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1 **Design and development of a portable β - spectrometer for** 2 **^{90}Sr activity measurements in contaminated matrices**

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10

11 **ABSTRACT**

12 Determining the nature and activity of radionuclides is an important task in the nuclear industry. Pure
13 β - emitters are hard to characterize because the mean free path of electrons in dense matter is very
14 short. Most measurement techniques are therefore based on destructive laboratory analyses. This
15 article presents a non-destructive approach based on a portable β - spectrometer, designed to provide
16 qualitative and quantitative information on pure β - emitters, notably ^{90}Sr . Building on existing
17 methods focused on the measurement of ^{90}Sr activity in natural soil contaminated by the Chernobyl
18 accident, the aim of this project is to develop a detector for the radiological characterization of
19 different types of contaminated matrices, in particular the contaminated concrete structures typically
20 found in nuclear facilities. A measurement device equipped with an EJ200 plastic scintillator was
21 designed using Monte Carlo simulations with the MCNP6 and PENELOPE calculation codes. The
22 energy calibration and the response of the detector were determined using experimental measurements
23 and MCNP simulations of laboratory configurations of standard β - sources. These data were used to
24 validate the model of the detector, as well as to determine calibration coefficients by numerical
25 simulation for various on-site measurement configurations. The device was then used to characterize
26 samples of sand from a decommissioning site in France (Fontenay-aux-Roses). The resulting ^{90}Sr
27 activities are within experimental uncertainties of those obtained using destructive measurements.
28 These results are promising for wider applications on nuclear decommissioning sites.

29

30 *Keywords:* ^{90}Sr ; Beta spectrometry; Decommissioning; Contaminated matrices; Monte Carlo;

31 Scintillation detector

32 *Non-standard abbreviations:* none.

33 *Declarations of interest:* none.

34 **1. Introduction**

35 The number of nuclear decommissioning sites is ever increasing worldwide. The
36 management of radioactive information is a crucial aspect of many of the major components
37 of these projects: radioactive waste management, choice of decontamination solutions,
38 operator radioprotection. The radiological state of buildings and equipment can be established
39 by combining non-destructive on-site radioactivity measurements with numerical modeling.

40 The characterization of γ emitters can be done by gamma-ray spectrometry, combined
41 in some cases with gamma imaging. In simple situations in which the characteristic spectrum
42 is known, the activity of all the other radionuclides, notably pure β - emitters, can be
43 determined using data on the main γ emitters such as ^{137}Cs and ^{60}Co , which act as radioactive
44 tracers. This procedure is unfeasible however in more complicated cases, such as for soils or
45 buildings, in which the $^{137}\text{Cs}/^{90}\text{Sr}$ ratio can vary substantially. In the soils and rivers
46 contaminated by the Chernobyl accident for instance, $^{137}\text{Cs}/^{90}\text{Sr}$ ratios were found to vary
47 from 0.5 to 0.01 [1], in part because the two radionuclides have different transport rates in
48 these materials. The radiological inventory becomes very uncertain or even impossible to
49 draw up and an alternative approach is required.

50 Pure β - emitters such as ^{90}Sr cannot be detected using gamma spectrometry. Currently,
51 measurements of ^{90}Sr activity are almost exclusively performed by radiochemical analysis of
52 extracted samples. These radiochemical analyses have several drawbacks. First, they are

53 destructive and involve several sample preparation steps (dilution, concentration, separation
54 of the interfering radionuclides) that are problematic for the representativity of the samples.
55 For the results to be interpretable moreover, the analyses have to be repeated on a large
56 number of samples, which can be difficult or impossible to obtain. Finally, the analyses are
57 lengthy and costly. To complement these destructive measurements therefore, there is
58 considerable interest in developing spectroscopic methods to detect and quantify ^{90}Sr on site.
59 ^{90}Sr is one of the main pure beta emitting radionuclides found in nuclear dismantling sites and
60 given its radiotoxicity, locating and quantifying ^{90}Sr activity is essential to evaluate
61 radiological impact on workers. It is in this context that non-destructive techniques to measure
62 ^{90}Sr activity in contaminated soils were developed by different research laboratories
63 worldwide [1–13].

64 Building on existing methods, the objective of the study reported here was to develop
65 a device for on-site characterization of soils, building structures (e.g. concrete shells) and
66 samples contaminated with ^{90}Sr . More specifically, the project was to develop a dedicated β -
67 spectrometer to detect and quantify ^{90}Sr via its decay product ^{90}Y by rigorously modeling the
68 detector's response and accounting for contribution from γ photons and unwanted β - particles.

69 The first part of the article presents the measurement method, the dimensioning studies
70 carried out using the particle transport codes MCNP6 [14] and PENELOPE [15], and the
71 design of the detector with an EJ200 plastic scintillator. The electronics of the device and the
72 spectrometric chain are also described. The second part focusses on the energy and efficiency
73 calibration of the β - spectrometer as well as on the determination of calibration coefficients by
74 numerical simulation for various on-site measurement configurations. This involved
75 comparing the theoretical response calculated using MCNP simulations to experimental
76 measurements obtained using standard β - sources. The third part of the article reports and
77 discusses the results obtained using the device for the quantification of ^{90}Sr activity in

78 samples of Fontainebleau sand from the CEA (French Alternative Energies and Atomic
79 Energy Commission) site at Fontenay-aux-Roses in comparison with radiochemical analyses.

80 **2. Materials and methods**

81 *2.1. β - spectrum acquisition and interpretation*

82 The measurement technique used here is based on the work of Chesnokov and al. [2]
83 and Potapov and al. [3] to characterize soils contaminated by the Chernobyl nuclear accident.
84 More recently, Slaninka and al. [4] have compared this non destructive method with
85 radiochemical analyses. The principles are outlined here as they underpin the work reported in
86 this article. More detailed descriptions can be found in the original references.

87 *2.1.1. ^{90}Sr decay*

88 Strontium-90 is an unstable nucleus with an excess of neutrons that undergoes
89 spontaneous β^- decay. The parent nucleus decays to ^{90}Y with a half-life of 28.8 years,
90 emitting electrons with a maximum kinetic energy of 0.546 MeV [16]. This intermediate
91 nucleus then decays with a half-life of 64 hours either to an excited state of ^{90}Zr by emitting
92 an electron with a maximum energy of 0.518 MeV (~0.017% probability), or directly to stable
93 ^{90}Zr by emitting an electron with a maximum energy of 2.280 MeV. In the latter case, the β^- -
94 radiation emitted is more energetic and therefore more easily detected and measured than the
95 initial emission from ^{90}Sr . Since both nuclides are at secular equilibrium, ^{90}Sr activity can be
96 evaluated through the β^- particles emitted by its daughter nucleus, ^{90}Y .

97 *2.1.2. Spectrum acquisition*

98 The nuclear activity of ^{90}Sr can be determined by acquiring two separate spectra with a
99 scintillation detector. For the first, the main measurement, the detector is placed very close (a

100 few millimeters) from the surface of the studied material so that the β^- and γ rays are
101 absorbed by the scintillating material in the detector. This deposition of energy leads to the
102 emission of light photons that are directed through an optical waveguide to a photomultiplier
103 tube. The second or auxiliary spectrum is recorded with an aluminum cover placed between
104 the detector and the sample. This blocks the β^- particles but not the γ rays emerging from the
105 studied area such that only the latter enter the detector. Subtracting the second spectrum with
106 appropriate processing yields the net β^- signal from which the ^{90}Sr activity can be estimated.

107 *2.1.3. Spectrum processing and interpretation*

108 *2.1.3.1. Unwanted signals from other radionuclides*

109 Since ^{90}Sr is a fission product, it is almost always found in nuclear installations with
110 other fission products, notably ^{137}Cs , which undergoes β^- decay to metastable $^{137\text{m}}\text{Ba}$ with a
111 half-life of 30,05 years [16]. The other main artificial radionuclide that emits both β^- and γ
112 rays is ^{60}Co , produced by neutron activation from ^{59}Co in nuclear reactors [16]. Natural
113 radionuclides may also interfere with the measurements. In soil and concrete, the main natural
114 radioisotopes are ^{40}K and those in the ^{238}U , ^{235}U and ^{232}Th series which are all mixed alpha,
115 beta and gamma emitters. According to the IAEA, the concentration ranges of ^{40}K , ^{232}Th and
116 ^{226}Ra in common types of concrete are about 5–1,570 $\text{Bq}\cdot\text{kg}^{-1}$, 1–190 $\text{Bq}\cdot\text{kg}^{-1}$ and 1–250
117 $\text{Bq}\cdot\text{kg}^{-1}$, respectively [17]. Signals from natural radionuclides, are thus only important to
118 consider for low levels of ^{90}Sr contamination, in particular the $^{234\text{m}}\text{Pa}$, a ^{238}U daughter, which
119 emits an electron with a maximum energy of 2.280 MeV [16], i.e. same as ^{90}Y . The clearance
120 level set by the European Commission for building rubble from nuclear installations is of the
121 order of 1000 $\text{Bq}\cdot\text{kg}^{-1}$ [18]. For walls and floors, the difficulty lies in locating the
122 contamination and identifying its surface or volume within the background contribution from
123 mixed β^-/γ emitters, retaining only the contribution from ^{90}Sr .

124 2.1.3.2. *Spectrum analysis*

125 To eliminate this background contribution, two key facts are that the components of
126 the β - and γ spectra below 0.7 MeV come mainly from ^{137}Cs contamination, while those
127 above 0.7 MeV come from natural radionuclides and ^{60}Co if present. Therefore, the high-
128 energy β -- background in the spectra from natural radionuclides can be estimated by
129 correlating their β -- spectrum with the γ component above 0.7 MeV. The number of electrons
130 and photons in the background noise over an energy interval ΔE_γ are correlated as follows
131 [2,3]:

$$N_e = \sigma^* N_\gamma + \Delta\sigma N_\gamma^U \quad (1)$$

132 with $\sigma^* N_\gamma$ being the correlation between the number of γ photons and electrons from the
133 background noise and $\Delta\sigma N_\gamma^U$ the correlation that takes account of natural radioactivity.

134 2.1.4. *Determining ^{90}Sr activity*

135 Once the background contribution has been eliminated, the activity of ^{90}Sr can be
136 calculated using Eq. (2):

$$A_{^{90}\text{Sr}} = \{ [N_{e\gamma}(\Delta E_e) - N_\gamma(\Delta E_e)] - [\sigma^* N_\gamma(\Delta E_\gamma) + \Delta\sigma N_\gamma^U] \} \times C \quad (2)$$

137 Here, $N_{e\gamma}(\Delta E_e)$ and $N_\gamma(\Delta E_e)$ are the count rates measured respectively without and with the
138 aluminum cover in the energy range ΔE_e , $[\sigma^* N_\gamma(\Delta E_\gamma) + \Delta\sigma N_\gamma^U]$ is the count rate from
139 background noise in the same energy domain, and C is a calibration coefficient that converts
140 the measured count rate into ^{90}Sr activity. It is equal to the count rate that would be measured
141 for unit activity of $^{90}\text{Sr}/^{90}\text{Y}$ in the studied concrete or soil.

142 Potapov and al. [3] defined the following energy ranges : $\Delta E_{e1} = 0.9\text{--}2.0$ MeV, $\Delta E_{e2} =$
143 $1.2\text{--}2.0$ MeV and $\Delta E_\gamma = 1.09\text{--}2.00$ MeV. The first interval (ΔE_{e1} , shown in green in Fig. 1) is

144 designed to minimize statistical uncertainties in the measured ^{90}Sr activity due to the presence
145 of ^{137}Cs [3]. This was the range used by default in the work reported here. The second interval
146 (ΔE_{e2}), with a lower limit shifted up to 1.2 MeV is more appropriate in the presence of high
147 ^{60}Co concentrations. The γ -ray interval (ΔE_{γ}) is designed to minimize background β -
148 radiation noise from natural radionuclides.

149 *2.2. Dedicated detector design*

150 *2.2.1. Main β - particle detection techniques*

151 Beta particles can be detected destructively in dissolved samples using a liquid
152 scintillation counter [19]. Another standard approach involves two superposed passivated
153 implanted planar silicon detectors in a vacuum chamber [9]. This is routinely used in α
154 spectrometry and has been developed for environmental matrices to detect and quantify
155 natural and artificial β - emitters [10]. In the same way, Sokolov and al. [11] have developed a
156 SiLi detector inside a vacuum chamber to detect both beta and X-ray radiation from different
157 kind of samples. However, the design of these detection unit severely limits their
158 applicability for in situ measurements. Kadenko and al. have developed a portable β -
159 spectrometer consisting of an activated polystyrene scintillator, a photomultiplier tube, and a
160 multichannel analyzer [12]. The ^{90}Sr activity can be determined from the count rate measured
161 in the region of interest 1311-2282 keV. The US Department of Energy [8] have developed a
162 BetaScint fiber-optic sensor for detecting ^{90}Sr and ^{238}U in soil. It consists of a multi layers of
163 scintillating fiber optic arrays, which affords some discrimination against low energy beta
164 particles, gamma rays and cosmic-rays by measuring coincident events. Due the large area of
165 the BetaScint, the measurements are more representative of the contamination in the soil
166 against laboratory measurements on samples. The main limitation of the BetaScint is that the
167 beta radiation from ^{90}Sr and ^{238}U cannot be discriminated. Finally, the combined scintillators

168 with different pulse shape characteristics in phoswich (phosphor sandwich) detectors allow
169 low-energy radiation (γ , X-ray, α or β -) to be distinguished from the background noise thanks
170 to pulse shape (in particular response time) analysis. Farsoni and Hamby have developed a
171 real-time detector for simultaneous γ and β - spectrometry, with three scintillators—a plastic
172 scintillator in front of $\text{CaF}_2(\text{Eu})$ and $\text{NaI}(\text{Tl})$ crystals—and a customized digital pulse
173 processor [13].

174 2.2.2. *Study design*

175 The objective of this part of the study was to dimension a device able to provide
176 analyzable β - spectra to estimate ^{90}Sr activity in the studied object (concrete structure,
177 contaminated sample, filter, etc.), by characterizing the source term, choosing the detector,
178 and designing the aluminum cover. For the source term, the task was to predict the paths and
179 energy distribution of electrons emerging from concrete structures contaminated with ^{90}Sr and
180 potentially interacting with the detector. This involved calculating the path length of electrons
181 emitted by ^{90}Y in various concrete matrices of different densities in comparison with
182 previously published results for contaminated soils [2,3]. Choosing an appropriate detector is
183 crucial as it governs the measurement performance of the detector. The main criteria
184 considered were the detection efficiency, sensitivity to γ rays, and the electron backscattering
185 coefficient. Several materials were considered and electron and γ responses of two plastic
186 scintillators, the BC400 (Saint-Gobain, France) and EJ200 (Eljen Technology, TX, USA),
187 were compared using Monte-Carlo simulations. Finally, the design and dimensions of the
188 aluminum cover used to block electrons during the auxiliary measurement were optimized.
189 The influence on incident β -- radiation of the window protecting the scintillator from ambient
190 light and humidity (the material is hygroscopic) was quantified and minimized. Simulations
191 were performed mainly with MCNP6 [14] and validated using PENELOPE [15].

192 2.2.3. *Electron path length in soil and concrete contaminated with ^{90}Sr*

193 To estimate the maximum measurable depth of ^{90}Sr contaminations, electron paths
194 through various soil or concrete shells were modeled using MCNP6 simulations. A surface
195 source of β^- radiation ($10 \times 10 \text{ cm}^2$) was placed under layers of non-contaminated material
196 (soil or concrete) with different thicknesses. The β^- particles were emitted vertically with an
197 energy distribution corresponding to those emitted by ^{90}Y ($E_{\text{max}} = 2.280 \text{ MeV}$). An F1 surface
198 current tally was performed on the upper surface of the uncontaminated matrix, giving the
199 number of electrons that pass through the matrix layer. Fig. 2 shows that the proportion τ of
200 electrons generated by the source that penetrate through the layer depends on its thickness and
201 density. Above 5 mm of concrete and 7 mm of soil, the proportion of electrons passing
202 through the layer becomes negligible. These results of simulation were confirmed by
203 calculating the range of electrons with an energy $Q_{\beta} = 2.280 \text{ MeV}$ (the maximum energy of
204 electrons emitted by ^{90}Y) using the Flammersfeld equation [20] which gives respective path
205 lengths of 0.71 and 0.47 cm in soil ($1.52 \text{ g}\cdot\text{cm}^{-3}$) and concrete ($2.3 \text{ g}\cdot\text{cm}^{-3}$).

206 2.2.4. *Components and dimensions of the β^- spectrometer*

207 2.2.4.1. *Active part of the detector*

208 Plastic scintillators like BC400 (Saint-Gobain, France) or EJ200 (Eljen Technology,
209 TX, USA), both based on polyvinyltoluene, are well adapted for the detection of electrons
210 [21,22]. Indeed, since they are composed mainly of carbon and hydrogen, they have a low
211 mean atomic number, meaning that very few incident electrons are scattered, γ photons do not
212 deposit much energy, and bremsstrahlung is negligible.

213 2.2.4.2. *Optimal plastic scintillator thickness*

214 Electron interactions in the plastic scintillator were simulated using MCNP6 and
215 PENELOPE to calculate the energy deposited in the detector as a function of its thickness.
216 The model consisted of a square 10×10 cm² concrete slab, 0.5 cm thick (the maximum range
217 of β^- particles from ⁹⁰Y), with different thicknesses of scintillating plastic placed on top,
218 separated by a 1 mm thick layer of air. The ⁹⁰Y source term was homogeneously distributed in
219 the concrete. Fig. 3 shows how the energy deposited varies as a function of the thickness of
220 the scintillator as estimated using an F8 tally with MCNP6. The results with EJ200 and
221 BC400 are almost identical because their compositions are similar. The energy deposited in
222 the scintillator increases before leveling out at a thickness of about 10 mm, which corresponds
223 to the path length of β^- particles with an energy of 2.280 MeV in the scintillator material
224 [20]. However, since 95% of the energy is deposited in the first 4 mm, this thickness was
225 chosen as a good compromise between maximizing the detection efficiency of β^- particles
226 from ⁹⁰Y and minimizing the sensitivity to gamma photons.

227 2.2.4.3. *Optimal window design*

228 The window acts as a protective film, limiting interference from ambient light and
229 preventing direct contact between the sensitive part of the detector and the studied material. It
230 must nonetheless be thin and as transparent as possible to electrons. To limit electron energy
231 loss on entering the device, we investigated the effects of a 18 μ m Mylar film as a potential
232 improvement on the 50 μ m titanium sheet used in the device developed in references [2,3].
233 Mylar (biaxially oriented polyethylene terephthalate) is a polyester film commonly used to
234 protect scintillation detectors. We performed MCNP6 simulations with a cylindrical detector
235 ($\varnothing = 8$ cm) and a 5 mm thick cylindrical layer of concrete emitting β^- particles with the
236 energy distribution of ⁹⁰Sr/⁹⁰Y electrons. Fig. 3 shows that Mylar is more transparent to

237 electrons than titanium, absorbing about 10 keV less energy at the same thickness. As
238 expected furthermore, the most transparent film is the thinnest, so the best choice is a Mylar
239 film with a thickness of 18 μm which also enable the detector to be light-tight.

240 *2.2.4.4. Optimal aluminum cover thickness*

241 As mentioned above, aluminum shielding is required to take auxiliary readings of the
242 background γ radiation. The low atomic number of aluminum is advantageous for this
243 purpose as it interacts weakly with γ photons, with little bremsstrahlung, while being effective
244 at stopping electrons. According to Flammersfeld equation [20], the path length of β^-
245 particles with an energy of 2.280 MeV in aluminum ($2.698 \text{ g}\cdot\text{cm}^{-3}$) is 0.408 cm. A 4 mm
246 thick sheet of aluminum should therefore stop all the electrons emitted by ^{90}Y . To verify this,
247 the simulations reported in the previous section were repeated with aluminum layers of
248 different thicknesses inserted between the concrete and the Mylar film. The results obtained
249 with MCNP6 and PENELOPE, compared in Fig. 4, are very close. The energy deposited in
250 the detector decreases from about 60 keV without the shield (Fig. 3) to 1 keV with a 2 mm
251 thick shield and becomes negligible for thicknesses of 4 mm and above (Fig. 4). A thickness
252 of 4 mm was therefore chosen for the aluminum cover used in practice.

253 *2.2.4.5. Optimal β^- spectrometer design*

254 The detector used in the rest of the study was custom-built by the manufacturer
255 Scionix (Bunnik, The Netherlands) using the results of the dimensioning studies described
256 above. The device consists of a 4 mm thick EJ200 plastic scintillator with an 18 μm thick
257 Mylar window. The scintillator is attached to a PMMA light guide that directs the photons to
258 the photocathode of a photomultiplier. The detector was attached via a DSA1000
259 multichannel analyzer (MIRION Technologies, CA, USA) to a computer running the software

260 Génie 2000 [24], which was used to calibrate the device and acquire and analyze the spectra
261 (Fig. 5).

262 **3. Calibration and characterization of the EJ200 detector's response**

263 The next stage of the study was to build a numerical model of the detector with
264 MCNP6 and validate it using spectra measured using calibration sources in controlled setups,
265 minimizing the relative error between simulated and measured spectra. After qualifying the
266 model, Monte Carlo simulations were performed to determine the calibration coefficient C in
267 Eq. (2) for different measurement setups likely to be encountered in the field.

268 *3.1. Experimental measurements using calibration sources*

269 The reference spectra were measured using surface calibration sources, mainly pure β -
270 emitters (^{14}C , ^{36}Cl , $^{90}\text{Sr}/^{90}\text{Y}$ and ^{204}Tl) and a mixed β -/ γ emitter (^{137}Cs). The dynamic range of
271 the spectrometer was calibrated beforehand using a $^{90}\text{Sr}/^{90}\text{Y}$ source placed up against the
272 detector, to limit losses in air. The gain of the device was thus adjusted so that the β -
273 spectrum of ^{90}Y would cover all the 1024 channels of the 10 bits dynamic range.

274 The characteristics of the sources are summarized in Table 1. Their radioactivity was
275 measured at the time of the measurements, namely on 23 June 2015. The ^{36}Cl , $^{90}\text{Sr}/^{90}\text{Y}$, ^{204}Tl
276 and ^{137}Cs sources consisted of a 30 mm diameter active layer obtained by evaporating a
277 solution between two 75 μm thick gold-coated plastic sheets held by a 3 mm thick metal ring
278 [25]. The ^{14}C source had a 51 mm diameter thick active layer, obtained by evaporating a
279 solution deposited on an aluminum sheet, covered by a protective layer [25].

280 The detector was placed at a fixed distance (as close as possible) vertically above each
281 source. To prevent direct contact with the fragile Mylar window, the detector was placed on
282 two 8.5 mm spacers, leaving 9.65 mm of air between the detector and the source. With the

283 ^{137}Cs source, a mixed β -/ γ emitter, auxiliary measurements were taken with the aluminum
284 cover to obtain the β - contribution alone after subtraction. The acquisition time for each
285 spectrum was 10 min to ensure good counting statistics.

286 *3.2. Numerical model of the detector*

287 The method described above for the measurement of ^{90}Sr activity in concrete and soil
288 involves calculating calibration coefficients from Monte Carlo simulations of various
289 measurement setups. The reliability of these simulations depends in turn on the reliability of
290 the numerical model of the detector. The following section describes how the model of the
291 EJ200 detector was built and validated using MCNP6 simulations and the reference spectra.

292 *3.2.1. Modeling the EJ200 detector using MCNP6*

293 The model was built using data provided by Scionix on the nature and dimensions of
294 the different components and by LEA (Pierrelatte, France) on the characteristics of the
295 calibration sources. Since the path length of the electrons is short, the parts of the detector
296 beyond the light guide have little effect on the measurements and were therefore not included.
297 The composition and density of the materials in the model were taken from the Pacific
298 Northwest National Laboratory database [26]. The surface-like radioactive source was
299 modeled as a very thin cylinder of air with a diameter specified by the manufacturer [25] and
300 a uniformly distributed activity. The characteristic β - spectra of each of the radionuclides
301 were modeled by emission probabilities in energy intervals obtained from the Joint Evaluated
302 Fission and Fusion radiological database [27]. A 5 keV energy step was used and continuous
303 spectra were obtained by interpolation using MCNP6.

304 Several features of the radioactive decays were not included in the model. First, for
305 mixed β -/ γ emitters such as ^{137}Cs , the electron spectrum also contains a separate component
306 from the internal conversion process, which occurs in competition with gamma emission.

307 There is also an internal conversion component in the electron spectrum of ^{90}Y , but it is too
308 weak to be included in the model. The emission of internal conversion electrons is followed
309 by a reorganization of the electron shell and the emission of X-rays; this process was ignored
310 in the Monte Carlo simulations. Second, for mixed β - γ emitters, the source of γ rays was not
311 included in the calculations because the corresponding contributions to the spectra are
312 subtracted from the experimental data using the auxiliary spectrum but not necessarily all
313 their beta components. Third, ^{36}Cl and ^{204}Tl decay by electron capture followed by the
314 emission of γ photons if the daughter nucleus is in an excited state, and by the emission of X-
315 rays and Auger electrons as the electron shell reorganizes. Because of the complexity of the
316 source term corresponding to this decay process, it was not included in the Monte Carlo
317 simulations.

318 Fig. 6 shows a cross-section of the setup used for MCNP6 simulations with a ^{137}Cs
319 source. An F8 tally was used to calculate the distribution of electron energy in the sensitive
320 part of the detector (representing the EJ200 plastic scintillator). The PE (photon and electron
321 transport) mode was used to account for the transport of secondary electrons produced by
322 bremsstrahlung radiation and fluorescence X-rays in the material down to a minimum particle
323 energy of 1 keV (the default setting in MCNP6 [6]). Electron transport was modeled using the
324 ITS (Integrated TIGER series) algorithm [28], as an add-on to the code, which results suggest
325 is more accurate than the default MCNP algorithm [29–31]. To ensure meaningful
326 comparisons between the experimental and simulated spectra, the detector was energy
327 calibrated using the same intervals as used for the F8 tally in MCNP6.

328 *3.2.2. Energy calibration of the detector*

329 Energy calibration involves linking each of the detector's channels to an energy. To
330 increase the precision of the calibration, several {energy; channel} pairings are typically used,
331 covering the whole dynamic range. Monoenergetic electrons such as those emitted along with

332 γ rays in internal conversion processes are well suited for this process. In the present work,
333 the detector was energy calibrated using ^{137}Cs and ^{14}C sources.

334 The electron spectrum of ^{137}Cs has characteristic internal conversion peaks at 0.624
335 and 0.656 MeV [27], which were used to define a first {energy; channel} pair. A second pair
336 was obtained from the beta emissions of ^{14}C , whose spectral density is concentrated at low
337 energies (maximum energy, 0.156 MeV) and is therefore well suited to calibrate the channel
338 with the highest counting rate. Note that since the two energy ranges required to calculate ^{90}Sr
339 activities in this approach (Eq. (2)) are quite broad ($\Delta E_{e1} = 0.9\text{--}2.0$ MeV or $\Delta E_{e2} = 1.2\text{--}2.0$
340 MeV), a rough calibration of the spectrometer using just two {channel; energy} pairs,
341 respectively at low and medium energies, was considered adequate.

342 Monte Carlo simulations were used to estimate the energy loss of electrons emitted by
343 the two sources in the air-filled gap separating them from the detector. These simulations
344 yielded the energies corresponding to the maximum counts for the ^{14}C spectrum in channel 18
345 and the internal conversion peak of the ^{137}Cs spectrum in channel 240. For the simulated ^{14}C
346 spectrum, the count rate maximum occurred at 0.049 MeV.

347 The internal conversion electrons emitted by $^{137\text{m}}\text{Ba}$ have energies of 0.624 or 0.656
348 MeV and respective intensities of 8.07 and 1.50% [27]. The beta spectrum of ^{137}Cs should
349 therefore contain two peaks, but only one was observed experimentally, indicating that the
350 energy resolution of the detector is lower than the separation of the two lines. As a first
351 approach, we assumed that the main component of the measured peak was the more intense
352 line at 0.624 MeV. A Monte Carlo simulation was performed without taking the resolution of
353 the detector into account. Fig. 7 shows the energy distribution in the scintillator from the two
354 types of internal conversion electrons (orange line). The maximum count rate occurs at 0.604
355 MeV, indicating that the electrons lose about 20 keV in passing through the layer of air and
356 the Mylar film.

357 The energy calibration equation deduced from the two calibration points, {channel 18;
358 0.049 MeV} and {channel 240; 0.604 MeV}, obtained from the experimental and simulated
359 ^{14}C and ^{137}Cs spectra is thus:

$$E(\text{MeV}) = 0.0025 \times \text{channel} + 0.004 \quad (3)$$

360 3.2.3. Energy resolution parameters

361 In MCNP6 simulations, the detector is assumed to be ideal and the different sources of
362 dispersion of charges are ignored. However, the finite resolution of the detector can be taken
363 into account in the code (using the Gaussian energy broadening (“GEB”) option in the input
364 file) via the following function that gives the resolution of the detector (the full width at half
365 maximum (FWHM) of the peaks) as a function of energy (E) [14]:

$$\text{FWHM} = a + \sqrt{b(E + cE^2)} \quad (4)$$

366 The coefficients a , b and c were determined using the experimental data measured for ^{137}Cs .
367 The experimental data were fitted [32] using a Gaussian function. The FWHM of the peak is
368 about 30 channels, or 0.075 MeV according to the energy calibration equation (Eq. (3)), this
369 means that the energy resolution of the detector is about 12.5%.

370 The three coefficients in Eq. (4) were then determined using three {FWHM; E } pairs,
371 the first at the origin {0 MeV; 0 MeV}, the other two corresponding to the two internal
372 conversion peaks {FWHM = 0.078 MeV; E = 0.624 MeV} and {FWHM = 0.082 MeV; E =
373 0.656 MeV}. Fitting Eq. (4) to these three data points then gives $a = 5.272 \times 10^{-6}$, $b = 6.718$
374 $\times 10^{-2}$ and $c = 1.901$.

375 The spectra simulated with and without this finite detector resolution are compared in
376 Fig. 7. The maxima of both spectra appear at 0.604 MeV, therefore in the same channel, and

377 the energy corresponds to that of the maximum count rate for the most intense internal
378 conversion peak. This comparison thus confirms the hypothesis that the dominant
379 contribution to the experimental peak measured for ^{137}Cs is from electrons emitted at 0.624
380 MeV, and also validates the energy calibration equation (Eq. (3)) obtained without accounting
381 for the energy resolution of the detector.

382 *3.2.4. MCNP6 model qualification*

383 Simulations of the electron energy distribution in the plastic scintillator were
384 performed using MCNP6 for all the calibration sources (^{14}C , ^{36}Cl , $^{90}\text{Sr}/^{90}\text{Y}$, ^{137}Cs and ^{204}Tl),
385 integrating energy intervals and detector resolution as described above. Figs. 8–10
386 respectively compare the measured and simulated spectra of ^{14}C , $^{90}\text{Sr}/^{90}\text{Y}$ and ^{137}Cs . The
387 simulated spectra reproduce the main features of the experimental spectra. The maxima occur
388 at the same energies and the high-energy tails of the simulated and experimental spectra
389 overlap, tending to zero at the same energies. These results indicate that the energy calibration
390 of the device is accurate over the whole measurement range.

391 The spectra were compared quantitatively in terms of integrated count rates for
392 different energy intervals. For ^{14}C , ^{36}Cl and ^{204}Tl , the count rates were integrated over the
393 entire measurement range, namely 0.031–0.156 MeV for ^{14}C , 0.031–0.709 MeV for ^{36}Cl and
394 0.031–0.763 keV for ^{204}Tl . For ^{137}Cs , the energy interval (0.508–0.700 MeV) was defined to
395 only consider the internal conversion peak. For $^{90}\text{Sr}/^{90}\text{Y}$ finally, the count rates were
396 integrated over the two energy ranges defined previously, $\Delta E_{e1} = 0.9\text{--}2$ MeV and $\Delta E_{e2} = 1.2\text{--}2$
397 MeV.

398 The relative deviations between the simulated and measured count rates over these
399 energy ranges are listed in Table 2. The simulated spectra tend to underestimate the count
400 rates between 0.03 and 0.7 MeV with a maximum relative deviation of 29%, and overestimate
401 those between 0.7 and 2.2 MeV with a maximum relative deviation of 33%. There are several

402 possible sources of uncertainty in this context: (i) uncertainties from approximating certain
403 calibration sources (^{137}Cs , ^{204}Tl and ^{36}Cl) as pure β^- emitters (see section 3.2.1), (ii)
404 uncertainties from the electron transport model—the transport algorithm used, uncertainties in
405 the nuclear data, in particular the effective interaction cross-sections, and the cut-off used for
406 electron and photon transport (1 keV, the default value), and (iii) uncertainties in the setup
407 geometry—in the thicknesses of the different layers and in the assumed uniformity of the
408 source term distribution.

409 Because of the multiple sources of uncertainty, a sensitivity analysis was carried out,
410 focusing on the most easily quantified uncertainties, namely the transport algorithm and the
411 modeling of the source term. There are three possible choices for the electron transport
412 algorithm in MNCP6 [14]: ITS, the one used for the above simulations; “mncp”, the default
413 option in MCNPX; and “Landau”, the default option in MCNP6. To quantify the uncertainty
414 from the choice of algorithm, the simulations with the $^{90}\text{Sr}/^{90}\text{Y}$ source were repeated using the
415 mncp and then the Landau transport algorithms (data not shown). For the latter, the energy
416 cut-off for electron and photon transport had to be decreased to 20 eV instead of the 1 keV
417 defined by default in MCNP6 [14]. Using the mncp algorithm reduces the relative deviation
418 between simulated and experimental spectra by a few percentage points in the interval 0.9–2
419 MeV and by a few tens of percentage points in the 1.2–2 MeV region. Using the Landau
420 algorithm did not reduce the relative deviation from the experimental data, except for energies
421 below 0.7 MeV, and the lower electron transport cut-off significantly increased the simulation
422 times.

423 The second source of uncertainty investigated was the modeling of the source term,
424 assumed to be uniformly distributed. To investigate the effect of a non-uniform source
425 distribution, simulations were carried out with $^{90}\text{Sr}/^{90}\text{Y}$ sources arranged in a regular pattern
426 of circular deposits on the substrate (data not shown). The results obtained differed little from

427 those with a uniformly distribute source, showing that this assumption has little influence on
428 the simulated spectra and cannot explain the observed deviations from the experimental data.

429 The conclusion of this sensitivity analysis is that the observed differences between the
430 observed and simulated spectra cannot be explained by a single source of uncertainty but are
431 likely due to a combination of the multiple identified sources of uncertainty. The MCNP
432 model of the detector can be considered qualified based on these results, with a maximum
433 relative deviation of 30% from the experimental data. This upper estimate was used thereafter
434 as the conservative relative uncertainty of the Monte Carlo simulations.

435 *3.3. Calibration coefficients*

436 The method adopted here to estimate the ^{90}Sr activity of contaminated soils rests on
437 Monte Carlo simulations of different measurement setups to establish calibration coefficients
438 (C in Eq. (2)) [2,3].

439 *3.3.1. ^{90}Sr -contaminated concrete*

440 The method, which was initially developed to study ^{90}Sr -contaminated soils, was first
441 applied here to concrete structures contaminated with ^{90}Sr . Sensitivity analyses were carried
442 out to determine the variation of the calibration coefficient as a function of the diameter of a
443 surface source and its depth in the substrate. As a first approach, the horizontal and vertical
444 distributions of the contaminant were both considered uniform.

445 *3.3.1.1. Sensitivity analysis in terms of the surface area of the contamination*

446 The calibration coefficient in Eq. (2) was calculated for surface sources ranging from
447 6–156 mm in diameter in 10 mm steps. The source was modeled as a cylinder with a height of
448 0.1 mm made of air with a uniformly distributed activity of $1 \text{ Bq}\cdot\text{cm}^{-2}$. The concrete was
449 assumed to have a standard composition [26] and a density of $2.3 \text{ g}\cdot\text{cm}^{-3}$. Calculations were

450 performed with the detector either in direct contact with the source, to optimize the β -
451 detection efficiency, or at distance of 4 mm (corresponding to the thickness of the aluminum
452 cover), to evaluate the effect of the detector's positioning with respect to the ground.

453 Fig. 11 shows how the calibration coefficient for the energy range 0.9–2.0 MeV varies
454 as a function of the diameter of the source in these two arrangements. When the detector is in
455 direct contact with the ground, the calibration coefficient plateaus at diameters above 76 mm.
456 This is because the electrons from the outer parts of the source are stopped by the aluminum
457 casing when the source becomes wider than the scintillator. The detection efficiency is lower
458 when the detector is placed 4 mm from the concrete floor. The difference is about 20% at a
459 source diameter of 76 mm. However, the calibration coefficient levels out more gradually,
460 tending towards the value in direct contact for sources wider than 130 mm. Similar data were
461 obtained for the other energy interval of interest (1.2–2.0 MeV, data not shown). These results
462 indicate that measurements in direct contact with the contaminated surface are more efficient
463 than at a distance when the contaminated area is no larger in diameter than the scintillator (76
464 mm). The calibration coefficients calculated in this configuration for a uniform surface
465 contamination are listed in Table 3.

466 *3.3.1.2. Sensitivity analysis in terms of the depth of the source*

467 The same approach was used to evaluate the effect of the depth of the source. A
468 cylinder of air 76 mm in diameter with a uniformly distributed volume activity of $1 \text{ Bq}\cdot\text{cm}^{-3}$
469 was placed at a depth of 0.5–6 mm in the concrete floor. The concrete was modeled as before
470 and the detector was placed in direct contact with the floor.

471 The variation of the calibration coefficient with the depth of the source is shown in
472 Fig. 12 for both energy intervals. The profiles are similar, an increase followed by a plateau,
473 but the maximum occurs at a depth of about 3 mm for the energy interval 0.9–2.0 MeV versus
474 about 2 mm for electrons in the energy range 1.2–2.0 MeV. This is expected because the

475 electron spectrum shifts to lower energies as the depth of the source increases and the
476 electrons lose more energy passing through the concrete. This means that electrons with
477 energies greater than 0.9 MeV do not contribute to the detector's response in the 0.9–2.0 MeV
478 interval if the source is covered by more than 3 mm of concrete. For electrons with an energy
479 greater than 1.2 MeV (the 1.2–2.0 MeV interval), the corresponding threshold is 2 mm. The
480 calibration coefficients determined under these conditions are listed in Table 3.

481 If the contamination is uniformly distributed down to a depth of more than a few
482 millimeters, the mass activity can be deduced from the measurements obtained for the first
483 few millimeters of contaminated concrete using the calibration coefficients in Table 3. The
484 difficulty then lies in defining the minimum depth at which this conversion is valid. For
485 surface or volume contaminations in the first few millimeters of concrete indeed, the mass
486 activities obtained from the calibration coefficients will substantially underestimate the true
487 values. These uncertainties are specific to substrates in which ^{90}Sr is liable to migrate (soils,
488 concrete, etc.). For surface-contaminated industrial waste, the use of surface calibration
489 coefficients provide close overestimates of the true activity.

490 *3.3.1.3. Estimated setup-specific detection limits*

491 The detection limit corresponds to the situation in which the signal from contaminated
492 concrete is the same as the background signal from non-contaminated concrete [33].

493 The background β - spectrum from radionuclides naturally present in concrete (^{40}K and
494 the daughter nuclei of ^{238}U and ^{232}Th) was obtained by taking the difference of the spectra
495 measured over 16 h with and without the aluminum cover in direct contact with a non-
496 contaminated concrete floor (Fig. 13). The count rates were then calculated in the two energy
497 intervals of interest, 0.9–1.2 and 1.2–2.0 MeV.

498 The detection limits of the system were calculated for different on-site measurement
499 times (10, 20, 30 and 60 min for each spectrum with and without the aluminum cover) and

500 then converted to mass activities using the calibration coefficient (Table 3) calculated for
501 concrete with a uniform contamination of ^{90}Sr down to a depth of 3 mm. The values obtained
502 are listed in Table 4.

503 3.3.2. ^{90}Sr -contaminated Fontainebleau sand

504 In view of applying this method to various types of soil encountered in
505 decommissioning sites, measurements were performed on samples of Fontainebleau sand
506 from the CEA (French Alternative Energies and Atomic Energy Commission) site at
507 Fontenay-aux-Roses. Furthermore, to assess the possibility of analyzing multiple samples
508 using an autosampler, measurements were also taken on sand samples conditioned in
509 standardized 500 cm³ polyethylene (SG500) containers, which are mainly used to analyze
510 liquid samples by gamma spectrometry.

511 3.3.2.1. Direct measurements on bare sand

512 The variation of the calibration coefficient in Eq. (2) with the depth of a volume
513 source was calculated as described above (section 3.3.1.2) with the concrete replaced by
514 Fontainebleau sand. As before, the detector was placed in direct contact with the substrate to
515 maximize the detection efficiency. The composition of the sand was assumed to be 100%
516 SiO₂, with a density of 1.5 g·cm⁻³ [34].

517 The depth profiles obtained for the two energy ranges of interest are shown in Fig. 14.
518 The trends are the same as those observed for contaminated concrete (cf. Fig. 12). However,
519 since sand is less dense than concrete, the electrons travel further and the maximum count rate
520 occurs at a depth of 4.5 mm (rather than 3 mm for concrete). The corresponding calibration
521 coefficients are listed in Table 3. The calibration coefficients for surface contamination are
522 similar to those obtained for concrete (Table 3). The nature of the substrate therefore has little
523 effect on electron transport, particularly in terms of backscattering from the surface.

524 3.3.2.2. Measurements on sand in SG500 containers

525 The aim of these measurements was to evaluate the sensitivity loss due to the
526 polyethylene container. Fig. 15 shows the setup used for the Monte Carlo simulations, with
527 the SG500 container placed centrally and directly against the aluminum frame of the detector.
528 The container was modeled as realistically as possible, with two separate compartments and a
529 wall density ($0.939 \text{ g}\cdot\text{cm}^{-3}$) corresponding to the mean of the reference values for low- and
530 high-density polyethylene (respectively $0.917\text{--}0.930 \text{ g}\cdot\text{cm}^{-3}$ [35] and $0.944\text{--}0.965 \text{ g}\cdot\text{cm}^{-3}$
531 [36]). The decrease in signal intensity due to the container was assessed by simulating the
532 $^{90}\text{Sr}/^{90}\text{Y}$ spectra of uniformly contaminated Fontainebleau sand in a SG500-type container with
533 either air or polyethylene walls (Fig. 16). Compared with direct measurements on bare sand,
534 the count rates with an SG500 container are approximately 8 and 16 times lower in the 0.9–
535 2.0 MeV and 1.2–2.0 MeV regions, respectively (Table 3). One possibility to increase the
536 detection efficiency if ^{137}Cs is present but not ^{60}Co would be to extend the measurement range
537 to lower energies (down to 0.3 instead of 0.9 MeV). In this configuration indeed, the beta
538 spectrum of ^{137}Cs is shifted to energies below 0.3 MeV. Samples co-contaminated with ^{60}Co
539 have not been studied so far.

540 4. Results for samples of contaminated Fontainebleau sand

541 To validate this method for quantifying ^{90}Sr activity, measurements were performed
542 on 10 samples of contaminated sand from the CEA decommissioning site at Fontenay-aux-
543 Roses. Previous laboratory-based radiochemical analyzes revealed that the main radionuclides
544 in the samples were ^{238}Pu , ^{237}Np , ^{233}Pa , ^{90}Sr , with ^{137}Cs and $^{239}\text{Pu}+^{240}\text{Pu}$ also present in some
545 samples (Table 5). The main reasons for choosing these samples were that the main emitter is
546 ^{90}Sr , that the mass activity of ^{90}Sr varies from a few to a few hundred $\text{Bq}\cdot\text{g}^{-1}$, and that some
547 of samples contain ^{137}Cs . Note that while some of the daughter nuclei in the decay chain of

548 ^{239}Pu , ^{238}Pu , ^{237}Np and ^{233}Pa emit β - particles with energies similar to those of ^{90}Y , these
549 contributions were considered negligible because the activity of these radionuclides in the
550 samples is two decades much lower than that of ^{90}Sr .

551 Two measurement setups were used, as described in section 3.3.2 and shown in Fig.
552 17; the detector was placed either in direct contact with the sand (for all ten samples), or with
553 the sample in a SG500 container placed on top of the detector (for four samples). Since the
554 samples had been stored in SG500 containers, the direct measurements involved spreading
555 some of their contents in a larger diameter container. To avoid contaminating the detector, a
556 layer of Parafilm M[®] (Bemis, Neenah, WI) was placed between the sand and the detector and
557 between the detector and the SG500 container. Parafilm M is a blend of paraffin and
558 polyolefin on a sheet of glassine paper. The manufacturer lists its thickness as 127 μm and its
559 density as $0.922 \text{ g}\cdot\text{cm}^{-3}$.

560 Two acquisition times (for the primary and auxiliary spectra) to obtain good counting
561 statistics in the region of interest (0.9–2.0 MeV) varied from 5 min for the most highly
562 contaminated samples (^{90}Sr activities of hundreds of $\text{Bq}\cdot\text{g}^{-1}$) to 20 min for the least
563 contaminated ones (^{90}Sr activities of just a few $\text{Bq}\cdot\text{g}^{-1}$). The auxiliary measurements showed
564 that the gamma emissions from the samples in the region of interest were very weak (Fig. 18).

565 Assuming negligible natural radioactivity, the ^{90}Sr activity of the studied samples can
566 be estimated by multiplying the count rate measured in the energy interval of interest,
567 $N_e(\Delta E_e)$, by the calibration coefficient determined using Monte Carlo simulations:

$$A_{\text{Sr}} = N_e(\Delta E_e) \times C \quad (5)$$

568 The calibration coefficients for the two measurement setups were determined (section
569 3.3.2) for a unit activity of ^{90}Sr uniformly distributed in pure SiO_2 with a density of 1.5
570 $\text{g}\cdot\text{cm}^{-3}$. The assumption that the ^{90}Sr is uniformly distributed in the samples is reasonable

571 given the small size of the samples with respect to the extent of the contamination and their
572 granular nature, which favors mixing. Nonetheless, to minimize uncertainties in the measured
573 activities due to uncertainties in the calibration coefficients, another set of more precise
574 simulations was performed, including the layer of Parafilm M and using the measured density
575 of the sand ($1.4 \pm 0.2 \text{ g}\cdot\text{cm}^{-3}$) instead of the previous estimate ($1.5 \text{ g}\cdot\text{cm}^{-3}$). The additional
576 layer between the detector and the sample shifts the beta spectrum to lower energies and thus
577 reduces the calibration coefficients in the energy regions of interest (Table 3). For the
578 measurements on bare sand, since the calibration coefficient varies with depth down to 4.5
579 mm (Fig. 14), a sufficiently thick layer of each sample ($> 4.5 \text{ mm}$) was poured to ensure the
580 validity of the maximum calibration coefficient. The calibration coefficients obtained for the
581 two measurement setups are presented in Table 3.

582 The uncertainty in the calibration coefficient was estimated by combining the
583 individual estimates of the uncertainties in the parameters in the model. For the measurements
584 on bare sand, the main sources of uncertainty are the uncertainties in the calibration
585 coefficient ($\pm 30\%$), in the densities of the sand ($\pm 0.2 \text{ g}\cdot\text{cm}^{-3}$) and of Parafilm M ($\pm 10\%$),
586 and in the position of the source with respect to the detector ($\pm 0.5 \text{ mm}$). For the
587 measurements on sand in SG500 containers, there is an additional contribution from the
588 density of the polyethylene walls, which falls somewhere in the range $0.917\text{--}0.965 \text{ g}\cdot\text{cm}^{-3}$.
589 These uncertainties (type B) were assumed to be independent and the overall relative
590 uncertainty in the calibration coefficient was obtained by combining the relative values
591 quadratically. The uncertainty range was then doubled to obtain the 95% confidence interval.

592 The ^{90}Sr activities calculated using Eq. (5) are compared with the values measured by
593 destructive radiochemical analysis in Table 6, corrected for the decrease in ^{90}Sr activity that
594 occurred between the laboratory measurements (in 2013 and 2015) and those with the
595 portable β - spectrometer (in 2016). The values obtained by beta spectrometry on bare sand

596 tend to be lower than the activities measured by radiochemical analysis, with a maximum
597 relative deviation of 52.9%, while the measurements on sand in SG500 containers tend to be
598 overestimates, with a maximum relative deviation of 57% (Table 6). These deviations can be
599 attributed to uncertainties in the geometry of two models and in the Monte Carlo simulations.
600 The activities measured by beta spectrometry are nonetheless within the estimated uncertainty
601 range of the values obtained by destructive analysis. These results are therefore promising for
602 the use of the portable β - spectrometer on decommissioning sites as a simple, fast and at least
603 semi-quantitative method to characterize difficult to detect radionuclides such as ^{90}Sr .

604 **5. Conclusion**

605 Portable β - spectrometry for the non-destructive detection and quantification of ^{90}Sr is
606 a relatively new research topic at the CEA, motivated by the need for on-site characterization
607 methods for pure β -- emitters. These radionuclides are indeed difficult to detect using
608 classical non-destructive techniques such as gamma spectrometry and are usually quantified
609 by liquid scintillation in the laboratory. The overall aim of the work presented here is to
610 optimize the decommissioning process, namely to improve radioactive waste management
611 and radioprotection measures by providing better information more rapidly on sources, and to
612 minimize costs by optimizing sampling schemes in terms of the number and location of
613 samples analyzed.

614 Building on a method developed to study soils contaminated by the Chernobyl
615 accident, we updated, adapted and qualified the different components using Monte Carlo
616 simulations before using the system to measure the ^{90}Sr activity of sand samples from a CEA
617 decommissioning site. The results are encouraging and indicate that this simple approach can
618 in the future be used on site to obtain semi-quantitative information on radionuclides that
619 typically require laboratory analyses to detect.

620 The aim of non-destructive measurement techniques is to complement rather than
621 replace destructive analyses, which will always provide the most accurate quantitative data
622 for decommissioning operations. However, the value of destructive measurements depends on
623 the number and representativity of the samples extracted from the site. The advantage of
624 portable non-destructive techniques is that they can be used directly, on site and on unaltered
625 structures, to identify the spatial distribution (uniform or with hot spots) of the contamination
626 and thus optimize the sampling scheme and the representativity of the samples for laboratory
627 analysis. On site measurements thus improve the sampling and the overall efficiency of the
628 analytical process.

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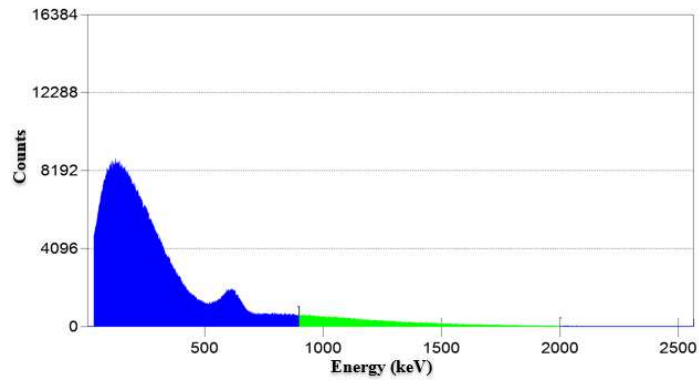
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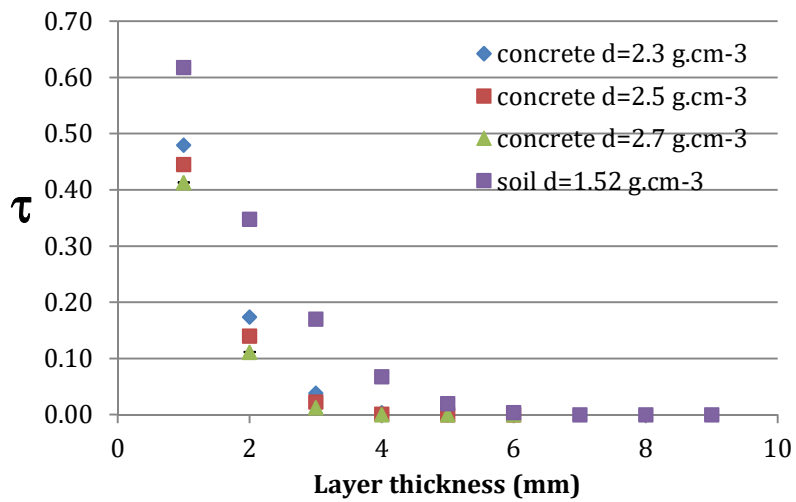
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1 Figures



3 **Fig. 1.** Example of the energy spectrum of β^- particles emitted by a ^{90}Sr and ^{137}Cs source
4 after subtraction of the contribution from gamma photons.

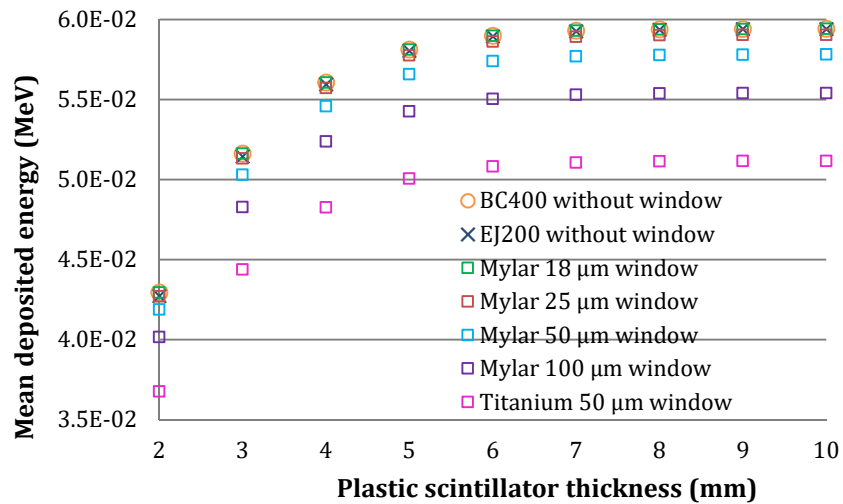


7

8 **Fig. 2.** Proportion τ of $^{90}\text{Sr}/^{90}\text{Y}$ electrons traveling through layers of concrete and soil with
9 different densities as a function of the thickness of the layer.

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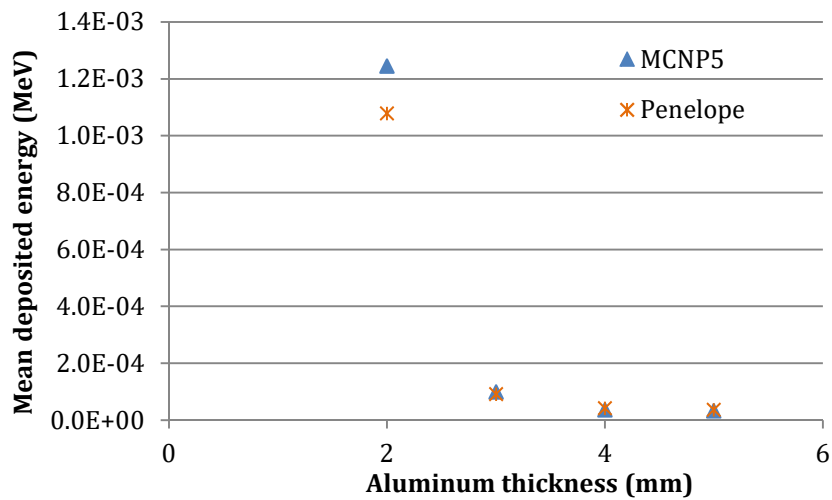


12

13 **Fig. 3.** Mean energy deposited by $^{90}\text{Sr}/^{90}\text{Y}$ electrons as a function of the thickness of the
 14 scintillator for two scintillating plastics, BC400 and EJ200 without window and when the
 15 EJ200 scintillator is covered by a layer of Mylar film (four different thicknesses) or a titanium
 16 sheet.

17

18

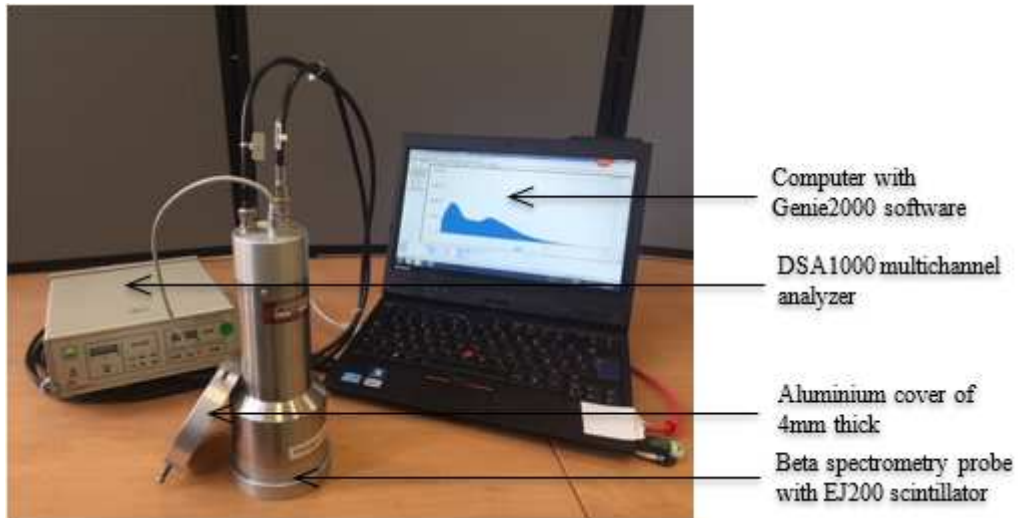


19

20 **Fig. 4.** Mean energy deposited by $^{90}\text{Sr}/^{90}\text{Y}$ electrons in the scintillator as a function of the
 21 thickness of the aluminum cover: comparison of the values obtained by MCNP5 and
 22 PENELOPE simulations.

23

24

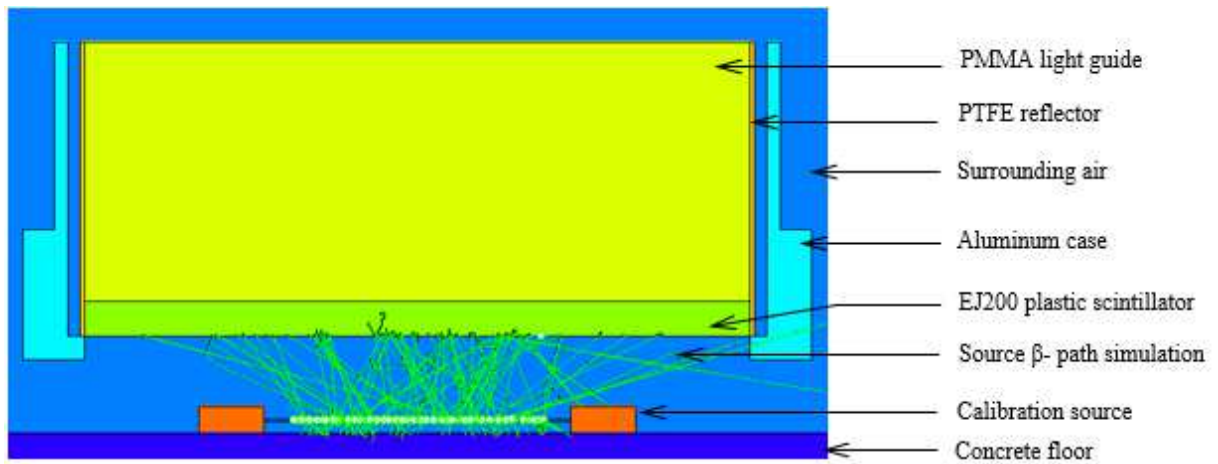


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26 **Fig. 5.** Photographs of the device for beta spectrometry.

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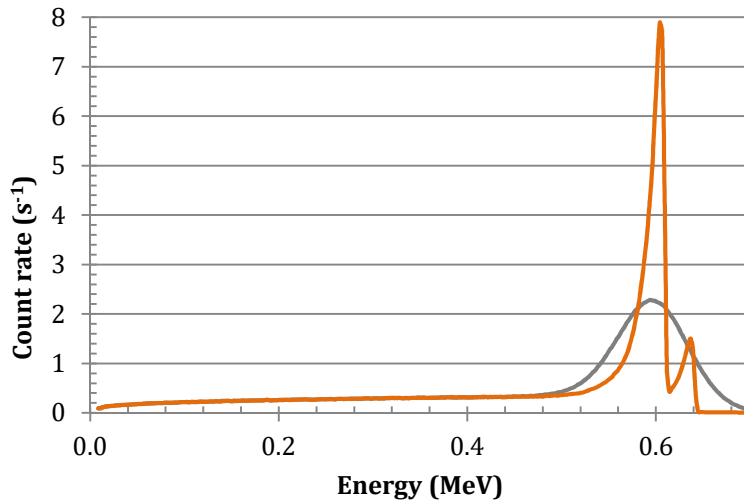
29

30 **Fig. 6.** Diagram of the setup and simulated electron paths in MCNP6 with a ^{137}Cs calibration

31 source.

32

33

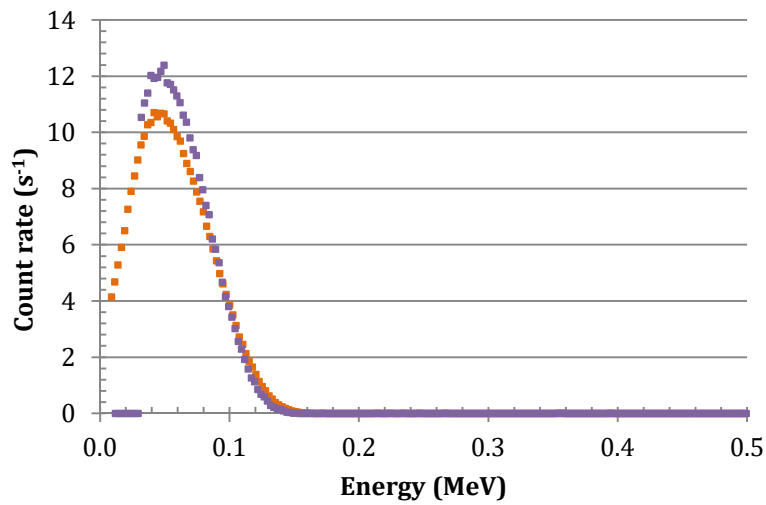


34

35 **Fig. 7.** Comparison of the simulated energy distributions in the scintillator arising from the
 36 internal conversion electrons emitted by ^{137}Cs at 624 and 656 keV accounting (grey line) or
 37 not (orange line) for the finite resolution of the detector.

38

39

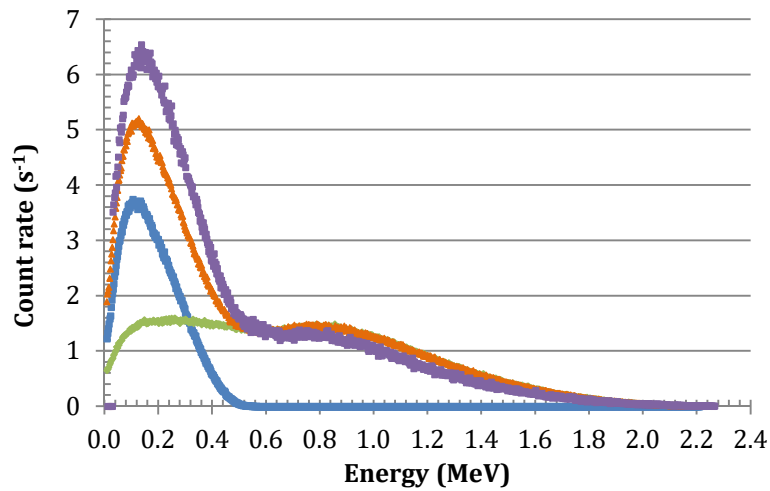


40

41 **Fig. 8.** Comparison of the simulated (in orange) and experimental (in purple) β^- spectra of the
 42 ^{14}C calibration source. Electron transport was simulated using the integrated Tiger series
 43 (ITS) algorithm.

44

45

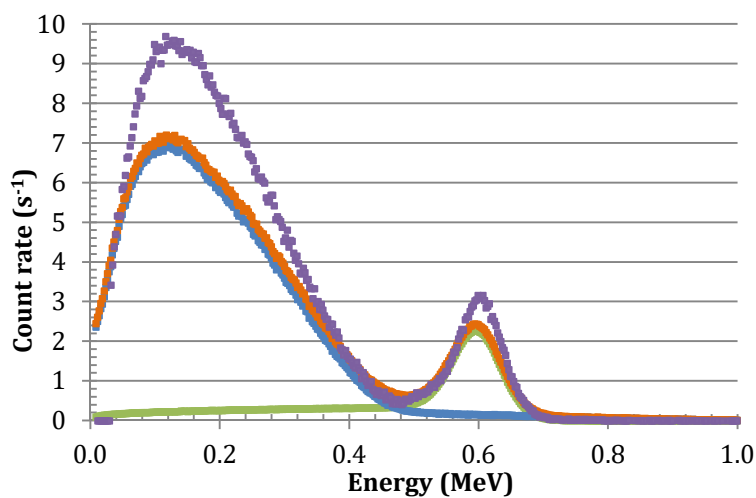


46

47 **Fig. 9.** Comparison of the simulated (in blue, the β^- spectrum of ^{90}Sr , in green, the β^-
 48 spectrum of ^{90}Y , in orange the total spectrum) and experimental (in purple) β^- spectra of the
 49 $^{90}\text{Sr}/^{90}\text{Y}$ calibration source.

50

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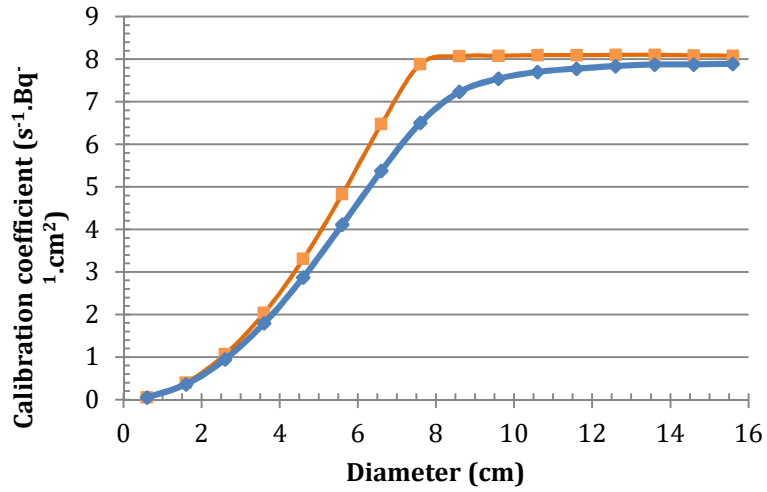


52

53 **Fig. 10.** Comparison of the simulated (in blue, the β^- spectrum of ^{137}Cs , in green, the Internal
 54 Conversion spectrum, in orange the total spectrum) and experimental (in purple) β^- spectra of
 55 the ^{137}Cs calibration source. IC, internal conversion.

56

57

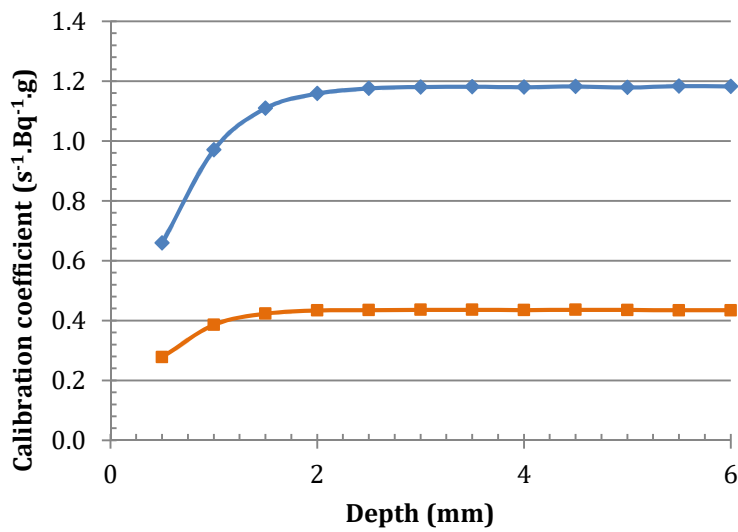


58

59 **Fig. 11.** Calibration coefficient in Eq. (2) for the energy interval 0.9–2.0 MeV as a function of
 60 the diameter of a surface source of uniformly distributed activity in a concrete floor, with the
 61 detector placed either in direct contact with the floor (orange line) or 4 mm above (blue line).

62

63

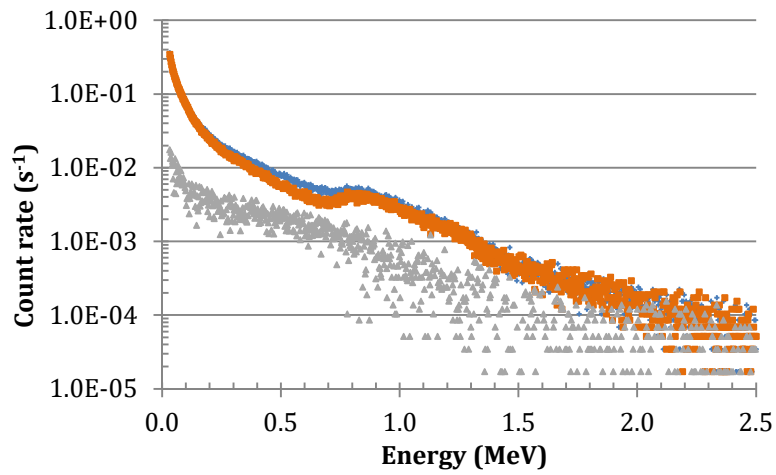


64

65 **Fig. 12.** Calibration coefficient in Eq. (2) for the two energy levels of interest (0.9–2.0 MeV
 66 in blue, 1.2–2.0 MeV in orange) as a function of the depth of a uniformly distributed volume
 67 source in concrete.

68

69

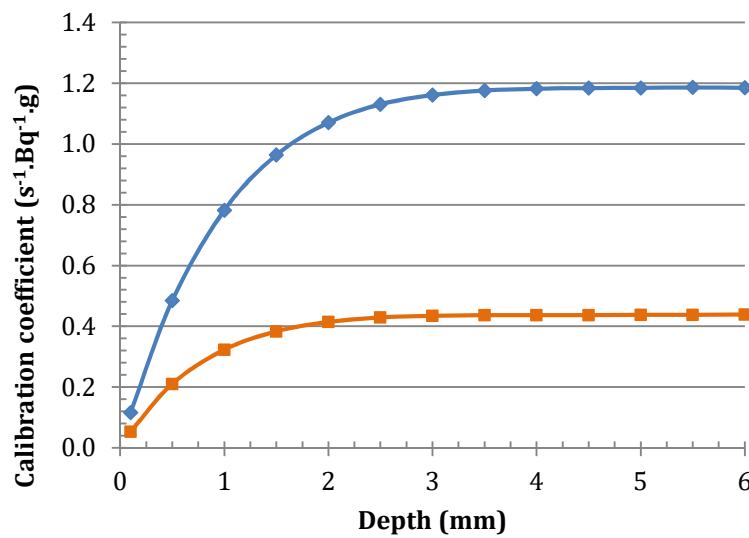


70

71 **Fig. 13.** Background spectra measured on non-contaminated concrete without the aluminum
 72 cover (in blue) and with (in orange), and the difference spectrum (in gray).

73

74

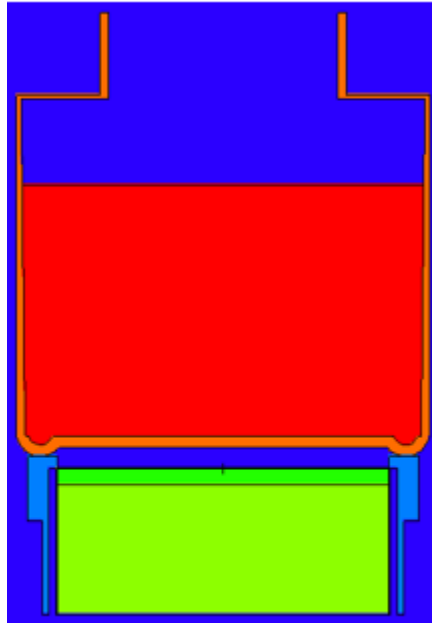


75

76 **Fig. 14.** Calibration coefficient in Eq. (2) for the two energy levels of interest (0.9–2.0 MeV
 77 in blue, 1.2–2.0 MeV in orange) as a function of the depth of a uniformly distributed volume
 78 source in Fontainebleau sand.

79

80

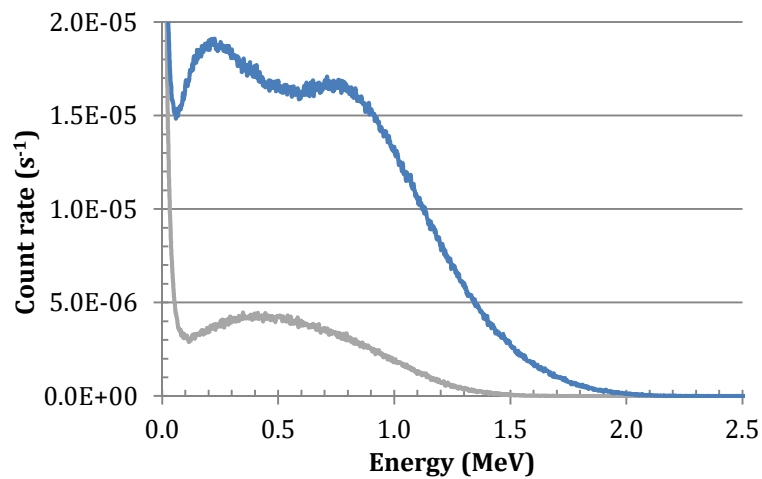


81

82 **Fig. 15.** Cross-section of the setup simulated in MCNP6 for measurements of ^{90}Sr activity in
 83 uniformly contaminated sand inside a standardized 500 cm^3 SG500 container.

84

85



86

87 **Fig. 16.** Comparison of simulated $^{90}\text{Sr}/^{90}\text{Y}$ β^- spectra of uniformly contaminated sand in a
 88 container with walls made of air (in blue) or polyethylene (in gray).

89

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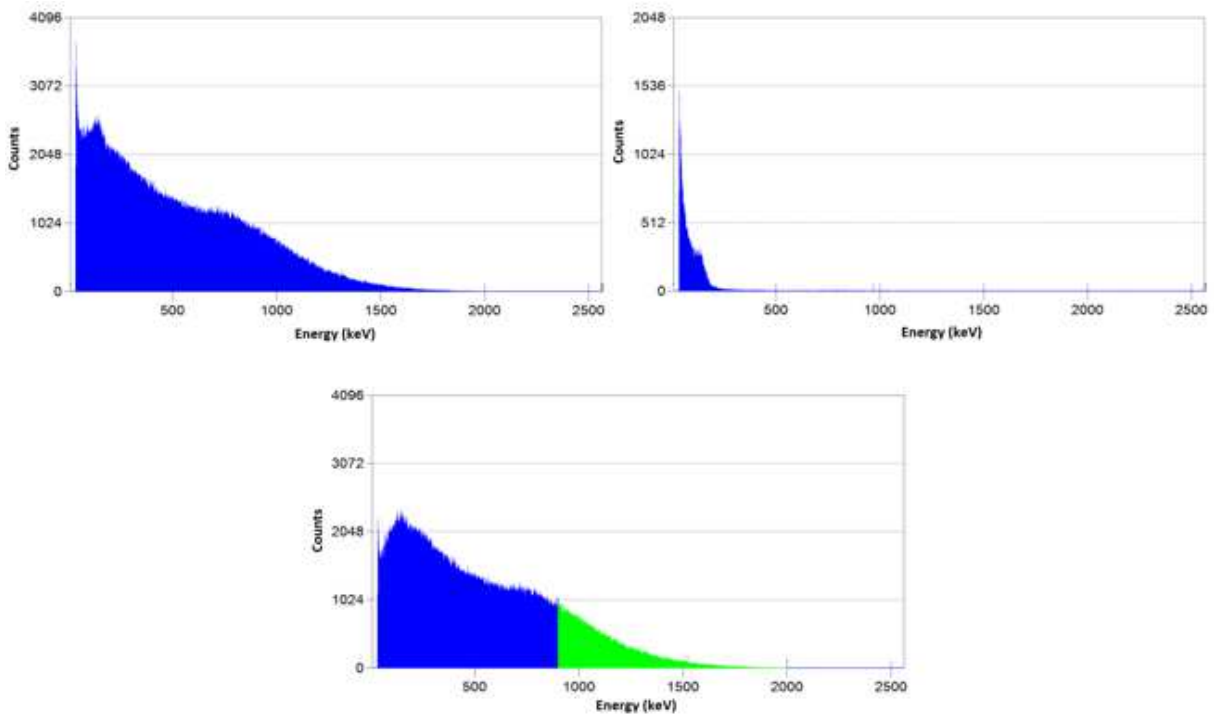
92

93 **Fig. 17.** Photographs of the beta spectrometer for measurements on sand samples in a

94 standardized container (bottom left) and in direct contact with the sand (bottom right).

95

96



97

98 **Fig. 18.** Examples of the β spectra acquired in 10 min by the detector in direct contact with

99 the sample 3 (Table 6): the main spectrum with beta and gamma components (upper left

100 panel), the auxiliary spectrum with only gamma component (upper right) and the final β
101 spectrum (bottom panel) after subtraction of the latter from the former.

102

1 Tables

2 Table 1

3 Characteristics of the calibration sources used to validate the numerical model of the β
4 spectrometer

Radionuclide	^{14}C	^{36}Cl	^{90}Sr	^{90}Y	^{204}Tl	^{137}Cs
Atomic number	6	17	38	39	81	55
Emission type	pure β^-	pure β (98.1% β^- & 1.9% β^+ or EC)	pure β^-	pure β^-	pure β (97.08% β^- & 2.92% β^+)	β^- & γ
Average energy (keV)	49.47	246.5	195.9	933	237.92	187.54
Maximum energy (keV)	156.47	709.3	546.2	2281.5	763.41	1175.1
Half life (years)	5.7×10^3	3.01×10^5	28.8	2.66 days	3.79	30.04
Activity	^{14}C	^{36}Cl	$^{90}\text{Sr} + ^{90}\text{Y}$		^{204}Tl	^{137}Cs
$A(t_0)$ (Bq)	1641	3269	3254		2839	3100
t_0	3 Mar 2015	1 Jul 2014	16 Jul 2014		4 Mar 2015	2 Mar 2015
$A(t_1)$ (Bq)	1641	3269	3181		2685	3078
t_1			23 Jun 2015			

5 EC, electron capture.

6 7 Table 2

8 Comparison of the measured and simulated β spectra of the calibration sources

Radionuclide	Energy range (keV)	Average count rate (measured, {M})	Average count rate (simulated, {S})	Relative deviation ({S} - {M}) / {M}
^{14}C	31–156.5 ^a	2.82×10^2	2.57×10^2	-9%
^{36}Cl	31–709.3 ^a	1.39×10^3	1.09×10^3	-21%

^{204}Tl	31–763.4 ^a	1.08×10^3	7.71×10^2	–29%
^{137}Cs	508–700 ^b	1.17×10^2	1.02×10^2	–13%
$^{90}\text{Sr} / ^{90}\text{Y}$	900–2000	2.03×10^2	2.55×10^2	+26%
$^{90}\text{Sr} / ^{90}\text{Y}$	1200–2000	8.78×10^1	1.17×10^2	33%

9 ^aTotal spectrum
10 ^bInternal conversion peak
11

12 **Table 3**

13 Calibration coefficient for different measurement setups determined by Monte Carlo
14 simulations

Substrate ^a	Depth (mm)	Surface (cm ²)	Volume (cm ³)	Energy range (MeV)	C (s ⁻¹ ·Bq ⁻¹ ·cm ²)	C (s ⁻¹ ·Bq ⁻¹ ·cm ³)	C (s ⁻¹ ·Bq ⁻¹ ·g)
Concrete ^a	0	45.4	–	0.9–2.0	7.89	–	–
Concrete ^a	0	45.4	–	1.2–2.0	3.69	–	–
Sand ^a	0	45.4	–	0.9–2.0	7.95	–	–
Sand ^a	0	45.4	–	1.2–2.0	3.72	–	–
Concrete ^a	3	–	13.6	0.9–2.0	–	0.514	1.18
Concrete ^a	3	–	13.6	1.2–2.0	–	0.190	0.436
Sand ^a	4.5	–	20.4	0.9–2.0	–	0.790	1.19
Sand ^a	4.5	–	20.4	1.2–2.0	–	0.292	0.438
Sand ^b	58.3	–	388.16	0.9–2.0	–	–	1.177
Sand ^b	58.3	–	388.16	1.2–2.0	–	–	0.438
Sand ^c	58.3	–	388.16	0.9–2.0	–	–	0.143
Sand ^c	58.3	–	388.16	1.2–2.0	–	–	0.027
Sand ^c	58.3	–	388.16	0.3–2.0	–	–	0.661
Sand ^d	10	–	113.1	0.9–2.0	–	–	1.11 ± 60%

Sand ^e	58.3	–	388.16	0.3–2.0	–	–	0.61 ± 60%
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15 ^aPlaced in direct contact with the detector.

16 ^bSand in 500cm³ container with air walls

17 ^cSand in 500cm³ container with polyethylene walls

18 ^dBare sand, detector covered by Parafilm M

19 ^eSand in 500cm³ container with polyethylene walls, detector covered by Parafilm M

20

21 **Table 4**

22 Detection limits calculated for different measurement times on non-contaminated cement

Energy range (MeV)	Count rate (s ⁻¹)	C (s ⁻¹ ·Bq ⁻¹ ·g)	DL, 2 × 10 min (Bq·g ⁻¹)	DL, 2 × 20 min (Bq·g ⁻¹)	DL, 2 × 30 min (Bq·g ⁻¹)	DL, 2 × 60 min (Bq·g ⁻¹)
0.9–2.0	7.13 × 10 ⁻²	1.18	3.70 × 10 ⁻²	2.60 × 10 ⁻²	2.10 × 10 ⁻²	1.50 × 10 ⁻²
1.2–2.0	2.29 × 10 ⁻²	0.436	5.60 × 10 ⁻²	4.00 × 10 ⁻²	3.30 × 10 ⁻²	2.30 × 10 ⁻²

23 C, calibration constant; DL, detection limit.

24

25 **Table 5**

26 Mass activities of samples of contaminated Fontainebleau sand as determined by

27 radiochemical analysis

Sample	Year	²³⁸ Pu+ ²⁴⁰ Pu (Bq·g ⁻¹)	²³⁸ Pu (Bq·g ⁻¹)	²³⁷ Np (Bq·g ⁻¹)	²³³ Pa (Bq·g ⁻¹)	¹³⁷ Cs (Bq·g ⁻¹)	⁹⁰ Sr (Bq·g ⁻¹)
1	2013	–	1.70 ± 0.73	2.00 ± 0.27	1.80 ± 0.21	–	27.0 ± 1.5
2	2013	–	21.0 ± 9.0	11.0 ± 0.9	11.00 ± 0.66	51.0 ± 3.0	190 ± 100
3	2015	–	4.5 ± 1.1	48.0 ± 5.0	43.0 ± 4.7	–	260 ± 140
4	2015	–	3.00 ± 0.75	67.0 ± 7.1	67.0 ± 7.7	–	560 ± 310
5	2013	–	32 ± 14	41.0 ± 4.4	43.0 ± 4.9	–	370 ± 200
6	2013	–	0.260 ± 0.024	0.099 ± 0.030	0.087 ± 0.015	–	76.0 ± 7.8
7	2013	8.3 ± 3.9 × 10 ³	1.50 ± 0.15	5.5 ± 1.4	5.50 ± 0.35	–	210 ± 22
8	2013	6.3 ± 3.6 × 10 ³	1.40 ± 0.12	5.1 ± 1.3	4.70 ± 0.30	170 ± 17	170.0 ± 8.6
9	2013	–	0.230 ± 0.023	0.90 ± 0.23	1.00 ± 0.07	–	9.40 ± 0.93
10	2013	–	0.036 ± 0.007	0.056 ± 0.018	0.071 ± 0.008	–	14.0 ± 0.14

28 The data are presented as A ± 2σ (activity ± twice the standard deviation).

29

30 **Table 6**31 Comparison of the ^{90}Sr mass activity values obtained by radiochemical analysis and non-

32 destructive beta spectrometry for samples of contaminated Fontainebleau sand

Sample	Bare sand {A} ($\text{Bq}\cdot\text{g}^{-1}$)	Sand in SG500 {B} ($\text{Bq}\cdot\text{g}^{-1}$)	RA {C} ($\text{Bq}\cdot\text{g}^{-1}$)	$(\{C\}-\{A\})/\{C\}$	$(\{C\}-\{B\})/\{C\}$
1	25 ± 15	–	25 ± 14	0.4%	–
2	83 ± 50	–	180 ± 97	52.9%	–
3	16 ± 9.9	–	250 ± 140	35.0%	–
4	33 ± 20	70 ± 42	550 ± 300	39.5%	–28%
5	43 ± 26	–	340 ± 190	–23.9%	–
6	42 ± 25	11.0 ± 6.6	7.00 ± 0.73	40.5%	–57%
7	13.0 ± 7.6	–	200 ± 21	36.9%	–
8	9.5 ± 5.7	21 ± 13	160 ± 16	39.5%	–35%
9	6.2 ± 3.7	–	8.80 ± 0.86	29.0%	–
10	0.8 ± 0.5	1.9 ± 1.2	1.30 ± 0.13	37.4%	–51%

33 The data are presented as $A \pm 2\sigma$ (specific activity \pm twice the standard deviation).34 SG500, a standardized 500cm³ polyethylene container; RA, radiochemical analysis.