calculation is not possible, we must use "a priori" knowledge of compositions by statistical methods on positive measurements, as described above in order to quantify mean average penalising compositions that correspond to the upper boundary of the confidence interval adopted with specific quantification of the risk of under estimation.

Consensual compositions from the literature

If the theoretical knowledge does not involve sufficient numbers, we must use values considered to be conservative by all those concerned (consensus is only valid if no other rigorous method is available). More often than not, such values are taken from the literature.

COMPOSITIONS FITTING BY DATA ASSIMILATION OR MELDING SCHEME

General explanation of the method

We shall see that the adjustment or data assimilation consisting in using $\frac{\textit{calculation}}{\textit{measurement}}$ ratios enables us to "withdraw" the effects of the activation flux where the measurement is taken to "retain" only the effect of the chemical composition which produced the RN and the variability of which is stochastically modelled. It should be noted that this stochastic modelling may give rise to two physical explanations specific to two types of materials:

- Materials that are constant on a large scale because they are due to a controlled manufacturing but which, by nature, are highly heterogeneous on a small scale (graphite and/or concretes). Concentrations on a scale smaller than the REV may only be modelled by a random variable.
- Materials that are homogeneous on a small scale but subject to random variations, as with uncontrolled compositions, due to different origins which correspond to different ingots or casting phases for metal parts made of the same or a similar alloy. Owing to lack of sufficient information, concentrations of uncontrolled compositions of such alloys may also only be modelled by a random variable.

There are of course cases where the chemical elements at the origin of the RN measured may also be measured. In some conditions, as set out below, the Data Assimilation or Melding method may also summarise the data from these two sources. There are therefore two distinct versions of RN measured: (1) "Restricted" version of the "non mixed" RN measured which does not correspond to any chemical element measured which would "explain" this RN production, (2) "General" version of the "mixed" RN measured which corresponds to at least one chemical element measured which would "explain" this RN production.

"Restricted" Melding scheme

In this "restricted" configuration, the term "calculation" means "the calculation of activity due to activation" on the basis of "explanatory" chemical elements given by "calculation-measurement" identification. On the basis of the elements obtained by direct measurement, we make direct activation calculations. Since by definition, these two sets of elements are disjoint, there is no confusion possible for the Data Assimilation for these RN, none of which are "mixed". In order to express the equality of two populations of N measurement and calculation points of a RN, even in a stochastic context, we must, and we only need to, establish the equality of the statistical averages (that is, make the differences between the average of the calculations and the average of the measurements equal to zero). We saw in the previous chapter that the statistical estimation of the average which remains meaningful for essentially positive values, such as radioactivity, involves a change of scale:

Just use the logarithms of the values calculated or measured. Therefore, the only meaningful differences between calculations-measurements are differences of logarithms; that is, the population of the logarithms of $\frac{\text{calculation}}{\text{measurement}}$ ratios of the same RN for the same calculation and measurement date. We know that the minimisation of the sum of the square roots of the differences involves identifying the two averages, which are averages of logarithms, which involves identifying the geometrical averages of values.

$$\bar{x} = \frac{1}{N} \sum_{n=1}^{n=N} \ln \frac{C_n}{M_n} \quad (\text{Eq. 8})$$

The adjustment of the initial chemical composition enables us to obtain the following where $\bar{x}_{RN \text{ measured}}$ is equal to zero for all RN which we shall call "simple adjusted RN" or in the worst case, enables us to minimise the norm of the vector subject to $\bar{x}_{RN \text{ measured}}$ kept positive

We therefore obtain the best solution for the wanted result of the identification of geometrical averages. By applying the techniques described above, we quantify the relative error of identification of the adjusted RN. For a simple adjusted RN, the relative error is

$$\Delta_{\alpha} x = \frac{\lambda_{(\alpha, N-1)}}{\sqrt{N}} \sqrt{\frac{\sum_{n=1}^{n=N} \left(\ln \frac{C_n}{M_n} \right)^2}{N-1} + \sum_{n=1}^{n=N} \left(\frac{\Delta[\ln M_n]}{1.96} \right)^2}. \quad (\text{Eq. 9})$$

Identification $\frac{\text{calculation}}{\text{measurement}}$ by adjustment of "observable" compositions is an expression synonymous with the "restricted" Data Assimilation or Melding. Since concentrations of certain impurities may be too low to be accessible to direct chemical measurements, they may be obtained by indirect determination using RN produced by neutron activation. In this case, the identification of the calculations and measurements of all RN measured is carried out by adjustment of the mean "observable" chemical compositions.

In this case, the process of identification is used to adjust the "observable" compositions to values known as "explanatory compositions" because these composition values "explain" the activation calculations in that the average of the calculations is made equal to the average of the measurements. It should be noted that this can only be an average explanation of a phenomenon, the stochastic nature of which means that it cannot anyway be explained from a deterministic point of view (principle of uncertainty). This process matches exactly a methodology for Data Assimilation using the activation model provided by the Bateman's equations, the criterion (to minimize) which corresponds to the sum of the squares of the differences of the averages of the logarithms of the calculations and measurements of each RN. For any given RN, it should be remembered that the uncertainty of adjustment $\Delta_{\alpha}x$ on N points is defined as the product of Student's variable with N-1 degrees of freedom (and which corresponds to the risk α of under estimation) with the standard deviation of the average of the logarithms of the N $\frac{\text{calculation}}{\text{measurement}}$ ratios of the RN.

Finally, $\Delta_{\alpha}x$ depends neither on flux conditions nor history, but only on the adjustment of compositions. The confidence interval of activity C averaged for a volume greater than the REV of the material, encompasses value $C_{\text{calculation}}$, calculated by the activation code using the flux conditions, irradiation history and the "adjusted" values of the compositions: C is in the interval $[C_{\text{calculation}} \cdot e^{-\Delta_{\alpha}x}, C_{\text{calculation}} \cdot e^{+\Delta_{\alpha}x}]$.

Therefore, for the quantification of the confidence interval of an RN to be possible, it is enough that a population of this RN is strictly greater than $N_{\text{min}}-1$ points ($N > N_{\text{min}}-1$) for the use of the CLT to be sufficient (general case making the use of (Eq. 9) meaningful for a simple adjusted RN) or a small uncertainty of adjustment $\frac{\text{calculation}}{\text{measurement}}$ compared to the uncertainty quadratic mean of measurement to the same level of confidence. Indeed, in this case, the adjustment may be considered done (very special favourable case where there are no problems of adjustment because the analyses were carried out on samples of the magnitude of the REV since in this case,

$$\Delta_{\alpha}x \approx \frac{\lambda_{(\alpha, N-1)}}{\sqrt{N}} \sqrt{\sum_{n=1}^{n=N} \left(\frac{\Delta[\ln M_n]}{1.96} \right)^2} \quad (\text{Eq. 10})$$

For metal alloys, the special very favourable case may correspond to the very high quality of control of a chemical element of the composition, a particularly homogeneous set of castings and/or ingots for the grade concerned or to a particularly high uncertainty of measurement for the RN concerned (this is a very artificially "favourable" case since it leads to increasing the estimated activity of the waste)

It should be noted that the only special case possible for graphite or concrete (inherently heterogeneous materials) can only correspond to uncertainties of measurement for the RN concerned that are so high that they may mask the inherent variability of the material. In this case, it is difficult to speak of favourable cases.

At the limits of the statistical approach, if we do not have a special favourable case, it is difficult to come to a result obtained with $N < N_{\text{min}}$ unless we give up any quantification of the uncertainty due to the chemical elements concerned and we simply compare the adjustment of the composition with the direct measurements available for the chemical elements concerned.

"General" Melding scheme

In this "general" configuration, the term "calculation" means "calculation of activity due to activation" on the basis of the "explanatory" chemical elements given by "calculation-measurement" identification or on the basis of data obtained by direct measurement. By definition, these two sets of data have common explanations for "mixed" RN and it is these RN that we shall consider. On the basis of data obtained by direct measurement, we **calculate** a "truncated measurement" for the RN, limited to the activation calculation of the predominant element with its "**measured**" value. Be careful, "truncated measurement" is the result of a calculation of RN starting from a measurement of impurity which explanatory mean value is looked for. With respect to this "truncated measurement", we make a "truncated calculation" of the RN limited to the activation calculation of this elements measured with its "**adjusted**" value. It should be noted that the calculated "truncated measurement" of the "mixed" RN does not use values from measurements of any other chemical element.

This missing complementary term may be calculated from both "adjusted" and "not adjusted" elements. It must not exceed the overall uncertainty of the "truncated measurement" which incorporates the uncertainty of calculation of the activation parameter and the uncertainty of measurement of the corresponding predominant explanatory element. If this is the case, we consider that the potential error given by the "truncated measurement" is too large and we do not take this "truncated measurement" into account for the final adjustment. In this case, the measurement of the underlying concentration is not therefore taken into account. For the RN measured which do not depend on the definition of "mixed" RN, we simply apply the "restricted" Data Assimilation scheme.

In order to express the equality of two double populations characterising a "mixed" RN, even in a stochastic context:

- of N1 measurement M_n and calculation C_n points (points similar to the restricted version),
- of N2 "truncated measurement" \tilde{M}_n and "truncated calculation" \tilde{C}_n points (generalisation),

we must, and we just need to, make the differences between the average of the calculations and the average of the measurements equal to zero. We have seen that the only meaningful calculation-measurement differences are the differences in logarithms, that is, the population of the logarithms of the ratios $\frac{\text{calculation}}{\text{measurement}}$ or $\frac{\text{truncated calculation}}{\text{truncated measurement}}$ of the same "mixed" RN with the same calculation and measurement date, provided that these ratios remain independent.

For example, one cannot use the measurement of Co-60 in the acid solution by gamma-ray spectrometry and the measurement of Co of the same solution by mass spectrometry, at the same time, to adjust the Co. Nevertheless, these two redundant measurements confirm the level of uncertainty of the calculation of flux, which may be useful.

According to

$$\bar{x} = \frac{1}{N1 + N2} \left\{ \sum_{n=1}^{n=N1} \ln \frac{C_n}{M_n} + \sum_{n=N1+1}^{n=N1+N2} \ln \frac{\tilde{C}_n}{\tilde{M}_n} \right\} \quad (\text{Eq. 11})$$

the same kind of adjustment of the initial chemical composition as in previous case is performed

For a simple adjusted "mixed" RN, the relative error is only a generalisation of (Eq. 9). Let consider also the case of no activity measured: As we have $\frac{\textit{truncated calculation}}{\textit{truncated measurement}} = \frac{\textit{adjusted element}}{\textit{measured element}}$, the measured element being the predominant one, if no activity is measured, we find again (as seen above) the calculation of the adjusted value of the chemical element as the geometric mean average of its direct measurements including the calculation of its confidence interval for a given risk. By simple generalisation, the identification of $\frac{\textit{calculation}}{\textit{measurement}}$ and/or $\frac{\textit{truncated calculation}}{\textit{truncated measurement}}$ by adjustment of "observable" compositions is an expression synonymous with the "general" Data Assimilation or Melding.

In this case, the process of identification is used to adjust the "observable" compositions to values known as "explanatory compositions" because these composition values explain the activation calculations where the average of calculations including truncated calculations is made equal to the average of measurements including truncated measurements (it is the way to merge activity data with chemical composition data).

For a given "mixed" RN, it should be remembered that the uncertainty of adjustment $\Delta_{\alpha}x$ on $N=N1+N2$ points is defined as the product of Student's variable with $N-1$ degrees of freedom (and which corresponds to the risk α of under estimation) with the standard deviation of the average of the logarithms of the N $\frac{\textit{calculation}}{\textit{measurement}}$ and/or $\frac{\textit{truncated calculation}}{\textit{truncated measurement}}$ ratios of the "mixed" RN. For the other RN, the "restricted" definition applies.

Finally, in all cases, $\Delta_{\alpha}x$ depends neither on flux conditions nor history but only on the adjustment of "observable" compositions. In all cases, the confidence interval of activity C when it is averaged for a volume of the same order of magnitude than the REV of the material, encompasses value $C_{\textit{calculation}}$, calculated by the activation code using the flux conditions, irradiation history and the "adjusted" values of the compositions. C is in the interval $\left[C_{\textit{calculation}} \cdot e^{-\Delta_{\alpha}x}, C_{\textit{calculation}} \cdot e^{+\Delta_{\alpha}x} \right]$.

Therefore, we can extend the "restricted" approach for the quantification of the confidence interval of a "mixed" RN.

Mapping of the levels of activity calculated for each RN

With a risk of underestimation selected, we have seen that the only final result possible corresponds to the upper boundary of the confidence interval of the average specific activity. This "envelope" calculation includes all specific activities of each RN for each volume (cell) of spatial computation. The determination of this spatial distribution is based on the evaluation of "best estimated" activation. It stems from the use (often massive via computer clusters) of an activation code:

- the initial "best estimated" compositions of which were determined indirectly by the methods of adjustment ("restricted" or "general" Data Assimilation) described above
- the initial, directly available compositions of which have been quantified as geometric mean average or, if this is not possible, as consensual penalising values which usually include the uncertainty.

ASSESSMENT OF UNCERTAINTIES

As seen above, directly available compositions have been quantified as geometric mean averages. The uncertainty of them has been explained to calculate their confidence intervals.

Uncertainties of adjusted compositions

We have seen that the adjustment of "explanatory chemical elements" was expressed in terms of relative uncertainty for a RN measured. This term of uncertainty (by-product of the method of adjustment), enables us to calculate, initially, upper limits for the relative uncertainties of the elements of the explanatory composition adjusted by considering the worst case, that each is the cause of all relative uncertainty found experimentally.

The method may, naturally, be improved to produce lower upper uncertainties. For this, we solve conventional error calculation equations to return to the relative uncertainties for each item of adjusted explanatory chemical element. This implies taking into account several contributions to the same RN from different adjusted chemical elements. The fact of considering the contribution of non-adjusted data as non-existent is an inevitable penalty.

Uncertainties due only to the calculation

Sources of uncertainty affecting the flux and activation calculations (inaccuracy of cross sections, simplification of geometry and/or sources) should be assessed in the form of relative uncertainty due only to the calculation. For an adjusted RN, the quality of identification of the average of the calculations with the average of the measurements enables us to incorporate all uncertainty, by allocating all sources of uncertainty of calculations to the "explanatory" composition.

For a RN not measured, the identification of the average of the calculations with the average of the measurements of RN which have the same "explanatory" chemical element in common, allows for the propagation of the resulting uncertainty. Nevertheless, if there is a non-adjusted chemical element value which makes a significant contribution to this non-adjusted RN, it will not be possible to consider that the uncertainty of adjustment of explanatory compositions includes the uncertainty of unmeasured RN. We should consider the greatest relative uncertainty between adjusted relative uncertainties and the relative uncertainty adopted only for the calculation.

Uncertainty of the final result

The relative uncertainty of the RN calculated for each computation volume or cell may propagate from:

- the relative uncertainty of the elements that produce the RN
- the relative uncertainty due only to the calculation.

SCALING FACTOR CALCULATION IN RELATION TO A KEY RN

Unlike purely stochastic methods used to quantify RN that are difficult to measure using the measurement of a gamma tracer RN (or key RN), it is not possible to use the term "correlation factor" as it is very often correct to do so in the context of contaminated waste. Since the overall gamma key RN inventory for the whole facility shall be assessed by adding the inventories together, package by package, by a direct gamma-ray measurement, the requirements lie in knowledge of the overall ratio (at a given date) of the global inventories for each RN compared to the overall inventory of the gamma tracer RN.

As a by-product of the inventory calculation, this scaling factor (SF) is in no way a correlation factor but must be referred to as a simple ratio. Its confidence interval must be assessed to be able to use the upper boundary with a given risk α of under evaluation. If A^{RN} and A^{KEY} represent "best estimated" activities from the activation code for each computation of the current RN and key RN, by definition of the "best estimated" scaling factor:

$$SF_{RN} \Big|_{best\ estimated} = \frac{A^{RN}}{A^{KEY}} \quad (\text{Eq. 12})$$

A risk of under estimation α is selected ex ante. The approach for the calculation of each RN in each volume therefore produced a multiplication factor that only depends on the RN to go from the "best estimated" calculation to the upper boundary of the confidence interval. This property is retained by going to the barycentre of these levels of specific activity [Bq/g] weighted by the masses involved, since summation is extended to all the active waste concerned. Let us use the logarithmic uncertainty $\Delta_{\alpha} x$ calculated above and let $\Delta_{\alpha}^{KEY} x$ and $\Delta_{\alpha}^{RN} x$ be the logarithmic uncertainties of the key RN and of any RN, respectively. We obtain:

$$\ln(SF_{RN}|_{best\ estimated}) = \ln(A^{RN}) - \ln(A^{KEY}) \quad (\text{Eq. 13})$$

and therefore a logarithmic uncertainty of at most $\sqrt{(\Delta_{\alpha}^{RN} x)^2 + (\Delta_{\alpha}^{KEY} x)^2}$ if assuming independency between these two RN.

This is not always the case if the element that results in the tracer also produces the RN concerned and, in this case, the assumption of independence is penalising in most practical cases, but we can always refine the calculation of uncertainty using the usual mathematical techniques. Since a value for the risk of underestimation α has been selected, the higher assessment (or "upper boundary") of the confidence interval is therefore as follows, in the worst case:

$$SF_{RN}|_{SUP} = SF_{RN}|_{best\ estimated} \cdot \exp\left(\sqrt{(\Delta_{\alpha}^{RN} x)^2 + (\Delta_{\alpha}^{KEY} x)^2}\right) \quad (\text{Eq. 14})$$

We note that the uncertainty for the adjustment of the impurity which produced the key RN (Co for Co-60) may significantly increase the uncertainty of the scaling factor and therefore increase the assessment of the RN which corresponds to the SF concerned.

CONCLUSION

This paper has given an overview of the characterisation of the radioactive waste and the main pitfalls to be avoided to remain efficient. In so far as the assessment of activity and the corresponding uncertainties concerning RN produced by neutron activation in materials and/or waste from nuclear power plants, a general methodology of "Stochastic Activation Melding" applied to chemical and radioactive data was described.

It explains the principles leading to the best possible exploitation of both chemical and radiochemical measurements, on the basis of a sufficiently precise evaluation of the mapping of neutron flux and of the history of such fluxes.

The final result, RN by RN, or in the form of SF concerning RN tracers, does not give a "dry" value, the meaning of which is always problematical, but the confidence interval indicating the level of uncertainty which characterises this quantification and which makes it credible (a physical value always has an uncertainty) for all possible applications.

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