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An algorithmic software model for the characterization of radioactive waste

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Abstract
Non-consolidated information base, repetitive work, and data unreliability are some of the issues that may hamper a radioactive waste characterization program. Therefore, an algorithmic software model for the characterization of radioactive waste is proposed to boost waste management in nuclear-based facilities. The data obtained have enabled the construction of a guide code in the pseudotechnical language. The developed system fully covered the characterization stage, and met the needs identified in the radioactive waste management service. Here, we propose systematic documentation of the characterization methods that are employed during the radioactive waste management processes, besides formulating the system's data recording.

Keywords Algorithm model · Radioactive waste management · Nuclear data · Characterization

Introduction

The radioactive waste management (RWM) has a particular characteristic that is, in general, absent in the considerations made in engineering projects: the actions carried out must contemplate centuries or millennia ahead, just as the information generated today must be available in equal time horizons. One of the necessary information, if not the main one, is the set of characteristics of the waste, in particular the content of radionuclides. This is what underlies the actions of protecting man and the environment today or in distant futures.

The characterization of radioactive waste, although practiced regularly for operational and radiological safety reasons since the inception of the commercial nuclear era, has only been the subject of specific recommendation and guidelines from the International Atomic Energy Agency (IAEA) more recently, resulting from the need to meet the requirements of quality assurance. The characterization, as a systematic activity of the RWM steps, is therefore still evolving [1, 2]. Yet, some sort of an automatic approach of the characterization process and related activities are scarce in the literature.

The safety standards of the International Atomic Energy Agency (IAEA) recommend that the radioactive content of each waste package be known. With this information, it is possible to classify the waste, define transportation and storage requirements, and immobilize the waste such that the final waste form meets the acceptance criteria for disposal [3, 4]. This recommendation was incorporated into the national regulations [5–7]. The safety analyses of repositories require the knowledge of the radioisotope inventory of all radioactive waste disposed of in the facilities to ensure radiological safety in the long term.

RWM is the set of activities that aims to maintain control over the waste and its associated risks, while reducing costs and simultaneously operational doses during all steps of the handling, including disposal. It is up to the RWM to control this material as long as it constitutes a potential source of danger.

One of the most important steps in the RWM cycle is waste characterization, which is in increasing demand for higher precision and sensitivity of the measurements related to the characterization of a multitude of radioactive wastes. The remarkable technological progress is

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noteworthy in many equipment and routine procedures. Examples of technological progress in this field are detectors, electronics, chemical and radiochemical analyses, and improvements in the calculations related to numerical simulations and data processing [8]. The characterization is done by obtaining the set of physical, chemical, and radiological properties of the waste. This is accomplished in two moments, one that is called primary characterization and the other that is the characterization of the final product of the waste treatment process, or waste form, as presented for disposal. In the primary characterization, the objective is to evaluate the raw waste and obtain the set of characteristics necessary to establish the radioisotopic inventory and the most appropriate treatment method. After the treatment, which usually includes immobilization, the final characterization of waste forms is performed so that the material can be accepted in the repository. The final characterization must ensure that the waste form meets the requirements of the regulatory body regarding disposal [9].

Given the construction of new nuclear-based facilities in Brazil, the main characterization methods must be integrated to facilitate and improve the RWM. The Angra III Nuclear Power Plant, the Brazilian Multipurpose Reactor (BMR) and the National Repository for Low- and Medium-Level Radioactive Waste (RLMRW) are the main nuclear facilities currently under development.

Therefore, the development of a computerized system for the management of the waste characterization stage is crucial not only to facilitate the waste management in the working facilities but also to apply it in new enterprises. It is pivotal that the data is of high quality so that it is possible to characterize varieties of waste and store information [10].

The system must have three main features. The contribution that a computerized system for characterization of radioactive waste brings to the waste management process is: a consolidated information base, reduction of repetitive work, data reliability, control of access to information, traceability, maintenance of long-term information, identification of responsibilities. To ensure the quality of the information generated and the technical specifications of the system, some features are necessary. These features should include a database structure, analysis of the necessary input data and expected output data, integration of the methods of characterization and formulation of the algorithms of these methods, evaluation and definition of access permission levels [11–14]. For instance, Sartori [10] describes four types of data needed in waste management, in which (1−3) are the activation cross sections, fission product yields and decay data libraries, respectively. In (3), the author highlights radioactive isotopes of interest, divided by four main decay chains (Th, Np, U, and Ac). Finally, the (4) is related to the inventory codes.

This paper proposes the investigation and documentation of the characterization methods used in the radioactive waste management process, and the formulation of the system documentation. With the complete documentation, it is expected that it will be possible, in the future, to implement a system that guarantees the quality of the information generated in the characterization step. The system will integrate the characterization methods so that the radioisotopic inventory and the total activity of radioactive waste and facility are supplied reliably and in accordance with the requirements established by the regulatory bodies. Finally, the system may also serve as a reference for application in different industry segments, such as in other nuclear fields, oil industry, and mining.

Methods

The conceptual and logical elaboration of the project consists in the identification of processes and procedures used in the operational routine of the RWM. Special attention is given to those activities used in the Radioactive Waste Management Facility (RWMF/IPEN-SP), in order to raise all the necessary steps for the construction of all modules of a computerized system specific to the activity of characterization of radioactive waste.

The definition of the modules that should be present in the system was done by realizing the flow of commands, information and materials currently used in characterization of radioactive waste. To achieve the development of the system, three lines of search, analysis and construction activities were adopted.

The first line of search was the evaluation of documentation: (a) Published articles; (b) Theses and dissertations on Management of Radioactive Waste; (c) National laws and regulations and recommendations of the IAEA.

The second line of search was the specification of system requirements and modules: (a) Analysis of situation and current process: data collection; (b) Sequencing of the waste characterization procedure; (c) Reporting models required by CNEN and other organizations; (d) Models for storing information.

Finally, the third line of search was the technical specification of the methods of characterization of waste referenced in the modules: (a) Algorithm models specified in pseudo-technical language (Portugol—specification model adopted due to the versatility of subsequent encoding in any standard programming language; (b) Specification of characteristic waste data libraries: Energies, yields, relevant radionuclides, density of the materials, build-up factors, attenuation coefficients; (c) Screen layouts and reports.

The methods that can be used in the radioactive waste management routine can be organized in ten types: (1)

generator information; (2) radiochemical analysis of samples; (3) simple radiometric analysis of samples and/or of the entire packages; (4) application of the dose versus activity relationship; (5) application of scaling factors and correlation functions; (6) gamma-scanning radiometric analysis; (7) tomographic radiometric analysis; (8) passive neutron interrogation; (9) active neutron interrogation; (10) Modeling of the process that generates the waste. From 4 to 9, the methods must be applied all over the packaging. Among the identified methods, the first six (1–6) were implemented in our system.

For all methods, necessary data for entry into the computerized system is the origin of the waste, date of generation, physical state and chemical composition, radiological protection information, packaging data, sampling procedures, representativeness issues.

For all the methods, mandatory information is raised to track the wastes and setup the radioisotopic inventory report. A check page was created so that the image of the declared label can be inserted and the confirmation fields are filled in by the facility operator. One common example of this instance is the disused sealed radioactive sources that are collected for treatment and disposal. In future routines, the operator will check the dose and add the value in the due field, declaring the detector used, date, detector calibration number, and date of calibration. The operator will register in the system the technique used for the chemical separation of the target elements and measurement, allowing the construction of a library of methods—avoiding loss or poor use of previous information. The proposed system allowed the generation of labels to track the historical data of each sample. Furthermore, the system was able to store the analysis of each of the samples, declaration of transfer (if applicable), final destination and declaration of those responsible for the samples.

For simple radiometric analysis (method 3) and scanning radiometric analysis (method 6), the material is registered and the system generates a label for the sample or the package. Reports obtained in alpha, beta, and gamma analyzes should be uploaded so that they remain accessible over time for further use. In addition, the record of dose rates for both samples and packages must be maintained. These records can be updated by a mobile module, based on the scanning of the labels generated by the system, including again the instrument used, date, calibration number detector and calibration date, as previously mentioned.

As for method 4 (dose ratio versus activity for the entire package), the first step was to identify the usual geometries in the management routine. The project included: point source, cylinder source, and infinite slab (cube). Given the complexity of the non-pattern behaviors seen in the reference material of Rockwell [15], the results obtained in the Microshield® software were unsatisfactory. Then, a set of approximately 5000 data points was manually interpolated to obtain the polynomial curves used within the algorithm. Build-up factor curves were also constructed by selecting the materials common to shielding: lead, water, iron, and concrete. A library of energies and yields was built from the identification and categorization of common radioisotopes arising from mining activities, operation of reactors, operation of accelerators, production of radioisotopes, and disposal of sealed sources. A material library was also built to calculate the attenuation coefficients and establish standardized densities in a single reference. The values of attenuation coefficients for the construction of the curves were taken from a NIST Standard Reference [16]. The constructed curves include energies from 1 keV to 20 MeV.

For non-contemplated or mixed materials, an approximation module was built in the system just entering the percentage of the composition. As a return, a base material was defined to make the calculation feasible. After the calculation, the corrected activity values and the corrected dose values were generated. These values were then compared to the measured values. The values obtained will be included in the radioisotopic inventory of the waste packaging, which may also have its dose rates updated with the mobile module.

For the construction of the scaling factor module, the mathematical model presented in [17] was used. The algorithm highlights the so-called 'key radionuclides' and the 'difficult to measure radionuclides'. If it is not possible to obtain the scaling factor when the dispersion of measurement data $D2\sigma \geq 10$, according to method and criteria of the [17], the user will be guided to calculate a correlation function. If it meets the criteria for accepting the existence of a correlation function, that is the value of the correlation coefficient $(r) \geq 0.6$, the appropriate looping of the algorithm is ended [17]. If not, it triggers the storage of the information so that, when inserting new information in the system, the calculation of scaling factors and/or correlation function occurs again considering previous and current values. Graphs will be generated to check for points that are distant from the average, allowing the exclusion of outliers, but not permanently. The reason for that is that new data can change the status of the former data, from outliers to consistent data as new information is gathered. In future versions, the system will allow the exclusion of the outliers automatically but, again, not permanently.

For keeping the inventory up-to-date, a decay module was developed based on the Bateman equation. The module is capable of calculating the decay of the initial radioisotope by updating its activity as a function of time, as well as allowing the estimation of the activity of the isotopes present in the decay chain. For the application of the Bateman equation, the chains of the cataloged radionuclides were considered so that, radionuclides that have very short half-lives compared to the parent radionuclide, have their activities

approximate as being equivalent to their parent radionuclide at any time. Expansions called Bateman 0 to Bateman 4 were made, which makes it possible to estimate the activity of radionuclide chains for over 300 thousand years [18].

Since they are not mathematical models, tests were not performed for the methods (1), generator information, (2) radiochemical analysis of samples, (3) radiometric analysis, and (4) gamma-scanning radiometric analysis. The methods are described in the documentation and in the prototyping of screens consolidating the information that is mandatory for storage and tracking of the radioactive waste history. For the methods (4) dose versus activity ratio and (5) scaling factors and correlation functions, nine tests were performed, using data extracted from the literature for the composition of the scenarios [19–21].

Six cases were considered to assess the performance of the algorithm: (1) radioactive sealed source with the application of the model for point source ^{192}Ir and ^{60}Co (Point Source-A); (2) the same as (1) but using, instead of point source, the cylindrical source model (Cylindrical-A); (3) several ^{192}Ir and ^{60}Co sources grouped in one plastic beaker (50 sources exactly) assessed by the cylindrical source model for determining ^{192}Ir and ^{60}Co activities independently (Cylindrical-B); (4) slab source for ^{137}Cs/^{137m}Ba contaminated paper bundles conditioned in cubic, steel boxes; (5) cylindrical source for 200 L drums containing spent ion-exchange resin with ^{137}Cs/^{137m}Ba and ^{60}Co (Cylindrical-C); (6) cylindrical source for drums containing two types of mixed waste with ^{137}Cs/^{137m}Ba and ^{60}Co (Cylindrical-D1 and -D2); (7) scaling factors, for the determination of the activity of ^{238}Pu, ^{234}U, ^{230}Th, ^{226}Ra (^{238}Pu decay series) in waste samples reported in ref. xxx (Supplementary Information, Table S1).

Results and discussion

Characterization methods

In the present work, waste characterization consists in the determination of the concentrations of activity of the radionuclides contained in the waste [1]. This information is necessary for the work in all steps of the waste management process to comply with current radiological protection regulations and the waste to comply with the acceptance criteria of the repository where it will be disposed of definitively [22–24]. The characterization methods included in this paper are (a) radiochemical analysis of waste samples, (b) measurement of dose rates of waste packages and calculation of activity concentration by the dose-to-activity method, (c) application of scaling factors and correlation functions. Other methods that are currently being used or about to be used are: (d) validation of data presented by the waste producer, (e) modeling of the process that generates the waste, (f) gamma scanning of waste packages, and (g) tomography of waste packages.

Radiochemical analysis

One of the major problems faced in the characterization of radioactive waste is the presence of pure alpha or beta emitters, which makes it impossible to perform direct measurements of the waste packaged [25]. Thus, to obtain radioisotopic inventory of waste packages, the use of radiochemical methods for characterization is necessary. These methods are to obtain and process radioactive waste samples and to apply techniques to isolate the chemical element, to which the target radioisotope belongs, for example, precipitation, solvent extraction, chromatography, ion exchange, etc.

The data necessary for entry into the computerized system are the origin of the waste, date of generation, physical state and chemical composition, radiological protection information, packaging data, sampling procedures, representativeness issues, results of gamma-ray and alpha-particle spectrometry, measurement of beta emitters, analytical standards and validation status of the method, equipment used, responsible personnel for each step in the characterization process, and any other information relevant to the quality assurance of waste history and sample processing.

Over the past decade, most of the relevant radionuclides have been contemplated in the development of radiochemical analysis methods as a result of the waste characterization program at the RWMF/IPEN-SP. These radionuclides are present in nuclear power plant waste and other nuclear fuel cycle facilities, research reactors, industrial radiopharmacy, oil and gas production rigs, and other mining facilities. Figure 1 shows the flowchart of the process that is performed to analyze spent filter samples from a research nuclear reactor, as an example of data that will be handled by the system.

Dose-to-activity

For gamma-ray emitters, one method that can be used to determine the activity of each radionuclide in a waste packaging with acceptable accuracy consists of measuring the dose rate at a certain distance from the external surface of the packaging [27], in combination with the modeling of the measurement process: the Point-Kernel analytical method or the Monte Carlo stochastic method [28].

These two models are used to estimate the rate of photons emitted by the radionuclides in the waste, with an initial guess of unitary activity concentration, passing through a unitary surface centered in the point where the measurement is made. The calculated dose rate resulting from this flow is then compared with the measured value and the ratio

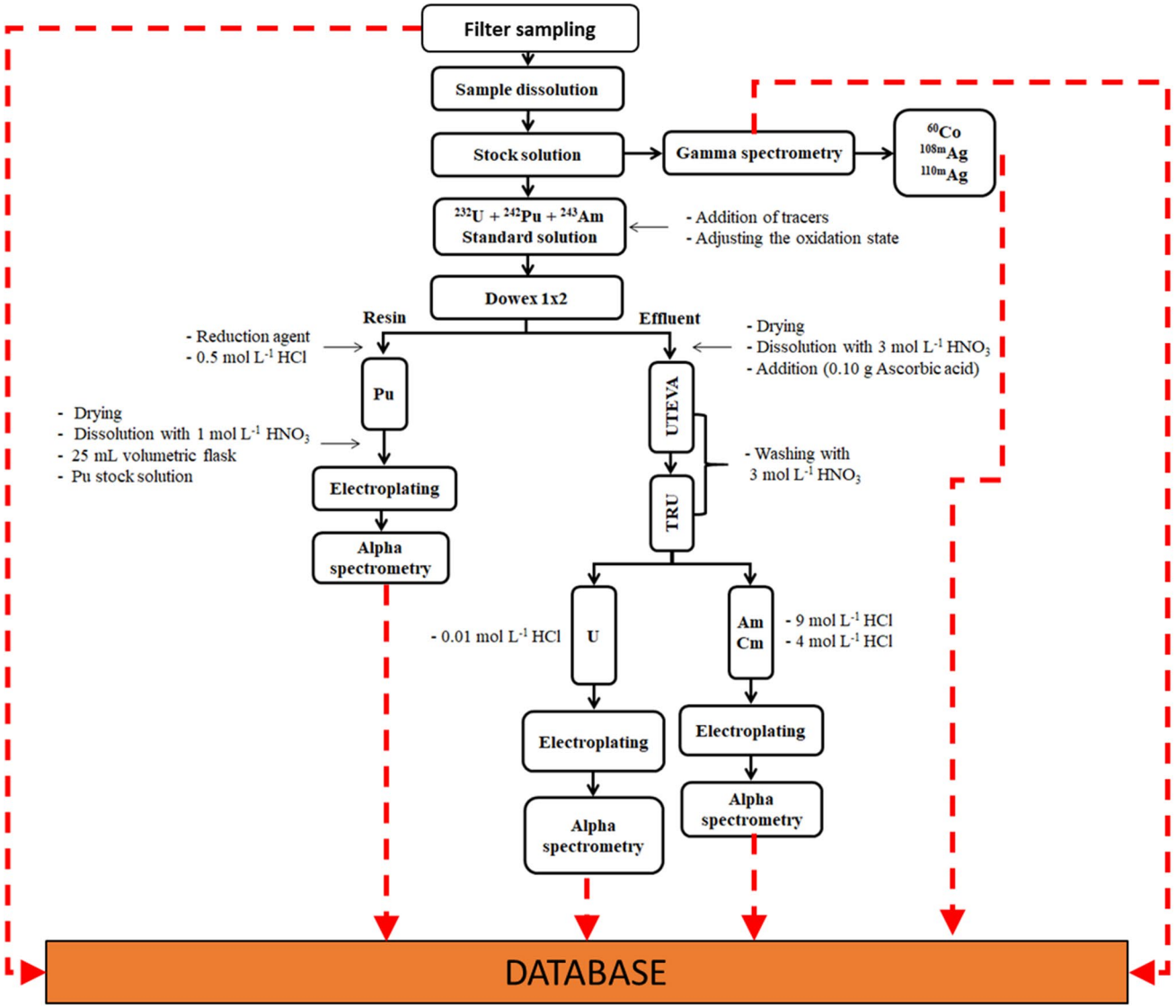


Fig. 1 Flowchart of the sequential determination of the isotopes of U, Pu, Am, and Cm in spent filters from the primary cooling water of a nuclear research reactor. Solid black lines represent the flow of materials and red long dashed lines represent the flow of information. Adapted from [26]. (Color figure online)

between them is used to correct the initial guessed value of the activity.

The 'dose-to-activity' method is widely used in the RWM routine due to the low cost, fastness, and because it results in a lower dose for the operators, especially when the waste has medium or high radiation levels [29, 30].

The Point Kernel method described by Rockwell [15] is a semiempirical method by which the photon flow at the measurement point is calculated for each geometry of the waste package, by integrating over its volume, the contribution of photons emitted in each unitary volume and attenuated by the waste material, package and shielding along the path to reach the point of measurement. The photon flow is then converted to dose rate by the expression:

$$\dot{D} = \sum_i \varphi_i \times G_i \tag{1}$$

where, $\dot{D}$ is the dose rate at the point of measurement, in $Gy{\cdot}h^{-1}$, φ_i is photon flux of each initial energy i at this point ($cm^{-2}{\cdot}s^{-1}$); and G_i is the gamma dose factor per unitary flow of photons of energy i, in $Gy{\cdot}h^{-1}{\cdot}cm^{-2}{\cdot}s$.

For each waste package geometry, there is an equation to calculate the photon flux at the point of measurement. For this work, three usual geometries were employed: point source (Eq. 2), which is used to model the measurement of disused sealed radioactive sources, infinite slab (Eq. 3), which is used to model the measurement of cubic

box-shaped waste packages, and cylinder (Eq. 4), which is used to model the measurement of waste drums.

For point source,

$$\phi_i = B\frac{A_i}{4\pi a^2}e^{-b_1} \tag{2}$$

where, B is the buildup factor of shielding, when present (dimensionless), A is the photon emission rate (s^{-1}), a is the distance between the measurement point and the source (cm), b_1 is the summation of the photon beam attenuation of all radiation shielding ($\sum_j^n \mu_{ij}t_j$), where μ_{ij} is the gamma radiation attenuation coefficient of the photon of energy i by each shielding material j (cm^{-1}) and t_j is the thickness of each shielding j (cm).

For rectangular boxes, the following expression, which models an infinite slab source, gives a good approximation of the photon flux at the point of measurement:

$$\phi_i = \frac{BS_{Vi}}{2\mu_{Si}}\left[E_2(b_1) - E_2(b_3)\right] \tag{3}$$

where, S_{Vi} is the emission rate of photons of energy i, in each unitary volume of the waste, (cm^{-3} s^{-1}), b_3 is equal to the summation of b_1, as defined earlier, and the product of the cross-session of the waste material, for photons of each energy i and the waste slab thickness h, ($b_1 + \mu_s h$), where μ_{si} is the attenuation coefficient of the waste material for the photons of energy I (cm^{-1}); E_2(b) is given by:

$$b^{n-1}\int_b^\infty \frac{e^{-t}}{t^n}dt, \quad n \geq 0 \quad E_0(b) = \frac{e^{-b}}{b} \tag{4}$$

For cylindrical drums, the following expression holds:

$$\phi_i = \frac{BS_{Vi}R_0^2}{2(a+Z_i)}F(\theta, b_2) \tag{5}$$

where B, $S_{Vi,}$ and a were defined previously, R_0 is the radius of the drum (cm); Z called an equivalent attenuation distance (cm), is calculated by interpolation in semi-empirical graphs for cylindrical geometry; $b_2 = b_1 + \mu_s Z$; and F(θ,b) is the function ($\int_o^\theta e^{-b}sec\theta' d\theta'$) for θ given for point P in the axial median plane of the cylinder.

The initial calculation of the dose rate in the measurement positions can be done with any Sv_i values, provided that the proportion between the emission rates of any pair of photons of different but reasonably close energies are in accordance with the rates detected by gamma spectrometry of the waste package or determined earlier. Reasonably close means here energies whose absorption in the path from the source to the measurement point do not differ more than a fraction of the unit.

Finally, each φ_i value is adjusted by multiplying it by the ratio D_{meas}/D_{calc} so that a new dose rate calculated according to expression (1) matches the measured value while respecting the proportions of emission rates. To improve accuracy, multiple measurements may be made at the same distance from the drum surface and the correction factor is calculated using the mean value of the ratios between the measured and the calculated dose rates.

To verify the accuracy of the method, the new calculated values of Sv_i are translated into the activity of the corresponding radionuclides, considering the yield of photon emission by decay. Activities are then used to calculate the dose rate at the measurement positions, using a computational package like the MicroShield (™) or by calculation using the Point-Kernel method.

Scaling factors and correlation functions

In the routine characterization of waste streams, the determination of difficult-to-measure radionuclides (DTM), which are the pure beta, pure alpha and very low-energy or low-yield gamma emitters, is usually made by using scaling factors (SF) or correlation functions (CF) previously determined by radiochemical methods [31]. The use of the method of SF and CF intends to avoid the costly and time-consuming radiochemical analysis of samples taken from multiple batches of a waste that are generated with reasonably constant composition over time.

In the method of SF&CF, the concentration of DTMs is evaluated by multiplying the concentration of a key nuclide (KN), a measurable gamma emitter, determined by the radiometric method, by factors that represent a constant ratio between the concentrations of the DTMs and the KN. In short, constant ratios, if they exist, are measured previously by the radiochemical method for a determined waste stream from a specific facility [2, 31]. The scaling factor can be applied in the same way as the characteristics of a large number of waste packages are inferred from a representative sample of them, both for existing waste and for those that will be generated in the future in the same facility and by the same process. Thus, the activity of a DTM is given by:

$$A_{DTM} = SF.A_{KN} \tag{6}$$

where, A_{DTM} is the activity of a difficult to measure radionuclide; SF is the scaling factor, and A_{KN} is the activity of the key radionuclide.

To determine the scaling factor, the geometric mean of the ratios P_i between the activities of the DTM and the KN is obtained by measurements of N waste samples. The geometric mean is used considering that these ratios vary orders of magnitude for different samples of the same waste stream. Therefore, the mean scaling factor is

obtained as the Nth root of the product of all P_i. A more friendly way to calculate the mean is to take the natural logarithm of the ratios measured in the samples, sum them all, divide the result by the total number N of measurements, and finally, exponentiate the base e of the natural logarithms to the result of the previous calculation. Thus, $\overline{SF}_i$ is given by:

$$\overline{SF}_i = \sqrt[N]{\prod_i P_i} \text{ or the equivalent expression } \overline{SF}_i = e^{\frac{\sum_{i=1}^{n} \ln P_i}{N}} \tag{7}$$

A calculated mean scaling factor can be considered applicable in the determination of DTM's activities if the standard deviation, $D_{2\sigma}$, of the N individual results of the proportions, is less than or equal to 10, that is, $D_{2\sigma} \leq 10$, meaning that the existence of a useful SF for the dataset is defined by a criterion that requires that 95.5% of the measured P_i are within the range:

$$\frac{\overline{SF}}{10} \leq (SF)_i \leq \overline{SF} \times 10 \tag{8}$$

The standard deviation $D_{2\sigma}$ is given by:

$$D_{2\sigma} = e^{2\left(\frac{\sqrt{\sum_{i=1}^{n}[ln(\overline{SF})_i - ln\left(\overline{SF}\right)]^2}}{N-1}\right)} \tag{9}$$

When no valid scaling factor is obtained, because the proportions vary more widely than the acceptable range, a second attempt is made to get a relationship between the activity of a DTM and a KN, which considers that the SF is not constant, but rather a function of the KN concentrations. In these cases, instead of an SF, we search for a correlation function (CF), which can be expressed as follows:

$$A_{DTM} = a(A_{KN})^b \tag{10}$$

where a and b are constants determined empirically by the results of the N measurements. More conveniently, this relationship can be written as follows:

$$ln\left(A_{DTM}\right) = ln(a) + b \times ln\left(A_{KN}\right) \tag{11}$$

Thus, a straight line can be adjusted to the values of the logarithms of the activities, whose parameters a and b are obtained with a linear regression of logarithms by the least square method, according to the matrix equation Eq. (12).

$$\begin{bmatrix} \ln(a) \\ b \end{bmatrix} = \frac{1}{\Delta}\begin{bmatrix} \sum_{i=1}^{N}\left(\frac{x_i^2}{\sigma_i^2}\right) & -\sum_{i=1}^{N}\left(\frac{x_i}{\sigma_i^2}\right) \\ -\sum_{i=1}^{N}\left(\frac{x_i}{\sigma_i^2}\right) & \sum_{i=1}^{N}\left(\frac{1}{\sigma_i^2}\right) \end{bmatrix} \times \begin{bmatrix} \sum_{i=1}^{N}\left(\frac{y_i}{\sigma_i^2}\right) \\ \sum_{i=1}^{N}\left(\frac{y_i x_i}{\sigma_i^2}\right) \end{bmatrix} \tag{12}$$

where Δ is given by:

$$\Delta = \left[\sum_{i=1}^{N}\left(\frac{1}{\sigma_i^2}\right)\right] \times \left[\sum_{i=1}^{N}\left(\frac{x_i^2}{\sigma_i^2}\right)\right] - \left[\sum_{i=1}^{N}\left(\frac{x_i}{\sigma_i^2}\right)\right]^2 \tag{13}$$

and

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

The measure of the degree of correlation between the activities of the DTMs and KNs are provided by the Correlation Coefficient r, given by (Eq. 14). The acceptable correlation function is here defined by the criterion $r \geq 0.6$.

$$r = \frac{\sum_{i=1}^{N} X_i y_i - \frac{\left(\sum_{i=1}^{N} X_i\right)\cdot\left(\sum_{i=1}^{N} y_i\right)}{N}}{\sqrt{\left[\left(\sum_{i=1}^{N} X_i^2 - \frac{\left(\sum_{i=1}^{N} X_i\right)^2}{N}\right)\left(\sum_{i=1}^{N} y_i^2 - \frac{\left(\sum_{i=1}^{N} y_i\right)^2}{N}\right)\right]}} \tag{14}$$

Different from the first two characterization methods, the SF&CF method can only be applied after enough data from the radiochemistry method has accumulated over time and the applicability criteria have been accepted. The number N of samples that must be analyzed before the SF&CF can be applied depends on the composition stability of a sequence of batches of a particular waste stream over time. According to Taddei et al. [21], while the required number of batches analyzed may be as high as 30, SF was obtained for various radionuclides present in spent ion-exchange resin and activated carbon from the primary cooling circuit of a research reactor with reduced number of batches. Therefore, the validity of the scaling factors obtained in the referenced material must be checked in the future as more waste is analyzed and analysis results are available.

Catalog of relevant radionuclides

One important step in any waste characterization program is to identify the set of radionuclides that are, or may be, present in the waste and that may constitute important contributors to the radiation doses incurred by operators, members of the public and, most important, potentially exposed people in the long term while the waste is disposed of in a repository with only passive safety barriers acting as protection against radiation risk. Such a set forms a catalog of radionuclides of the characterization program.

Radionuclides with half-lives below 100 days were excluded from the catalog except some that are daughter isotopes in a decay chain and that are important dose builders. Therefore, their contribution to activity and total radiotoxicity is considered in both safety assessments of the pre-disposal activities and of the long-term waste disposal. Tables 1, 2, and 3 bring the radionuclides significant to the RWM according to the practices that generate them.

Table 1 Radionuclides contained in waste from mining activities and accelerators

Mining waste			Accelerators waste		
Nuclide	T/2	Unit	Nuclide	T/2	Unit
^{210}Po	138	D	^{22}Na	2.6	Y
^{210}Pb	22.3	Y	^{26}Al	7.2×10^{5}	Y
^{226}Ra	1,600	Y	^{53}Mn	3.7×10^{6}	Y
^{228}Ra	5.75	Y	^{54}Mn	312.2	D
^{227}Ac	21.8	Y	^{56}Co	77.7	D
^{228}Th	1.9	Y	^{57}Co	272	D
^{230}Th	75,400	Y	^{60}Co	5.3	Y
^{231} Pa	32,760	Y	^{64}Zn	244	D
^{234}U	2.5×10^{5}	Y	^{134}Cs	2.1	Y
^{238}U	4.5×10^{9}	Y	^{152}I	13.3	Y
			^{154}I	8.8	Y
			^{207}Bi	31.55	Y

For running the code, radionuclide-related data such as dose factors, emission types, energies, and emitted radiation yields are included in a database and used in the system.

Polynomial adjustment

The step of data collection in this work consisted largely of the interpolation or extrapolation of data from printed tables and graphs. Thus, with the help of computational tools, curves were determined that describe with great similarity the relationship between reading data and its consequences. The reliability of these curves was determined by the coefficient of determination (R^2) with values higher than 0.88, in addition to the visual analysis of each important interval. When the generated curve showed anomalous behavior at different intervals, functions were drawn for small intervals where they showed to describe the behavior of the general curve. Thus, the uncompiled system contains, instead of tables, exponential and polynomial curves that describe the collected data with great similarity. The polynomials generated from both the materials and the point-kernel method were grouped into libraries related to each function and implemented in the computation source.

Bateman's equation

To maintain the quality of the information contained in the radioisotopic inventory over time, Bateman's equation is integrated into the system. According to [32], Bateman's equation can be derived by Laplace transforms, reaching the formula described in Eq. (14)

Table 2 Radionuclides contained in waste from the production of radioisotopes and use of sealed sources

Radioisotopes production			Sealed sources		
Nuclide	T/2	Unit	Nuclide	T/2	Unit
^{3}H	12.3	Y	^{3}H	12.3	Y
^{11}Be	1.51×10^{6}	Y	^{14}C	5,730	Y
^{14}C	5730	Y	^{22}Na	2.6	Y
^{55}Fe	2.7	Y	^{36}Cl	3.0×10^{5}	Y
^{59}Ni	7.6×10^{4}	Y	^{40} K	1.3×10^{9}	Y
^{63}Ni	100.1	Y	^{55}Fe	2.7	Y
^{60}Co	5.3	Y	^{60}Co	5.3	Y
^{79}Se	1.13×10^{6}	Y	^{63}Ni	100.1	Y
^{90}Sr	28.8	Y	^{75}Se	119.8	D
^{91}Zr	1.5×10^{6}	Y	^{85}Kr	10.8	Y
^{94}Nb	2.0×10^{4}	Y	^{90}Sr	28.8	Y
^{99}Tc	2.1×10^{5}	Y	^{133}Ba	10.5	Y
^{107}Pd	6.5×10^{6}	Y	^{137}Cs	30.07	Y
^{108m}Ag	418	Y	^{147}Pm	2.6	Y
^{110m}Ag	249.79	D	^{151}Sm	90	Y
^{121m}Sn	55	Y	^{152}Eu	13.3	Y
^{126}Sn	1.0×10^{5}	Y	^{192}Ir	74.0	D
^{125}Sb	2.8	Y	^{192m2}Ir	241.0	Y
^{129}I	1.6×10^{7}	Y	^{204}Tl	3.8	Y
^{134}Cs	2.0648	Y	^{226}Ra	1600	Y
^{135}Cs	2.3×10^{6}	Y	^{228}Th	1.9	Y
^{137}Cs	30.07	Y	^{232}Th	1.4×10^{10}	Y
^{151}Sm	90	Y	^{233}U	1.6×10^{5}	Y
^{154}I	8.6	Y	^{234}U	2.5×10^{5}	Y
^{155}I	4.8	Y	^{235}U	7.0×10^{8}	Y
^{235}U	7.0×10^{8}	Y	^{238}U	4.5×10^{9}	Y
^{236}U	2.3×10^{7}	Y	^{237}Np	2.1×10^{6}	Y
^{238}U	4.5×10^{9}	Y	^{238}Pu	87.7	Y
^{237}Np	2.1×10^{6}	Y	^{239}Pu	2.41×10^{4}	Y
^{233}U	1.6×10^{5}	Y	^{241}Am	432.2	Y
^{234}U	2.5×10^{5}	Y	^{244}Cm	18.1	Y
^{238}Pu	87.7	Y	^{244}Pu	8.1×10^{7}	Y
^{230}Th	7.5×10^{4}	Y	^{248}Cm	3.4×10^{5}	Y
^{239}Pu	2.4×10^{4}	Y			
^{240}Pu	6,563	Y			
^{241}Pu	14.4	Y			
^{241}Am	432.2	Y			
^{242}Pu	3.7×10^{5}	Y			
^{243}Am	7,370	Y			
^{243}Cm	29.1	Y			
^{244}Cm	18.1	Y			

*Most radionuclides contained in radioisotope production waste result from the production of ^{99}Mo by fission. Radionuclides contained in sealed radioactive sources are the nominal radionuclides, their decay products or unavoidable contaminants

Table 3 Considered radionuclides in waste from nuclear reactors

Nuclide	T/2	Unit	Nuclide	T/2	Unit
^{3}H	12.3	Y	^{137}Cs	30.0	Y
^{14}C	5730	Y	^{151}Sm	90	Y
^{55}Fe	2.7	Y	^{152}Eu	13.3	Y
^{59}Ni	7.6E+4	Y	^{154}Eu	8.8	Y
^{63}Ni	100.1	Y	^{155}Eu	4.8	Y
^{60}Co	5.3	Y	^{230}Th	7.5E+04	Y
^{79}Se	1.1E+06	Y	^{233}U	1.6E+05	Y
^{90}Sr	28.8	Y	^{234}U	2.5E+05	Y
^{93}Zr	1.5E+6	Y	^{235}U	7.0E+08	Y
^{94}Nb	2.0E+04	Y	^{236}U	2.3E+07	Y
^{93}Mo	3500	Y	^{238}U	4.5E+09	Y
^{99}Tc	2.1E+05	Y	^{237}Np	2.1E+06	Y
^{107}Pd	6.5E+06	Y	^{238}Pu	87.7	Y
^{108m}Ag	418	Y	^{239}Pu	24,110	Y
^{110m}Ag	249.79	D	^{240}Pu	6563	Y
^{121m}Sn	55	Y	^{241}Pu	14.4	Y
^{126}Sn	1.0E+05	Y	^{241}Am	432.2	Y
^{125}Sb	2.8	Y	^{242}Pu	3.7E+05	Y
^{129}I	1.6E+07	Y	^{243}Am	7370	Y
^{134}Cs	2.1	Y	^{243}Cm	29.1	Y
^{135}Cs	2.3E+06	Y	^{244}Cm	18.1	Y

$$A_n(t) = \sum_{i=1}^{n} \left[A_i(0) \times \left(\prod_{j=1}^{n-1} \lambda_j \right) \times \left(\sum_{j=1}^{n} \left(\frac{e^{-\lambda_j t}}{\prod_{p}^{n} = i, p \neq j(\lambda_p - \lambda_j} \right) \right) \right] \quad (15)$$

where, $A_n\,(t)$ is the time-dependent activity for radioisotope n of the decay chain (Bq); t is time (years); λ_j is the decay constant of any daughter isotope j in the series, and λ_p is the decay constant of its parent isotope p.

For the application of the Bateman equation, some chains and sub-chains of the cataloged radionuclides were simplified so that radionuclides that have very short half-lives compared to the parent isotopes, have their activities approximate as being equivalent to their parent activities for all times. This reduces the complexity of the equations for long decay chains without loss of accuracy for any practical purpose. For instance, the activity of ^{210}Pb (22.3 y) can be calculated with great accuracy for all times after 1.5 months, from the initial activity of ^{226}Ra, ignoring all intermediate isotopes in the chain: ^{222}Rn (3.825 days), ^{218}Po (3.05 min), ^{214}Pb (26.8 min), ^{214}Bi (19.9 min), ^{214}Po (164 µs). This is an example where the activity of ^{210}Pb can be calculated as if it were direct daughter nuclide of ^{226}Ra, and the time interval of 1.5 months is that required for equilibrium be established between ^{226}Ra and ^{222}Rn.

Proposed system specification

The system is conceived as a managerial tool that allows automation of all administrative activities and some technical activities of the waste characterization process, for instance, the recording of data, control of access to data, QAS-related records, generation and distribution of reports, updating of waste inventory, statistical analyses, etc. Therefore, the following are some features of the proposed code.

Product goals

The product aims to meet the needs of automation of complex methods used to characterize radioactive waste and to control the radioisotope inventory of radioactive waste from each generating facility.

Specific product objectives

The system integrates the characterization methods identified in the operational routine of radioactive waste facilities, supporting the control of the waste inventory. For that, one must store data regarding the waste, as shown in Fig. 2. These data include (a) radiological, chemical and physical characteristics; (b) material classification; (c) data regarding the updated status of the waste (untreated, discharged, treated, stored, transferred to final disposal); (d) grouping of information related to the methods used to characterize (mainly in the case of radiochemical and radiometric methods). For the expansion of the modules presented in Fig. 2, please refer to the Supplementary Information, Figure S1.

The system was designed with web and mobile interfaces. The reason is due to the need to perform routine monitoring in the field. To avoid losing any piece of information, the operator and the facility manager can access the system via mobile to update the data in real time. This type of access was included in the project so that portable detectors with previously registered labels are nonessential to update the information in the field. Currently, detectors are purchased with previously registered tags, increasing the cost of these operations. In the case of the system, we will have a label specific to each installation, allowing the update to be made using any detector.

The mobile system, after login, should open a tool to scan the label of the package in the field to enter the waste IDs. After identification, the operator enters measurement data such as the measured dose rate, distances taken for measurement, the model of the detector used, and the calibration data of the detector. The system will save the information in the waste history and update the report. To access other information and tools, the user must make use of the web interface.

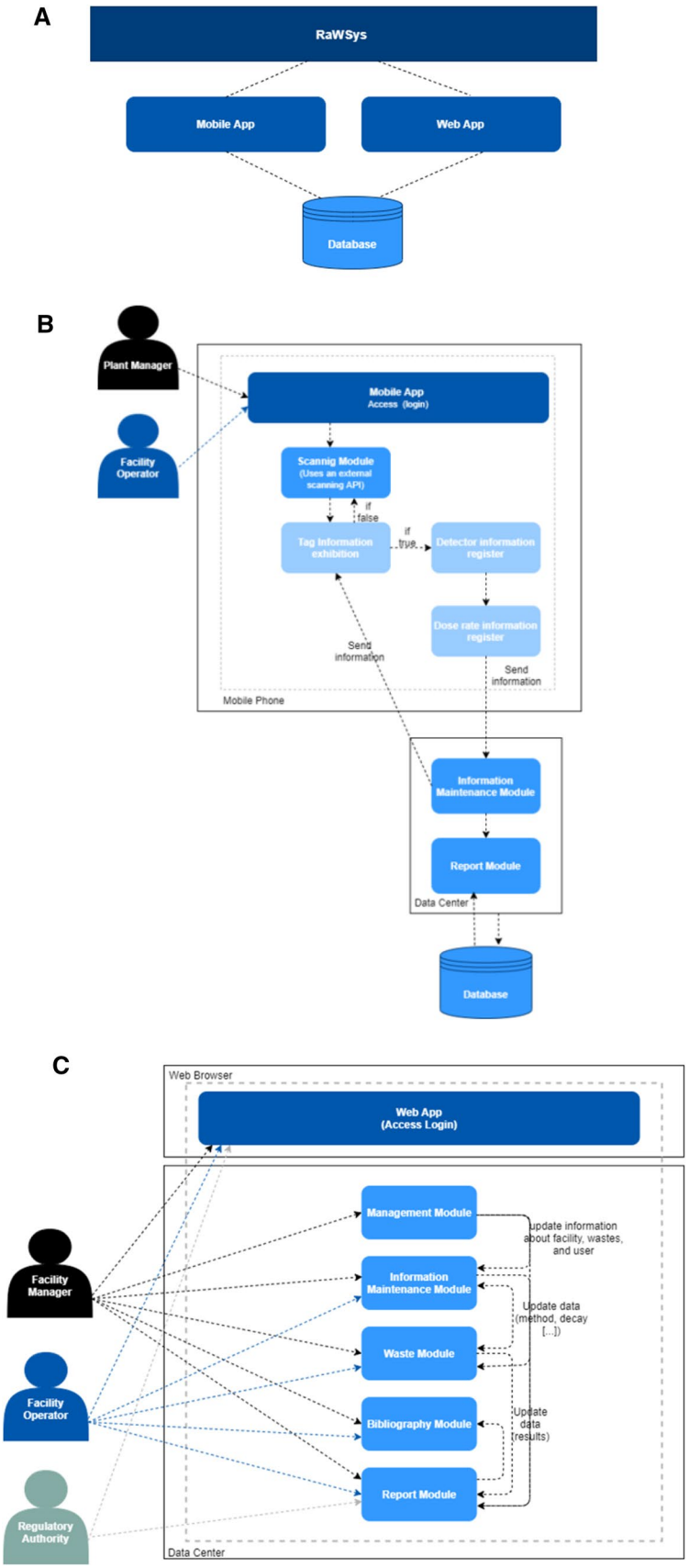


Fig. 2 Collection of data for each waste package: **a** general view; **b** context diagram of the mobile system; **c** context diagram of the web system

State of the art/current situation/problem description

No integrated system has been identified for the characterization of radioactive waste with the automation of administrative and technical activities. After evaluation, it was recognized that several methods can be applied in the characterization stage, but there is no grouping of information or quality assurance about the information generated in the literature.

In addition, the need to formalize reports related to the radioisotopic inventory was identified, with due responsibility and traceability of the information provided. These reports must be available in the system so that they can be accessed by managers and regulatory inspection agencies whenever necessary. There is a lack of security related to the access to information currently generated, and it is necessary to define access levels to guarantee the security of information in the long term. As a result, a flow is proposed, which considers the actions of the manager, the regulatory authority, and the inclusion of such information in the front-end interface. All the steps from the actions to the cloud database are illustrated in Fig. 3.

Product scope and boundary

The system so far aimed at covering the characterization stage, be it primary or final, and to meet the needs identified in the radioactive waste management services related to treatment, storage or final disposal. This system will be integrated into other modules for automation of the complete management of radioactive waste (Refer to the Supplementary Information, Fig. S1–S5).

Return/functions

- Registration for accessing the system with limited functions according to user level of access authentication (access and security levels);
- Registration of the waste;
- Record of historical actions taken to characterize each waste individually or in batches;
- Data storage related to radiochemical and radiometric methods;
- Application of pre-established methods: dose-to-activity, scaling factor and correlation function;
- Updating of the inventory using the Bateman equation;
- Generation of a standardized label with essential information and bar code system;
- Generation of partial or final reports, including radioisotopic inventory;
- Dose rate update using barcode and mobile system.

Application of the algorithm in real cases

Table S2 lists the base information used for generating all the data that are presented in Tables S3–S11. The data generated by the application of the algorithm reproduced the data of the literature used [19, 21, 33].

Conclusions

The main objective of the work was to generate the specifications/documentation so that it is possible to implement a characterization system that can be used by areas that routinely manage radioactive waste. The data obtained have enabled the construction of the guide code in pseudotechnical language, which is necessary for the programming and implementation of the system, besides the functional specifications document, with a detailed description of functions, fields for completion, number of characters, and grouping of developed libraries, and the interface prototype (wireframes) that complement the system documentation.

The system is currently under development for implementation and testing. The application of the complete system will allow storing the quality of the information generated, data traceability and those responsible for the actions taken. After implementation, the costs of operations will be reduced, since the system will avoid rework or loss of information, maintaining the archiving of radioisotopic inventories in the long term. In addition, it will allow the mapping of methods that have already been used, facilitating the management of the wastes already analyzed.

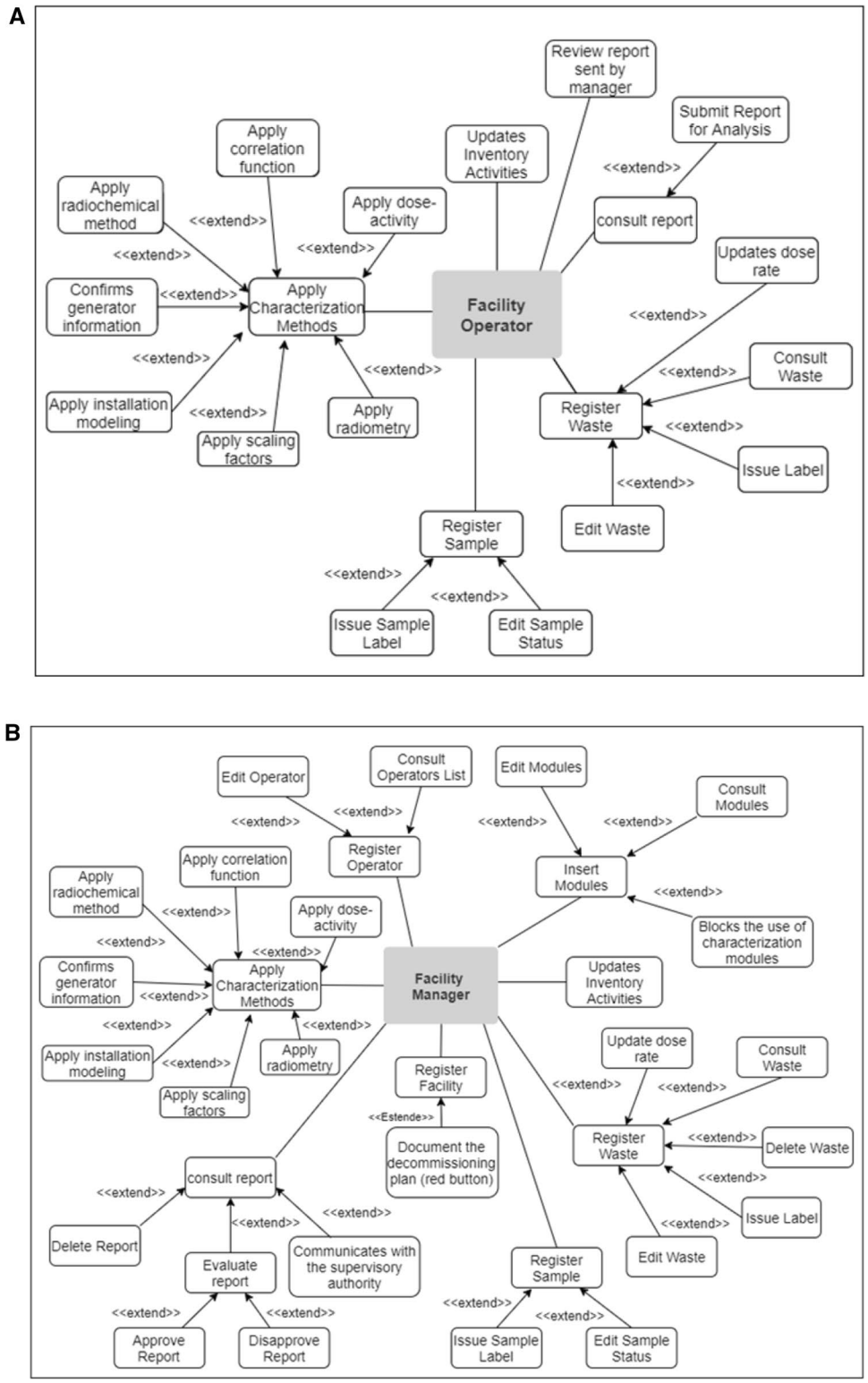
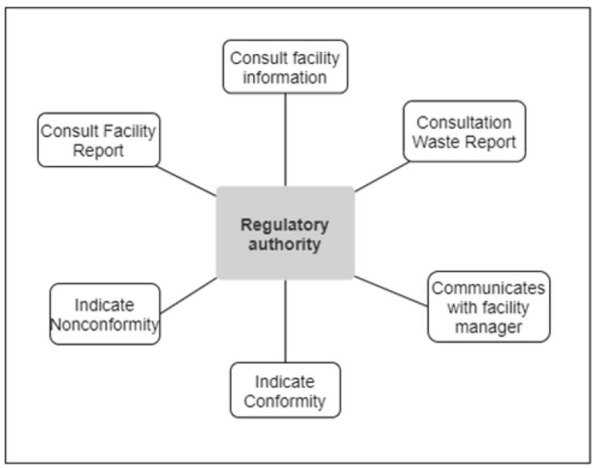


Fig. 3 Collection of data for each waste for mobile and web applications in the perspective of the **a** facility operator; **b** facility manager; **c** regulatory authority

Fig. 3 (continued)



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