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Radiological characterisation for the clearance of burnable waste produced at CERN

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ABSTRACT

Burnable waste produced at CERN during upgrading, maintenance and dismantling campaigns may be contaminated with radioactive nuclides produced through activation of accelerator components. Here, we present a methodology for the radiological characterisation of burnable waste, which takes into account the wide range of potential activation conditions (beam energy, material composition, location, irradiation and waiting time). Waste packages are measured using a total gamma counter, with the sum of clearance limit fractions estimated using the fingerprint method. Gamma spectroscopy was found to be unsuitable for classifying this waste due to the long counting times required to identify many expected nuclides, but was retained for quality control purposes. Using this methodology, a pilot campaign was performed in which we were able to clear 13 m^3 of burnable waste as conventional non-radioactive waste.

1. Introduction

The radiological activation of particle accelerator components is an important radiation protection concern (International Atomic Energy Commission, 2020). Where accelerator materials, including cables, supports, superconducting magnets, circuitry and concrete tunnel walls, are exposed to primary or scattered particles, stable nuclides may become radioactive (Vincke 2011), requiring the eventual disposal of these materials as radioactive waste. CERN operates a family of accelerators, ranging in energy from 160 MeV (Linac4) to 7 TeV (Large Hadron Collider, LHC). The components of these accelerators are exposed to a range of particles, including protons, neutrons and charged pions, meaning activation, and the requirement for full radiological characterisation prior to disposal, is inevitable.

Burnable waste items, including overalls, gloves, masks, overshoes, wipes and ventilation filters, represent a unique category of potentially radioactive waste. Typically, these materials are not activated themselves, rather are contaminated with dust and small fragments from directly activated accelerator components during upgrading, maintenance and dismantling campaigns. Burnable waste is (for obvious reasons) a fire hazard, thus it is desirable to minimise the amount of burnable waste stored at CERN at any given time. The radiological characterisation of this waste is challenging because the precise contaminating materials and irradiation conditions may be unknown. In addition, the typically low mass density of burnable waste items increases the gamma spectroscopy counting time required to detect the expected radionuclides.

In principle, slightly or potentially radioactive burnable waste produced at CERN can either be disposed of as very-low-level (Très Faible Activité, TFA) radioactive waste in France if it meets the associated acceptance criteria (Agence Nationale pour la Gestion des Déchets Radioactifs, 2013), or be cleared from regulatory control in Switzerland if it meets the Swiss clearance requirements (Swiss Federal Council, 2018). Clearance offers considerable cost savings and avoids taking up unnecessary space at a radioactive waste repository. However, it requires an especially robust characterisation methodology due to the relatively strict clearance limits applied to specific activities, and because characterization mistakes can lead to the clearance of material that is actually radioactive.

The burnable waste clearance pathway at CERN is named B-FREE (Burnable waste for Free Release). The primary characterisation approach used in B-FREE is measurement with a total gamma counter, with gamma spectroscopy used as an additional quality control. If the activity levels of a given waste package exceed the Swiss clearance limit,

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the package is compacted into metallic drums and disposed of as TFA waste in France. While some of the details described here are specific to CERN, the overall approach could be applied to other similar situations involving clearance of burnable waste potentially contaminated with activated metals and concrete originating in particle accelerators.

2. Waste candidate criteria

The methodology described below applies to burnable waste produced at hadron machines with accelerating potentials of 160 MeV and above. For a waste package to be considered suitable for clearance, the following criteria need to be met.

- No metallic fragments larger than 3 cm (or >5% of waste bag volume) detected by radiography. Smaller metallic items are accepted by the incineration facility if they are part of a larger burnable item, e.g. zips in overalls.
- Waste must not originate from facilities or experiments where contamination with alpha emitting nuclides is possible (e.g. n_TOF, ISOLDE).
- No liquids or liquid containers.
- Dose rate at contact must be below 30 nSv h^{-1}.
- Surface contamination must be below 0.4 Bq cm^{-2} Co-60 equivalent on the external surface of the waste bag.

In addition, waste must meet the criteria for clearance in Switzerland, as defined by the Swiss Federal Council (2018). As part of these criteria, each nuclide has a “Limite de Libération", or clearance limit (CL), in Bq/g. The sum of CL fractions (CL_{sum}) for all nuclides (Equation (1), below) must be below unity.

$$CL_{sum} = \sum \frac{A_i}{CL_i} \leq 1 \qquad \text{Equation 1}$$

If this limit is exceeded for a given waste package, it is considered to be radioactive and disposed of as radioactive waste.

3. Radionuclide inventory

The first stage of the characterization strategy was to define the inventory of nuclides expected to be present. Burnable waste may be contaminated with a range of materials, each subject to various irradiation conditions (energy, location, irradiation time and waiting time). As these parameters are typically unknown, a wide range of activation scenarios must be considered and the most conservative scenarios identified. Here, we define ‘conservative’ scenarios as those yielding a large CL_{sum} per detected gamma emission, therefore leading to a conservative radiological classification.

Activation products, with their associated activity per unit mass of the material (Bq/g), were estimated using the software tool ActiWiz (Vincke and Theis 2014, 2018). This application uses a proprietary nuclear data library based on extensive simulations performed using the Monte Carlo radiation transport code FLUKA (Fassò et al., 2004) as well as data originating from evaluated nuclear libraries such as JEFF, both arranged in group-structures for reasons of efficiency. Validation studies have shown good agreement between the results of ActiWiz and direct Monte Carlo scoring, as well as ActiWiz and experimental data (Duchemin 2019). The code was used to estimate the radionuclides produced for five different primary particle energies/momenta, seven locations with respect to the beam line, four irradiation times and five waiting times (also known as cooling time, i.e. the time since last irradiation) (Table 1). These scenarios were chosen to ensure that the exposure situations cover a large majority of cases typically encountered at CERN’s high energy accelerators. The various combinations of these parameters gave a total of 700 scenarios.

During analysis, the scenarios were divided into three waiting time periods: 6 months to 3 years (420 scenarios covering short to long maintenance periods), 3–10 years (280 scenarios covering waste with medium-term storage periods) and 10–30 years (280 scenarios covering waste with long-term storage periods). Note that the 3 year and 10 year waiting times appear in more than one waiting time period.

Table 1
Irradiation parameters. The different combinations of location, energy, irradiation time and waiting time give a total of 700 scenarios.

Location	[1] Beam impact area, [2] 10 cm lateral to target, [3] Within bulky materials, [4] Adjacent to bulky materials, [5] Behind massive shielding, [6] Close to wall with beam on target, [7] Close to wall with beam losses.
Energy	[1] 160 MeV, [2] 1.4 GeV, [3] 14 GeV/c, [4] 400 GeV/c, [5] 7 TeV
Irradiation times	[1] One, [2] Three, [3] Ten, [4] Thirty years
Waiting times	[1] 6 months, [2] One, [3] Three, [4] Ten, [5] Thirty years

Calculations were performed for four contaminating material types considered typical of materials used at CERN, namely aluminium (6060), stainless steel (304L), copper (CuOFE) and concrete. The elemental composition of each material is shown in Tables 2–5. Lead was not expected to be present in burnable waste, however a separate radionuclide inventory was produced for lead (HPPb4N) (Table 6) to enable us to be alert to the presence of characteristic activation products during quality controls.

For each radionuclide listed in the ActiWiz output file for a given irradiation scenario, the activity was divided by the nuclide-specific clearance limit (CL). The sum of CL fractions for all nuclides was then calculated, and the nuclides required to reach 90% of CL_{sum} identified. ‘Expected’ nuclides were those required to reach 90% of CL_{sum} for ≥80% of scenarios for a given waiting time period, while ‘potentially present’ radionuclides for a given waiting time period were defined as those required to reach 90% of CL_{sum} for at least one scenario, but less than 80%. Nuclides not required to reach 90% of CL_{sum} for any irradiation scenario were considered relatively unimportant from a clearance perspective. An example of this process is provided in Table 7. In this case, 90% of CL_{sum} can be reached by Zn-65 and Na-22 alone, thus only these nuclides are considered relevant for this scenario. The radionuclide inventory, including expected and potentially present nuclides, is shown in Table 8.

4. Total gamma counting methodology

A total gamma counter (TGC) (RTM644lnc, Mirion Technologies) was used to measure the total activity of each waste package. The recorded activity was then scaled using a ‘fingerprint’, defined as a list of radionuclides with associated activities expressed as a percentage of detected activity. Fingerprints for each material and waiting time period were defined by first calculating a figure of merit (FOM) for each of the 700 scenarios described above, in Equation (2), below.

$$FOM_j = \frac{\sum A_{i,j}/CL_i}{\sum A_{i,j} * LNC_i} \qquad \text{Equation 2}$$

Where FOM_j is the figure of merit for scenario j, $A_{i,j}$ represents the activity of nuclide i for scenario j, and CL_i represents the clearance limit of nuclide i. The LNC_i (leading nuclide correlation) factor indicates the detectability of gamma rays for nuclide i, relative to Co-60. As such, the FOM can be understood as a measure relating the radiotoxicity of a material versus its measurability via total gamma counting. Figs. S1–S12 in the supplementary materials of this paper show the distribution of figures of merit and the impact of waiting time and beam energy.

The fingerprint for a given material and waiting time period was defined as the list of nuclides required to reach 90% of CL_{sum} for the scenario with the highest FOM, with their associated activity proportions. In the example shown in Table 7 (which happens to be the most conservative scenario for the 6m-3y waiting time period, for any material), Zn-65 and Na-22 are required to reach 90% of CL_{sum} and thus

Table 2
Weight fractions for Aluminium (6060), density = 2.7 g cm^{-3}.

Element	Al	Cu	Cr	Fe	Mg	Mn	Si	Ti	Zn
WF (%)	98.375	0.1	0.05	0.2	0.475	0.1	0.45	0.1	0.15

WF = weight fraction.

Table 3
Copper (CuOFE), density = 8.9 g cm^{-3}.

Element	Bi	Cd	Cu	Pb	Hg	O	Se	S	Te	Zn
WF (%)	0.001	1E-4	99.99	0.001	1E-4	5E-4	0.001	0.0018	0.001	1E-4

Table 4
Stainless steel (304L), density = 8.0 g cm^{-3}.

Element	C	Cr	Co	Fe	Mn	Ni	P	Si	S
WF (%)	0.03	18.5	0.1	67.08	2	11.25	0.0225	1	0.015

Table 5
Concrete, density = 2.4 g cm^{-3}.

Element	Al	Ba	Ca	C	Eu	Fe	H	K	Mg	O	Si	Na	S	Ti
WF (%)	2.11	0.1	23.93	4.38	1E-4	1.26	0.56	0.83	1.51	48.22	16.18	0.45	0.39	0.17

Table 6
Lead (HPPb4N), density = 13.4 g cm^{-3}.

Element	Sb	As	Bi	Cd	Cu	Fe	Pb	Ag	Sn	Zn
WF (%)	0.0051	9E-4	4E-4	1E-4	1E-4	1E-5	99.99	3E-4	1E-4	2E-4

Table 7
Nuclides produced through activation of aluminium (located close to tunnel wall with beam losses, 160 MeV beam, 1 year irradiation time at 1 particle per second, 6 months waiting time). Only the top 10 contributors to CL_{sum} are shown.

Nuclide	Activity (Bq/g)	CL (Bq/g)	Fraction of CL	Contribution to CL_{sum}	Needed to reach 90% of CL_{sum}?
Zn-65	1.73E-12	0.1	1.73E-11	85.67%	Yes
Na-22	2.20E-13	0.1	2.20E-12	10.87%	Yes
Mn - 54	6.49E-14	0.1	6.49E-13	3.21%	No
Sc-46	3.59E-15	0.1	3.59E-14	0.18%	No
Co-60	7.64E-16	0.1	7.64E-15	0.04%	No
Fe-59	3.24E-15	1	3.24E-15	0.02%	No
Co-56	1.77E-16	0.1	1.77E-15	0.01%	No
Co-58	5.78E-16	1	5.78E-16	0.003%	No
H-3	5.50E-14	100	5.50E-16	0.003%	No
Co-57	4.61E-16	1	4.61E-16	0.002%	No

Table 8
Radionuclide inventory for burnable waste at CERN. All these nuclides contribute to 90% of the sum of clearance limit fractions for at least one activation scenario (out of 700).

Material	Potentially present radionuclides
Aluminium	H-3*, C-14*, Na-22§, Al-26, Ti-44, Co-60, Zn-65
Copper	Ti-44, Sc-46, Mn-54§, Co-56, Co-57, Co-58, Co-60§, Zn-65
Concrete	H-3*, C-14*, Na-22§, Ti-44, Mn-54, Cs-137*, Ba-137m, Ba-133, Eu-152, Eu-154
Steel	Na-22, Ti-44, Sc-46, Mn-54§, Co-56, Co-57, Co-60§,
Lead	H-3*, Sc-46, Mn-54, Co-60, Zn-65, Rb-83, Y-88, Nb-95, Rh-102, Ru-106*, Cd-109*, Sn-113, Sb-124, Ba-133, Ce-139, Pm-143, Eu-146, Gd-146, Gd-148*, Lu-172, Hf-172, Lu-173, Hf-175, Ta-179*, W-181, Os-185, Pt-193*, Hg-194§, Au-195*, Pb-202*, Tl-204*§, Bi-207§

§Expected nuclide, *Difficult to measure nuclide.

form the fingerprint, with their activities normalised to give 100% (Table 9). The FOM for this scenario is 27.9, while the average for all scenarios is 11.9. This means the fingerprint shown in Table 9 overestimates CL_{sum} by an average factor of 2.3. For the 3y-10y waiting time period, the most conservative scenario corresponded to activated stainless steel (Table 10), while for the 10y-30y waiting time period, the most conservative fingerprint corresponds to activated concrete (Table 11).

In cases where the waiting time of a waste package is not known, the most conservative fingerprint for all waiting time periods (i.e. those shown in Table 9) was used.

5. Gamma spectroscopy methodology

In toto Gamma spectroscopy was performed at a dedicated radioanalytical laboratory at the Radioactive Waste Treatment Centre (RWTC) at CERN, using two Falcon 5000 high purity germanium detectors (Mirion technologies). The detector arrangement is shown in Fig. 1. A source-to-detector distance of 50 cm was used. Each detector undergoes regular on site quality assurance and calibration.

ISOCS (In Situ Counting Object Software) [Venkataraman 2003,

Table 9
Most conservative fingerprint for the 6m-3y waiting time period (FOM = 27.9). Figures represent activated aluminium (160 MeV, located close to wall with beam losses on bulky materials, 1 year irradiation time and 6 months waiting time).

Nuclide	% activity
Zn-65	88.8
Na-22	11.2

Table 10
Most conservative fingerprint for the 3y-10y waiting time period (FOM = 20.6). Figures represent activated stainless steel (160 MeV, beam impact area, 1 year irradiation time, 3 years waiting time).

Nuclide	% activity
Mn-54	100

Table 11
Most conservative fingerprints for the 10y-30y waiting time period (FOM = 32.8). Figures represent activated concrete (160 MeV, 10 cm lateral to target, 30 years irradiation time, 30 years waiting time).

Nuclide	% activity
H-3[a]	98.2
C-14[a]	1.0
Eu-152	0.2
Ba-133	0.2
Ba-137m	0.2
Na-22	0.1
Ti-44	0.1

[a] Difficult to measure nuclides.

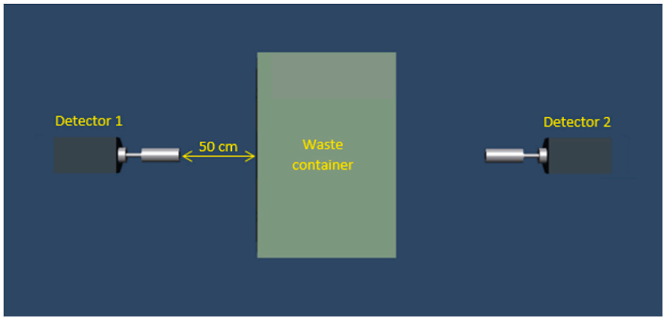

Fig. 1. Scanning geometry for gamma spectroscopy.

Bronson et al., 1997, and Menaa et al., 2011] from Mirion Technologies (Canberra) is used in the laboratory for creating customized efficiency calibrations. The tool undergoes a full factory characterization at the production factory using NIST-traceable radioactive sources. The detector modelling uncertainties range from 5% at 50 keV to 3% at 1332 keV. ISOCS overcomes the limitations of traditional efficiency calibration techniques by replacing the need to use radioactive sources by practical modelling of the assayed item. The geometry parameters such as dimensions, material compositions, densities, detector position and orientation, container filling heights and activity distribution within the item, can be accurately modelled to reduce, as much possible, the uncertainties related to the geometry description.

The minimum detectable activity (MDA) by gamma spectroscopy (International Organization for Standardization, 2019, Joint Committee for Guides in Metrology, 2008) for a given nuclide, is a function of various parameters, including the assay geometry, mass density of material and counting time. Bearing in mind the summation rule for clearance (Equation (1)), the MDA for individual nuclides must be well below the clearance limit (ideally <10% of the limit). Otherwise, the sum of CL fractions for a waste package could exceed unity based on MDA activities alone, even if no radioactive nuclides are present. Due to the low mass density of burnable waste packages, the counting time required to bring the MDA for some nuclides (especially Zn-65 and Eu-154) down to <10% of the clearance limit could exceed 48 h. This was considered impractical, given the number of B-FREE waste containers to be measured and the competing demands for the gamma spectroscopy laboratory. For this reason, gamma spectroscopy could not be used to classify B-FREE waste, but was retained to provide further quality assurance for a sample of the most active grid boxes. In this case, the aim was to check for the presence of unexpected nuclides which could indicate problems with the methodology and radionuclide inventory.

The activity of nuclides considered difficult to measure (DTM) by gamma spectroscopy (e.g. pure beta emitters such as H-3) was estimated using scaling factors. These are defined as the ratio of activity between a given DTM nuclide and an easy to measure 'key nuclide' (KN). In this case, Na-22 was chosen as key nuclide due to its reliable production in activated metals and concrete, and easily detectable gamma emissions. Both DTM and KN activities were obtained from the ActiWiz simulations used to define the radionuclide inventory and fingerprints. Scaling factors were only calculated for DTM nuclides included in the radionuclide inventory shown in Tables 8 and *i.e.* H-3 and C-14. Activity ratios were calculated for each of the 700 irradiation scenarios, in each of the three waiting time periods described in the Radionuclide Inventory section above. The distribution of activity ratios varied considerably according to the combination of DTM nuclide, KN, material, waiting time and particle energy, with some displaying a log-normal distribution, while others exhibited pronounced multi-modal behaviour (see Figs. S13–S16, supplementary materials).

Scaling factors were therefore calculated from the 99th percentile of the distribution of activity ratios (Table 12), as this is less sensitive to the distribution. Where a scaling factor was defined for multiple materials (*e.g.* H-3/Na-22 for both aluminium and concrete), the higher of these was chosen. In basing scaling factors on the 99th percentile activity ratio, our aim was to be conservative, rather than accurate. For example, the 99th percentile activity ratio is higher than the medium by a factor of 4–19 for H-3/Na-22.

6. TFA burnable waste validation study

To validate the radionuclide inventory shown in Table 8, a sample of 50 drums of compacted burnable waste earmarked for TFA disposal were scanned with *in toto* gamma spectroscopy. The counting time was 15 min, which was sufficient for TFA elimination. An additional 12 drums were scanned in groups of four using two gamma spectrometers with an extended counting time of 900 min (overnight). This was done to reduce the minimum detectable activity and increase the ability to detect unexpected nuclides. Activities were quoted with 2σ uncertainties, which take into account numerical approximations related to the efficiency calibration, along with uncertainties from peak area and emission intensities.

A summary of the results for the first batch of 50 drums is shown in Table 13. All detected nuclides were included in the radionuclide inventory, except for Sb-124. The activity (+2σ) of this nuclide was very low (2.58×10^{-2} Bq/g) and well below its clearance limit (1.0 Bq/g) [Swiss Federal Council, 2018]. Potentially, Sb-124 could originate from lead, but none of the other characteristic lead activation products (Table 8), most notably Bi-207, were detected. A more likely source of Sb-124 could be traces of antimony found in circuitry, cable sheathing or fire retardants.

In the second batch with the extended counting time, Na-22, Mn-54

Table 12
Scaling factors for potentially clearable burnable waste produced at CERN. Figures represent the 99th percentile of the H-3/Na-22 or C-14/Na-22 ratio for all scenarios in each waiting time (WT) period.

WT period	H-3	C-14
6 m – 3 y	13.3	3.06E-02
3y – 10 y	65.5	0.20
10 y – 30 y	4359	40.6

Table 13
Summary of gamma spectroscopy of 50 drums of compacted burnable waste, with a counting time of 15 min each.

Nuclide	Number of drums with recorded activity	Mean activity +2σ (mBq/g) [min - max]	Mean MDA (mBq/g)
Na-22[a]	21 (42%)	98.8 [16–518]	21.5
Mn - 54[a]	6 (12%)	48.3 [12.1–126]	21.5
Co-57	2 (4%)	21.1 [0–45.7]	25.2
Co-60[a]	23 (46%)	93.6 [6.4–908]	19.2
Zn-65	1 (2%)	69.6 [69.6–69.6]	56.1
Sb-124	1 (2%)	25.8 [25.8–25.8]	19.3
Cs-137	1 (2%)	14.9 [14.9–14.9]	21.9
Ba-133	1 (2%)	62.7 [62.7–62.7]	28.6
Eu-152	2 (4%)	251 [54.8–446]	65.9

[a] Expected nuclide (see Table 7).

and Co-60 were detected by both spectrometers for each set of four drums, while Zn-65 was detected by both meters for one set of drums, and Eu-152 and Ba-133 were detected by a single meter for two sets of drums (Table 14). No other nuclides were detected.

7. B-FREE pilot study

Twenty five 0.75 m^3 metallic cages known as 'grid boxes' containing burnable waste meeting the B-FREE candidate criteria were measured using total gamma counting and *in toto* gamma spectroscopy. The fingerprints for the 10–30 years waiting time period (Table 11) were used for all grid boxes. This fingerprint was chosen due to the potential waiting time of waste, rather than because we believed the waste to be contaminated with activated concrete (which the 10–30 year fingerprint represents).

Fig. 2 shows CL_{sum} for both methodologies. All values include 2σ uncertainties. In the case of gamma spectroscopy, minimum and maximum values are given. The former is calculated using the activities of detected nuclides only. If the detected activity *without* 2σ was lower than the MDA, the latter value was used instead. The maximum GS value in Fig. 2 corresponds to either detected (+2σ) or MDA activities for nuclides not detected.

For TGC, CL_{sum} was <1 for 18 grid boxes (13 m^3, 505 kg). For gamma spectroscopy, the lower limit of CL_{sum}, based on measured values only, was <1 for 25 (100%) of grid boxes, while the upper limit, based on measured/MDA GS activities, was <1 for 12 (48%) grid boxes. This means that for the remaining 13 boxes, the range of CL_{sum} values included unity, meaning gamma spectroscopy was inconclusive despite the long containing time. Importantly, CL_{sum} for TGC always exceeded the corresponding figure for measured-only gamma spectroscopy. In two grid boxes, CL_{sum} for TGC was >1, while the upper limit for gamma spectroscopy was <1. This suggests that TGC was overly conservative in these cases, meaning clearable waste was classed as radioactive.

Including activities of difficult to measure (DTM) nuclides increased CL_{sum} for GS by an average factor of 3.7. The contribution of DTM nuclides to CL_{sum} depends on how conservative their scaling factors are (in our case, they were set to the 99th percentile), and on the detected activity of their respective key nuclides. In particular, the application of these scaling factors can lead to values of CL_{sum} that are considerably higher than obtained via TGC. Indeed, for TGC we selected and applied the fingerprint associated with just one activation scenario, *i.e.* the most conservative one in terms of clearance limits and signal in TGC. However, in the case of GS and scaling factors, for each pair of key nuclide and DTM nuclide we chose the 99% most conservative scenario. Overall, in the case of clearance, we believe that the risk of being over-conservative is more acceptable than the risk of being under-conservative.

Table 14
Summary of gamma spectroscopy of 12 drums of compacted burnable waste scanned in three groups of four with, a counting time of 900 min.

Nuclide	No. times detected by at least one spectrometer	Mean activity +2σ (mBq/g) [min - max]	Mean MDA (mBq/g)
Na-22[a]	3	40.2 [7.77–144]	3.1
Mn - 54[a]	3	19.3 [2.57–76.2]	2.2
Co-60[a]	3	15.7 [7.68–25.7]	2.1
Zn-65	2	10 [6.99–13.1]	6.1
Ba-133	1	8.79 [8.79–8.79]	3.4
Eu-152	2	15.4 [7.36–23.4]	7.5

[a] Expected nuclide.

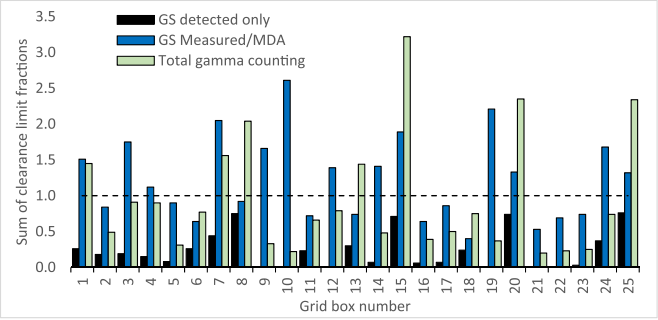

Fig. 2. Sum of clearance limit fractions for a pilot sample of 25 waste packages. The dashed line shows the clearance limit of unity.
GS: gamma spectroscopy.

Table 15 shows the number of grid boxes in which a given nuclide was detected by gamma spectroscopy, along with the maximum recorded activity (+2σ). The nuclides Ba-133, Cs-137, Eu-152 and Eu-154 are suggestive of the presence of activated concrete. The presence of Bi-207 could be due to the presence of lead, though a more likely source is bismuth impurities in metal or grease. No other activation products uniquely characteristic of lead where observed. The presence of Ag-110m is most likely due to silver in copper or circuitry [Zaffora 2016].

8. Discussion

We have developed a methodology for the radiological characterization of burnable waste potentially contaminated with activated particle accelerator materials. While conservative (in the sense that it likely overestimates the sum of clearance limit fractions), this methodology still allowed the clearance of the majority of burnable waste in our pilot project. By identifying the most conservative activation scenario, in terms of clearance limit fraction per clearance monitor response, and including 2σ measurement uncertainties, our methodology does not require additional safety margins. As long as LL_{sum} is below unity, the waste can be cleared.

Although we do not recommend using the same scaling factors and fingerprints outside CERN, the general approach described in this paper could be applied to other similar facilities. The finding that the lowest energy currently used at CERN (*i.e.* 160 MeV for Linac4) yields the most conservative activation scenario is therefore of particular relevance.

There are several sources of uncertainty in this study. Variation in the chemical composition of materials, compared to the compositions in Tables 2–6, may result in additional nuclides being included in the radionuclide inventory, and different fingerprints and scaling factors. To investigate this, we performed a sensitivity analysis by running ActiWiz simulations with various other types of aluminium, copper and stainless steel known to be used at CERN. Of the steel (304L, 316L and 316LN) and aluminium types (6060 and 6082) most commonly used at CERN, the figure of merit defined using Equation (1) was almost identical. For 316Ti steel, Fe-55 and Ni-63 were required to reach 90% of the sum of clearance limit fractions for some scenarios, thus would join the radionuclide inventory. Usage of this type of steel at CERN is thought to be

Table 15
Results of gamma spectroscopy of 25 burnable waste packages. Activity figures are presented as mean [min, max].

Measured only	Grid boxes with recorded activity	GS Detected only (mBq/g)	GS MDA (mBq/g)	GS MDA or detected** (mBq/g)	Total gamma counting
Na-22	15	12.0 [2.1–37.8]	6.9 [1.4–18.0]	10.6 [2.1–37.8]	–
Al-26	0	–	4.3 [1.0–10.9]	4.3 [1.0–10.9]	–
Ti-44	0	–	7.9 [1.9–19.6]	7.9 [1.9–19.6]	–
Sc-46	0	–	6.3 [1.6–15.4]	6.3 [1.6–15.4]	–
Mn-54	3	6.4 [2.4–10]	6.4 [1.6–15.7]	6.7 [1.6–15.7]	–
Co-56	1	2.1	6.0 [1.5–14.9]	6.0 [1.5–14.9]	–
Co-57	2	6.1 [4.8–7.5]	5.8 [2.1–11.1]	5.8 [2.1–11.1]	–
Co-58	0	–	6.1 [1.5–14.4]	6.1 [1.5–14.4]	–
Co-60	18	9.1 [2.5–22.3]	6.0 [1.1–15.1]	8.6 [2.5–22.3]	–
Zn-65	2	10.7 [6.4–15]	17.5 [3.6–44.3]	17.7 [4.3–44.3]	–
Ag-110m	1	9.1	6.3 [3.5–9.1]	6.3 [3.5–9.1]	–
Cs-137	1	20.1	6.2 [3.1–9.4]	14.7 [9.4–20.1]	–
Ba-133	4	10.2 [2.2–28.7]	8.0 [2.2–18.1]	8.9 [2.2–28.7]	–
Eu-152	4	19.7 [11.3–27]	15.9 [5.5–33.1]	16.6 [5.5–33.1]	–
Eu-154	4	12.2 [6.1–16.8]	11.8 [4.1–23.4]	11.9 [6.1–23.4]	–
Au-194	0	–	8.0 [2.3–18.1]	8.0 [2.3–18.1]	–
Bi-207	2	3.8 [3.3–4.4]	5.7 [1.4–13.2]	5.8 [2.5–13.2]	–
H-3*	–	52206 [9301–164984]	30000 [6198–78506]	46038 [9301–164984]	–
C-14*	–	486 [87–1537]	279 [58–731]	429 [87–1537]	–
CL_{sum} no DTMs		0.2.0 [0–0.77]	1.13 [0.30, 2.62]	1.22 [0.42, 2.62]	0.95 [0.20–3.22]
CL_{sum} with DTMs		0.84 [0–3.95]	1.71 [0.42, 4.14]	2.11 [0.60, 5.06]	NA

*Difficult to measure (DTM), ** Detected activities are used if higher than MDA, otherwise MDA activities are used. GS = gamma spectrometry.

limited, however. With the exception of CuZn05 and CuAI10FeNi5C copper, which include around 0.5% zinc, the FOM for other types of copper was almost identical. None of these other types of aluminium, copper or steel would result in more conservative fingerprints than those presented in Tables 9–11. Overall, these results suggest our classification methodology is robust with respect to deviation from expected material composition.

The choice of fingerprints was based on the assumption that no lead is present in burnable waste packages (with the exception of small lead impurities, *e.g.* in copper). Activation of lead results in the production of large quantities of Tl-204, leading to exceptionally conservative fingerprints (max FOM of 334). The no-lead assumption is supported by the results of gamma spectroscopy. Of the nuclides produced in activated lead but not aluminium, copper, steel or concrete (see Table 7), only Bi-207 was detected (in very small quantities).

A more likely explanation for the presence of Bi-207 is the presence of impurities of bismuth itself in metals or grease. Isolated signals of nuclides not included in the radionuclide inventory (including Sb-124 and Ag-110m) were not considered sufficient to justify changing the fingerprints, *e.g.* to those based on lead. We must, however, be alert to the potential presence of lead activation products, which is why we have implemented gamma spectroscopy quality controls. We were not able to use gamma spectroscopy to classify potentially clearable waste, *i.e.* calculate CL_{sum}, as the MDA values for some nuclides was too high, even with extended counting times.

9. Conclusion

The radiological characterization of burnable waste with the goal of clearance is especially challenging due to (1) the varied nature of potential contaminating materials and associated irradiation conditions, (2) the long gamma spectroscopy counting times required to ensure minimum detectable activities are well below the clearance limit. The methodology described in this paper, while radiologically conservative, still allowed the free release of the majority of candidate burnable waste at CERN. With suitable adaptations for local conditions and materials, our methodology could potentially be used for similar clearance campaigns at other accelerator facilities with isotope production predictive tools and total gamma counting equipment.

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CRediT authorship contribution statement

Richard Harbron: Writing – review & editing, Writing – original draft, Methodology, Investigation, Formal analysis, Data curation. **Renaud Charousset:** Resources, Methodology, Data curation. **Gerald Dumont:** Writing – review & editing, Supervision, Project administration, Conceptualization. **Matteo Magistris:** Writing – review & editing, Writing – original draft, Supervision, Methodology, Formal analysis, Data curation, Conceptualization. **Nabil Menaa:** Writing – review & editing, Validation, Formal analysis. **Paolo Giunio Pisano:** Writing – review & editing, Writing – original draft, Project administration, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Christian Theis:** Writing – review & editing, Writing – original draft, Supervision, Software.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.apradiso.2023.110782.

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