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1 **From Bq cm⁻³ to Bq cm⁻² (and conversely) - Part 1: a useful**

2 **conversion for autoradiography**

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9 **Abstract**

10 Autoradiography technique generally provides an image in cps cm⁻², the conversion in Bq g⁻¹
11 remaining a challenge. The Geant4 tool has been used to establish a conversion procedure based
12 on the estimation of a key parameter, the Emission Fraction F_E . The conversion method was
13 applied on three samples containing ³H, ¹⁴C and ²³⁸U respectively, allowing then to assess the
14 detection efficiency of a digital radioimager, BeaQuant™. Particular attention was paid to
15 matrix effect of contaminated material that could affect the detection efficiency. This article is
16 coupled with a companion paper, providing support to apply the conversion method to
17 decommissioning investigations.

18 **Keywords**

19 autoradiography, surface/volumetric activities, detection efficiency, Geant4, emission fraction.
20
21

Introduction

23 Autoradiography is a two-dimensional image of the radioactive emissions from a solid material.
24 It appears to be one of the most suitable technique to detect, quantify and map radioactive
25 particle emissions, with the significant advantage to be a non-destructive method [1]. The usual
26 methods use films (silver halide emulsions or phosphor screens) which are just blackened under
27 radiations: standard specimens are needed to quantify the blackening level. In opposition to
28 films, the radioimagers, a new technology of Digital Autoradiography (DA), allow an
29 acquisition in real time giving an individual particle counting (cps cm⁻²). Developed first in
30 biological, medical and pharmaceutical fields to track molecules labeled with radioactive
31 tracers [1], autoradiography is now a valuable technique to study solid materials such as rock
32 samples in geological field, building materials or personal protective equipment in a
33 dismantling context. DA is of primary interest to study properties of rock targeted for the final
34 disposal of spent nuclear fuel, such as the sorption and diffusion of radioactive elements into
35 rocks [2-4]. The connected porosity of rocks which controls the diffusion process [5], can also
36 be fruitfully investigated with DA after impregnation with the ¹⁴C-PolyMethylMetAcrylate
37 (¹⁴C-PMMA), a radioactive resin. The radioactive tracer is mapped on decimeter scale samples
38 [6-7]. In the front-end activities of the nuclear fuel cycle, DA technology is being used now to
39 locate precisely the uranium present in a given ore, as well as the remaining radioactive
40 elements in uranium mill tailings [8-9].

41 The investigation of radionuclide contaminations under dismantling processes has also been
42 recently developed using autoradiography. A concrete floor of an old nuclear laboratory has
43 indeed been studied with phosphor screens, to map and quantify the radioactive contaminations
44 [10]. A full-scale visualization of the contaminations was obtained, and half of the estimated
45 activities measured by autoradiography was consistent with the liquid scintillation counting
46 measurements, which is a reference technique in the measurement of beta emissions [1;10-14].

47 For the other half of the autoradiography measurements, the observed discrepancy was due to
48 calibration problems including two successive unit changes:

49 1- From DLU cm^{-2} (Digital Luminescence Unit) to Bq cm^{-2} for the analyzed surface of the
50 contaminated sample. Contrary to digital radioimagers providing a direct particle counting per
51 area and per time, autoradiography technique using phosphor screens gives a signal per area in
52 the arbitrary unit DLU [10]. To convert DLU cm^{-2} into Bq cm^{-2} , standard specimens with known
53 activities must be exposed during the same time than samples of interest.

54 2- From Bq g^{-1} to Bq cm^{-2} for the standard specimens. The most often, standard specimens for
55 autoradiography consist of a set of strips whose activities are given in Bq g^{-1} (ART0123A for
56 ^3H , ARC146B for ^{14}C , from ARC Inc.). So, the surface activities of these strips (Bq cm^{-2}) need
57 to be estimated before performing the first conversion from DLU cm^{-2} to Bq cm^{-2} .

58 For many applications, there is also a need to estimate an activity per unit mass, when activity
59 per unit area can be determined with autoradiography technology or with Surface
60 Contamination Meters (SCMs), these last being widely used for decommissioning operations.
61 Until now, this conversion remains a challenging task.

62 The aim of this paper is thus to provide a useful conversion technique allowing to switch simply
63 and properly between surface and volumetric activities. This method is based on Geant4
64 simulations which provide the fraction of particles reaching a given surface, i.e. the Analyzed
65 Surface of a Contaminated Volume (ASCV), among all the particles emitting in this given
66 volume. This key parameter is known as the Emission Fraction F_E .

67 Three samples of rock and plastic, each contaminated with a different radionuclide (^3H , ^{14}C and
68 ^{238}U series), have been used in order to illustrate the conversion from Bq cm^{-3} to Bq cm^{-2} . The
69 emission fractions calculated with Geant4 for each sample, have allowed the conversion from
70 the known volumetric activities to the estimated surface activities. Surface activities have also
71 been determined experimentally using the gaseous radioimager BeaQuant™ (formerly known
72 as Beaver™). The detection efficiency ε of this detector, which corresponds to the number of

73 detected particles among the number of particles emitted from the ASCV, can thus be assessed
74 for three different radionuclides, just by comparing measured surface activities with the
75 estimated ones.

76 For a given device (radioimager or SCM), the detection efficiency varies from one radionuclide
77 to another according to the particle type and its energy of radiation. It is also possible that the
78 instrument detection efficiency for a given radionuclide depends on the material in which it is
79 emitted (known as “matrix effect”). More importantly, the particle counting measured with a
80 detector depends on the contamination thickness into the material. Both effects are carefully
81 investigated in the present work.

82

83 **1. Materials and Tools**

84 **1.1. Radioimager**

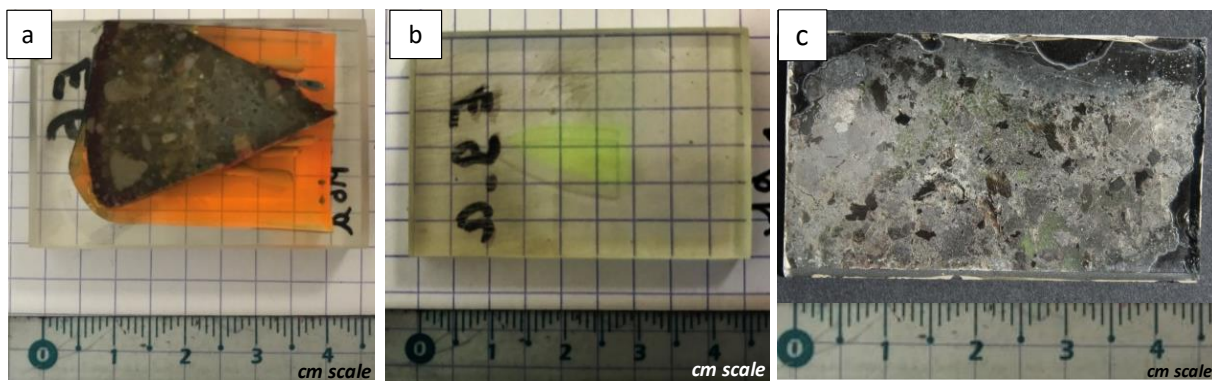
85 BeaQuant™ [15, 16], a gaseous device based on the combination of a parallel ionization
86 multiplier with a micro pattern gas detector, was used as a digital radioimager. Samples are
87 deposited in contact with the gas chamber, in such a manner that radioactive particles emerging
88 from a contaminated surface of the sample (i.e. the ASCV), interact with gas (ionization),
89 resulting in the creation of secondary electrons. Via an appropriate electric voltage applied
90 between sample (the cathode), micromeshes and anode, secondary electrons are multiplied and
91 then located onto the 2D pixelized anode. Then, each initial particle which triggers an electronic
92 acquisition is reconstructed providing an autoradiography image formation in real time.

93 The maximal spatial resolution obtained for both ^3H and alpha detection is 20 μm , while the
94 sensitivity can reach 8×10^{-4} cps cm^{-2} [15]. It can also be noted that BeaQuant™ is not affected
95 by X or gamma rays, and that the detection response is linear with the activities and covers 5
96 orders of magnitude. Moreover, BeaQuant™ is able to separate alpha and beta emissions, based
97 on a threshold applied on the energy deposited in gas (far higher for alpha than for beta), and

98 on the selection of adequate acquisition settings supplied by the manufacturer AI4R [15]. A
99 sample holder called GS with a rectangular area for analysis of 12 cm by 8 cm, providing a total
100 Detector Surface area $S_D = 96 \text{ cm}^2$, was used for the measurements.

101 1.2. Samples

102 Three laboratory samples of interest were used to experimentally illustrate the proposed
103 conversion method and determine the detection efficiency of BeaQuant™ (Figure 1).



104 **Fig. 1** Samples pictures. a- M6 sample is a triangular piece of mortar impregnated and
105 surrounded with ^3H -PMMA resin (orange). b- M9 sample is a triangular piece of ^{14}C -PMMA
106 (yellow). c- M8 sample is a thin section of granite, including β -uranophane (U-bearing
107 minerals)
108

109

110 The first sample (M6) is a triangular piece of mortar which has been impregnated with a ^3H
111 doped PMMA resin (pure beta emitter) [17]: a significant area of homogeneous pure ^3H -PMMA
112 resin is also present around the mortar (orange colored zone in Figure 1a). The second sample
113 (M9) is a sub-triangular piece (yellow zone in Figure 1b) of homogeneous resin of ^{14}C doped
114 PMMA (pure beta emitter), surrounded by non-radioactive colorless resin. Volumetric activity
115 of the PMMA resin is known for each of the two samples (Table 1).

116 Finally, the third sample (M8) is a granite thin section (rock thickness of 30 μm) stucked on a
117 glass slide (Figure 1c). It contains natural uranium bearing minerals known as β -uranophane
118 (54.4 wt% U) distributed in small clusters. The age of the rock (150 Ma) and alpha spectrometry
119 attest the secular equilibrium of ^{238}U series [9]. For each ^{238}U disintegration, the 13 daughters
120 disintegrate too, leading to the simultaneous emission of eight alpha and six beta particles, plus
121 all the associated emissions: gamma and X-rays, Auger and conversion electrons. However,
122 when analyzing with BeaQuantTM, the option allowing counting only alpha particles has been
123 chosen. Therefore, all the other emissions produced by ^{238}U decay chain are not investigated in
124 this work (anyway, BeaQuantTM is not sensitive to gamma and X-rays).

125 A theoretical Eq. (1) proposed in [18], was employed to estimate the activity of each alpha
126 emission from 1 cm^3 of β -uranophane crystal (i.e. the Volumetric Activity of one alpha $A_{V,\alpha 1}$):
127 a value of 26409 Bq cm^{-3} is found (Table 1) for each alpha emission, giving a total activity for
128 the eight alphas of 221271 Bq cm^{-3} .

$$129 \quad A_{V,\alpha 1} = \rho \lambda \left(\frac{C_x}{100} \frac{N}{A_x} \right) \quad (1)$$

130 Where ρ is the density of β -uranophane (3.9 g cm^{-3}), λ is the disintegration constant of ^{238}U
131 ($0.492 \times 10^{-17} \text{ s}^{-1}$), C_x is the weight fraction of uranium in β -uranophane (54.4 wt%) measured
132 by microprobe with a wavelength-dispersive spectroscopy system (Cameca SX100, Camparis
133 facilities, University Pierre and Marie Curie, Paris), N the Avogadro number (mol^{-1}) and A_x
134 the molar mass of uranium ($238.029 \text{ g mol}^{-1}$).

135 Detailed features and properties of these three samples are provided in Table 1.

136

137 **Table 1** Sample properties. For M8 sample, data are separated between granite rock under
138 exponent (1) and β -uranophane under exponent (2), granite being the whole sample, β -
139 uranophane a component of the sample

Sample	M6	M9	M8
Material	PMMA resin	PMMA resin	⁽¹⁾ granite (rock) ⁽²⁾ β -Uranophane mineral
Radionuclide	³ H	¹⁴ C	²³⁸ U series ⁽²⁾
Chemical formula	(C ₅ H ₈ O ₂) _n	(C ₅ H ₈ O ₂) _n	Ca(UO ₂) ₂ (SiO ₃ OH) ₂ ·5H ₂ O ⁽²⁾
Density ρ (g cm ⁻³)	1.19	1.19	3.9 ⁽²⁾
Contamination thickness d_C (μ m)	7000	500	30 ^{(1) and (2)}
Approximative surface area (cm ²)	5	1	10 ⁽¹⁾ , < 1 ⁽²⁾
Known Volumetric Activity $A_{V,K}$ (Bq cm ⁻³)	7 000 000	330 000	26 409 ^{(2) *}

140 Footnote to table 1 : * the volumetric activity given for M8 sample refers to each alpha emission
141 of the ²³⁸U decay chain. This value needs to be multiplied by the number of alpha (eight in ²³⁸U
142 series) to obtain the total activity of alphas in β -uranophane

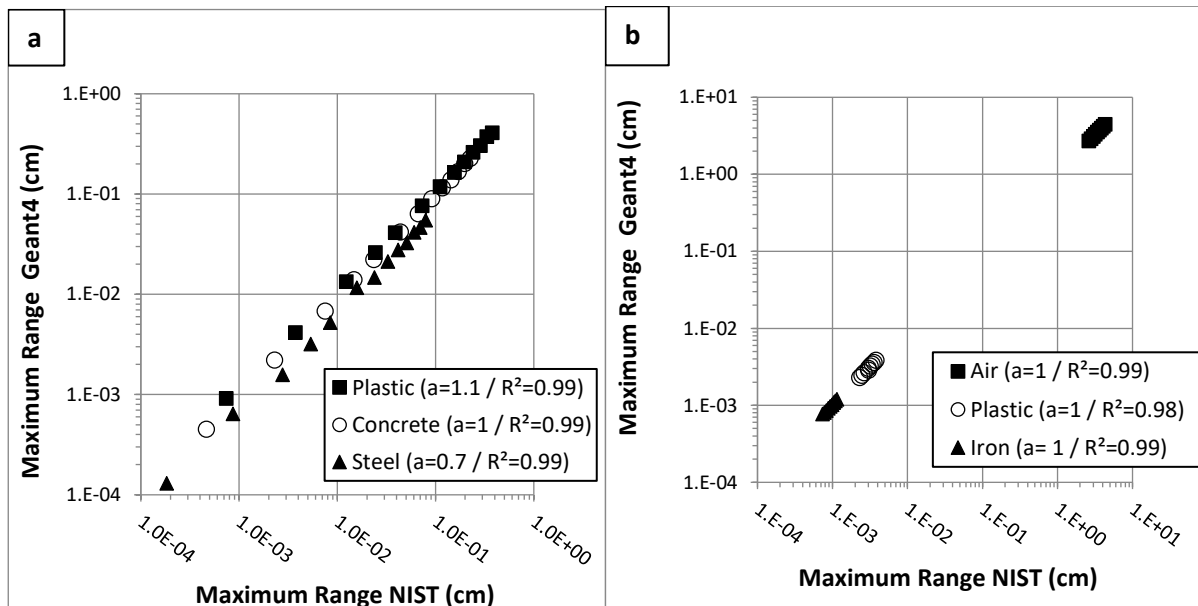
143 1.3. Geant4 simulations and validation

144 Geant4 (GEometry ANd Tracking 4) is a powerful and C++ based toolkit allowing to simulate
145 the transport of all kinds of particles through the matter [19]. It was used in this work to model
146 the transport of beta and alpha particles emitted from the three samples studied here. An
147 accurate list of the sample materials including chemical compositions (loaded in Geant4) is
148 given in supplementary data (Table 1). Energy spectra of beta emissions have been extracted
149 from database provided in the Rad Toolbox software developed by the Oak Ridge National
150 Laboratory [20]. The energy distributions for the alpha emissions, have been extracted from
151 standard [21]. Simulations have been achieved considering 10⁷ beta or alpha particles, ensuring
152 a valuable statistical analysis.

153 Geant4.10.02.p02 version was used for this study with a physics list based on the
154 electromagnetic standard processes and models (EM standard physics list). The creation of a
155 specific physics list allows to customize some settings instead of using the proposed default
156 values.

157

158 In order to validate the implemented physics in Geant4, the Maximum Range values R_{MAX} of
159 beta particles travelling in plastic (i.e. PMMA), concrete and steel (materials defined in
160 supplementary data Table 1) with kinetic energy ranging from 20 keV to 1 MeV, were
161 calculated using both ESTAR program from NIST database [22] (National Institute of
162 Standards and Technology), and Geant4 simulations. Both tools provide similar R_{MAX} values,
163 with linear correlations displaying a slope a close to 1 as well as a correlation coefficient R^2 of
164 0.99 (Figure 2a and supplementary data Table 2), except for beta radiations in steel where $a =$
165 0.7. This discrepancy might be due to additional physics processes involved in the Geant4
166 physics list for which the cross-sections are enhanced by high density material such as steel.
167 The same comparison has been performed for alpha particles travelling in air, plastic and iron,
168 using ASTAR program from NIST database [22] and Geant4: the obtained R_{MAX} values are very
169 close as illustrated by the slope $a = 1$ and the $R^2 > 0.98$ for the linear correlations (Figure 2b and
170 supplementary data Table 3).



171

172 **Fig. 2** Comparison of maximum range values (linear correlation with a slope “ a ” and a
173 correlation coefficient “ R^2 ”): NIST [22] Versus Geant4. a- beta emissions from 20 to 1000
174 keV. b- alpha emissions from 4000 to 5600 keV

175

176

2. Method

177

2.1. Detection efficiency calculation

178

Each radiation detector has a detection efficiency ε depending on the radiation energy and on

179

the radiation type (alpha, beta, gamma). The geometry of the detector has also a great influence

180

on the detection efficiency, because the solid angle of particles emissions depends on 1- the

181

distance between the Analyzed Surface of the Contaminated Volume (ASCV) and the sensitive

182

part of the detector, and 2- the ratio between contamination and detector surface areas. The

183

nature of the contaminated material, i.e. the material in which particles travel, could also impact

184

the detection efficiency. Indeed, the energy of particles, which decreases at each interaction

185

with matter, can be different at the analyzed surface of a contaminated material A, compared to

186

the analyzed surface of a contaminated material B, because of the matrix effect.

187

To determine precisely the efficiency of a SCM, the ISO standard 7503-3 [23] recommends to

188

use calibrated sources with $10 \times 10 \text{ cm}^2$ active area and certified alpha, beta or gamma Emission

189

Rate ER in 2π (provided in counts per second (cps) for the total contaminated Surface area of

190

the Source S_S), and to apply the following Eq. (2):

191

$$\varepsilon = \frac{(SCR_S - SCR_B)}{SER_S} \quad (2)$$

192

$$\text{with } SER_S = \frac{ER_S}{S_S}, SCR_B = \frac{CR_B}{S_D}, SCR_S = \frac{CR_S}{S_D} \text{ if } S_S > S_D, \text{ or } SCR_S = \frac{CR_S}{S_S} \text{ if } S_D > S_S$$

193

194

where SCR_S is the Surface Count Rate measured on the calibrated Source in cps cm^{-2} , SCR_B the

195

Surface Count Rate measured for the Background in cps cm^{-2} , SER_S the certified Surface

196

Emission Rate in 2π of the calibrated Source in cps cm^{-2} . A Surface Count Rate SER being a

197 Count Rate CR divided by the related surface area of counting: - S_D the Surface area of the
 198 sensitive part of the Detector (cm^2) or - S_S the Surface area of a calibrated Source in cm^2 .
 199 Then, the Surface Count Rate measurement (SCR_C) of a homogeneously Contaminated surface,
 200 for which the radionuclide is known, can be corrected by the factor $1/\varepsilon$ of the corresponding
 201 radiation type and energy region to obtain the Surface Emission Rate SER_C of the
 202 Contamination (Eq. (3)).

$$203 \quad SER_C = \frac{(SCR_C - SCR_B)}{\varepsilon} \quad (3)$$

$$204 \quad \text{with } SER_C = \frac{ERC}{S_C}, SCR_B = \frac{CR_B}{S_D}, SCR_C = \frac{CR_C}{S_D} \text{ if } S_C > S_D, \text{ or } SCR_C = \frac{CR_C}{S_C} \text{ if } S_D > S_C$$

205

206 The autoradiography technology allows to map a contaminated surface to calculate its surface
 207 area value S_C : it is thus possible to choose the relevant area to calculate SCR_C from the Count
 208 Rate of the contamination CR_C (cps) (Eq. (3)). For other radioactivity detectors, generally one
 209 of the two assumptions (S_D or S_C) has to be chosen.

210 The detection efficiency ε of the radioimager BeaQuant™ is investigated in this work for pure
 211 beta emitters (^3H and ^{14}C) and for alpha emitters from the entire decay chain of ^{238}U (eight
 212 alphas). To this purpose, the three samples (see Figure 1) were used: although they are not
 213 referenced sources with certified surface emission rate in 2π , volumetric activities of M6 and
 214 M9 samples were determined in laboratory by liquid scintillation counting (Table 1). For the
 215 M8 sample, the theoretical Eq. (1) proposed by [18] was used to estimate the volumetric activity
 216 of each alpha emitter of uranium bearing minerals, as already stated above. Surface activities
 217 have been estimated from these volumetric activities, in order to be compared with BeaQuant™
 218 surface measurements.

219 2.2. Concept of Emission Fraction F_E

220 The ISO standard 7503-1 [24] proposes to use an Emission Fraction F_E to convert Surface
221 Emission Rate SER (cps cm⁻²) into Surface Activity A_S (Bq cm⁻²) (Eq. (4)).

$$222 \quad A_S = \frac{SER_C}{F_E} = \frac{(SCR_C - SCR_B)}{\varepsilon F_E} \quad (4)$$

223 F_E is the ratio between the number of particles emerging from the ASCV (or in other terms the
224 face of a contaminated sample in contact with the detector window) and the number of particles
225 produced in the contaminated volume. However, the concept of F_E presented in [24] is based
226 on the assumption that the contamination layer is infinitely thin, eliminating the concept of
227 volume. Accordingly, the 2π geometry yields to a reduction of the detectable emissions by a
228 factor of 2 [23], providing an estimation of F_E equal to 0.5. However, in the case of thick
229 contaminations, which is the general case found for nuclear wastes or geological samples, such
230 an assumption induces a significant bias between Surface Activity A_S and Volumetric Activity
231 A_V . In other terms, Eq. (4) proposed in the ISO standard 7503-1 [24] to calculate A_S , is not truly
232 adapted to thick contaminated samples when considering $F_E = 0.5$.

233
234 Emission Probability P_E , described as the ratio between the number of particles created and the
235 number of decays of the same radionuclide, is equal to 1 for the three samples analyzed with
236 BeaQuant™. Indeed, M6 and M9 are pure beta emitters (³H and ¹⁴C). Concerning M8 in which
237 only alpha particles are considered, P_E could be considered equal to 1 for each of the eight alpha
238 emissions of the ²³⁸U decay chain. In this work, P_E equal to 1 (i.e. cps = Bq) allows the direct
239 conversion from the measured Surface Count Rate SCR_C (cps cm⁻²) to the Surface Activity A_S
240 (Bq cm⁻²), after correction from the corresponding instrument efficiency ε and from the
241 background measurement, without having to involve the Fraction Emission F_E (Eq. (5)).

$$242 \quad A_S = \frac{SCR_C - SCR_B}{\varepsilon} = SER_C \quad (5)$$

243

244 In the present study, an appropriate estimation of F_E is used to calculate a Volumetric Activity
245 A_V in Bq cm^{-3} (next section 2.3.), instead of a Surface Activity A_S in Bq cm^{-2} of a contamination
246 layer infinitely thin (as proposed in [24] and Eq. (4)).

247 The Emission Fraction F_E is difficult to estimate because it depends on the nature of the
248 contaminated material (matrix), the radionuclide (particle type and emission energy) and the
249 Contamination thickness d_C . The contamination is considered as homogeneous in the whole
250 material. For a given d_C , the denser the material is, the lower the number of particles of a given
251 type and energy emerge on the ASCV. For a same material, high energy beta particles are able
252 to cross over longer distances than low energy betas or alphas. This leads to a higher value of
253 F_E considering the same Contaminated thickness d_C . Finally, for a given combination of a
254 material and a radionuclide, the greater d_C is, the lower will be the fraction F_E of particles
255 emerging from the total volume, through the ASCV. Indeed, particles are more likely to be
256 stopped in a thick material, before to reach the ASCV, than in a thin one. A proper simulation
257 procedure using Geant4 has been performed to answer to these difficulties.

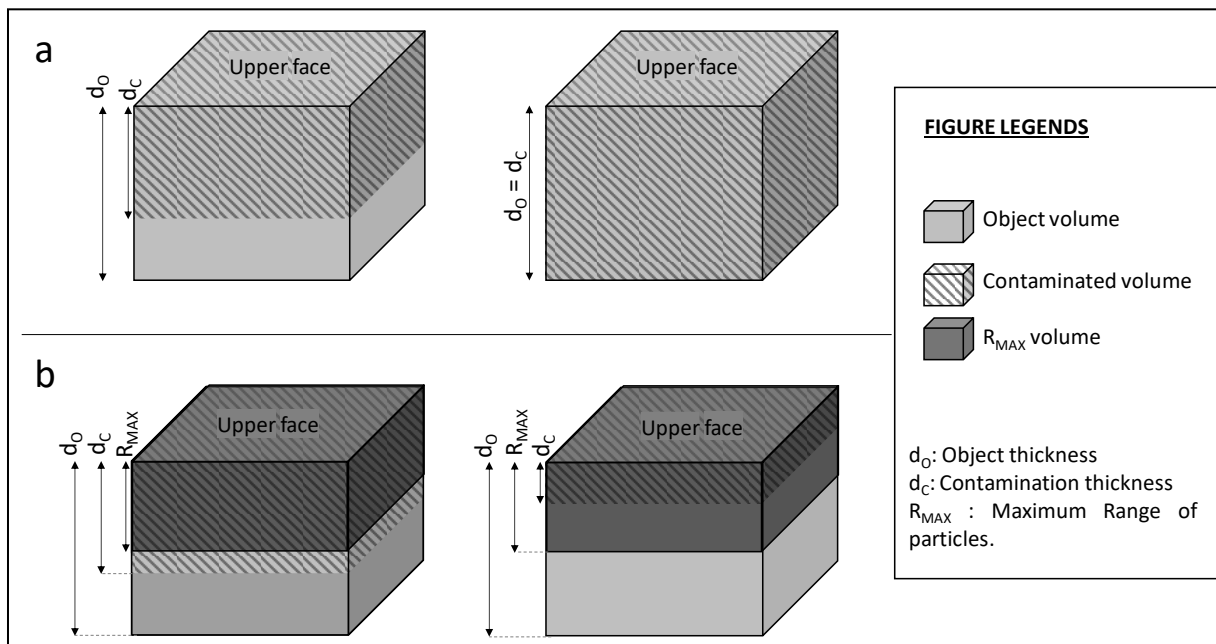
258 2.3. Estimation of the Emission Fraction F_E using Geant4 simulations

259 A contaminated material can be depicted as a parallelepiped object (Figure 3a), in which three
260 thicknesses are defined: 1- apparent Object thickness d_O (which can be considered as infinite in
261 the case of contaminated floor and/or wall), 2- Contamination thickness d_C (Figure 3a),
262 corresponding to the depth of the contamination, 3- Maximum Range R_{MAX} of the particles
263 responsible of the contamination in the considered material.

264 Knowing that d_O is thus necessarily greater or equal to d_C , d_O is not a significant thickness in
265 the present work, while d_C needs to be known. R_{MAX} can be lower, equal or higher than d_C . If
266 $R_{MAX} < d_C$, a part of the contaminated volume is not explored by the autoradiography technique

267 (or by any other SCM), because particles emitted deeper than the maximum range cannot reach
 268 the ASCV (Figure 3b). Thus, it is not relevant to estimate F_E for contamination thicknesses d_C
 269 $> R_{MAX}$, even if it could happen in real samples.

270



271

272 **Fig. 3** Definitions of important thicknesses for a contaminated sample. The upper face
 273 corresponds to the Analyzed Surface of the Contaminated Volume (ASCV). a- The
 274 Contamination thickness d_C is lower or equal to the Object thickness d_O . b- The Maximum
 275 Range R_{MAX} can be lower, equal or greater than the Contamination thickness d_C

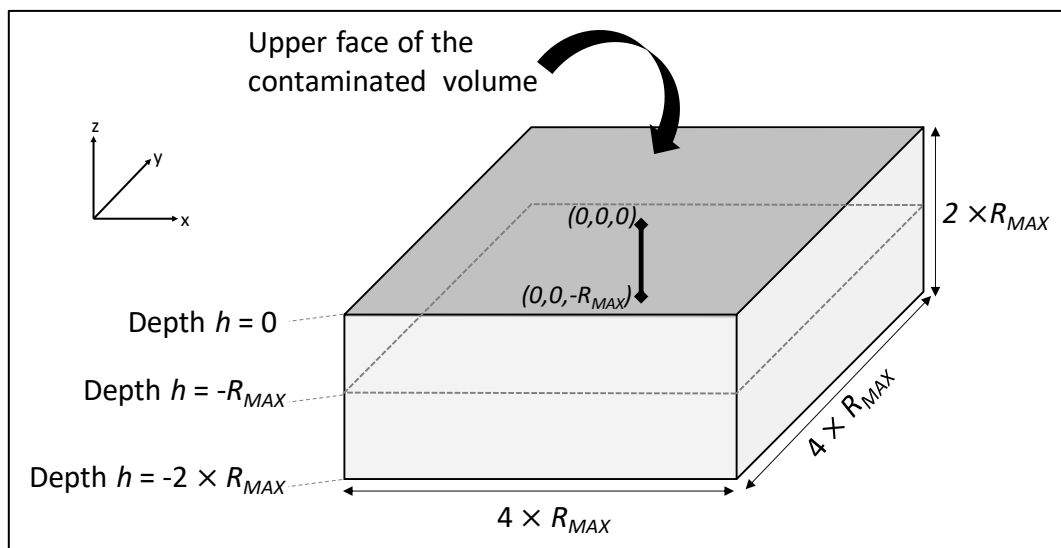
276

277 Geant4 was used to calculate the fraction F_E of the volumetric activity detectable on the ASCV,
 278 for the three studied samples previously described in section 1.2. F_E will then allow to switch
 279 from volumetric to surface activities.

280 The first step consisted to calculate with Geant4, the R_{MAX} values in the given material, using
 281 the maximal emission energy of the considered radiation [20, 21]. Then, a rectangular
 282 parallelepiped of $4 \times R_{MAX}$ in width, $4 \times R_{MAX}$ in length and $2 \times R_{MAX}$ in height, filled with the

283 material to consider, was build (Figure 4). Into this volume, 10^7 particles corresponding to one
 284 of the studied radionuclides have been generated with initial random directions and positions
 285 in a segment. This segment ranges from the origin $(0, 0, 0)$ to the point $(0, 0, -R_{MAX})$ (see Figure
 286 4). These conditions and dimensions were chosen in order that any emitted particle can possibly
 287 reach the upper face of the material volume, i.e. the ASCV. For each emitted particle, the initial
 288 kinetic energy is selected in the corresponding energy distribution [20, 21]. The fraction of
 289 particles emerging at the ASCV, according to their emission depth h , was calculated, h ranging
 290 from the surface ($h = 0$) to $h = -R_{MAX}$. Then, an integral calculation has been achieved to
 291 estimate the fraction of particles reaching the ASCV, i.e. the emission fraction according to the
 292 thickness d of the considered layer, $F_E(d)$. The numerical Steps s of $F_E(d)$ are adjusted according
 293 to the order of magnitudes of the R_{MAX} values: $s = 0.01 \mu\text{m}$ for ^3H , $s = 0.1 \mu\text{m}$ for ^{14}C and the
 294 alpha emitters of the ^{238}U decay chain.

295



296

297 **Fig. 4** Scheme of the geometry chosen in the Geant4 simulations. The parallelepiped object
 298 corresponds to the contaminated volume centered in $(0, 0, -R_{MAX})$. The upper face, i.e. the
 299 Analyzed Surface of the Contaminated Volume (ASCV), is centered in $(0, 0, 0)$. Particles are
 300 emitted from the segment ranging from $(0, 0, 0)$ to $(0, 0, -R_{MAX})$

301

302 With these sets of F_E values, it is then possible to evaluate the Surface Activity A_S (in Bq cm⁻²)
303 of a sample with known Volumetric Activity A_V (in Bq cm⁻³), and conversely. For a given case
304 (a material and a radiation type), if the Contamination thickness $d_C > R_{MAX}$, the activities need
305 to be calculated with a thickness equal to R_{MAX} , and $F_E(R_{MAX})$ has to be used (Eq. (6a)).
306 Otherwise, if $d_C < R_{MAX}$, activities have to be calculated with $F_E(d_C)$ (Eq. (6b)).

307
$$A_{S,G} = A_{V,K} \times F_E(R_{MAX}) \times R_{MAX} \quad or \quad A_{V,G} = \frac{A_{S,K}}{F_E(R_{MAX}) \times R_{MAX}} \quad if \ d_C > R_{MAX} \quad (6a)$$

308
$$A_{S,G} = A_{V,K} \times F_E(d_C) \times d_C \quad or \quad A_{V,G} = \frac{A_{S,K}}{F_E(d_C) \times d_C} \quad if \ d_C < R_{MAX} \quad (6b)$$

309

310 Where $A_{S,G}$ is the Surface Activity estimated with Geant4 simulation tool (Bq cm⁻²), $A_{V,K}$ the
311 Known Volumetric Activity (Bq cm⁻³), $A_{V,G}$ the Volumetric Activity estimated with Geant4
312 simulation tool (Bq cm⁻³) and $A_{S,K}$ the Known Surface Activity (Bq cm⁻²). Obviously, the
313 important assumption of the model is to consider that the activity is homogeneously distributed
314 in the volume.

315

316 Surface activities of calibrated sources are generally known to calculate the detection efficiency
317 of a detector (Eq. (2)), but for controlled samples (the case in the present work), surface
318 activities needed to be evaluated from their known volumetric activities (Eq. (6)): application
319 of the conversion from Bq cm⁻³ to Bq cm⁻². Then for any contaminated material with $P_E = 1$, the
320 measured Surface Count Rate SCR_C can be converted into volumetric activity (Eq. (6)), after
321 detection efficiency correction (Eq. (5)): use of the conversion from Bq cm⁻² to Bq cm⁻³.
322 However, this last assumption is accurate only if the detection efficiency of the instrument is
323 independent of any matrix effect.

324

325 2.4. Method for evaluating the matrix effects on detection efficiency

326 In this section, the impact of the contaminated material type on the detection efficiency of a
327 given instrument is examined. Indeed, it is not trivial to know if the detection efficiency of a
328 radionuclide obtained on a calibrated source or a well characterized sample (Eq. (2)) can be
329 employed to determine the surface activity of a contaminated object emitting the same
330 radionuclide (Eq. (5)), but composed with another material.

331 To clarify this point, it is necessary to determine the kinetic energy distribution of the emitted
332 particles when they reach the ASCV. Indeed, for almost all the common radiation detectors,
333 and in this work for autoradiography devices, the detection efficiency is directly linked to the
334 energy of the particles.

335 Geant4 was used to plot the frequency distribution (fd) of the kinetic energy of particles (beta
336 and alpha) just as they go out from the ASCV, considering three different materials: plastic,
337 concrete and steel. In these simulations, the contaminated material is still depicted as a
338 rectangular parallelepiped $4 \times R_{MAX}$ long and width, and a thickness (height) of $2 \times R_{MAX}$ (R_{MAX}
339 of the considered radionuclide in one of the chosen materials (Figure 4)). A second series of
340 simulations has also been achieved with contaminated volume presenting a constant
341 Contamination thickness d_C , regardless of the material.

342 Finally, it is important to emphasize that the matrix effect described here is only related to the
343 detection efficiency of a given instrument, i.e. the ratio of the number of particles detected to
344 the number of particles emitted at the ASCV. The number of particles emitted at the ASCV,
345 given by the Emission Fraction F_E , is however strongly dependent on the nature of the material,
346 as previously explained (end of the section 2.2.)

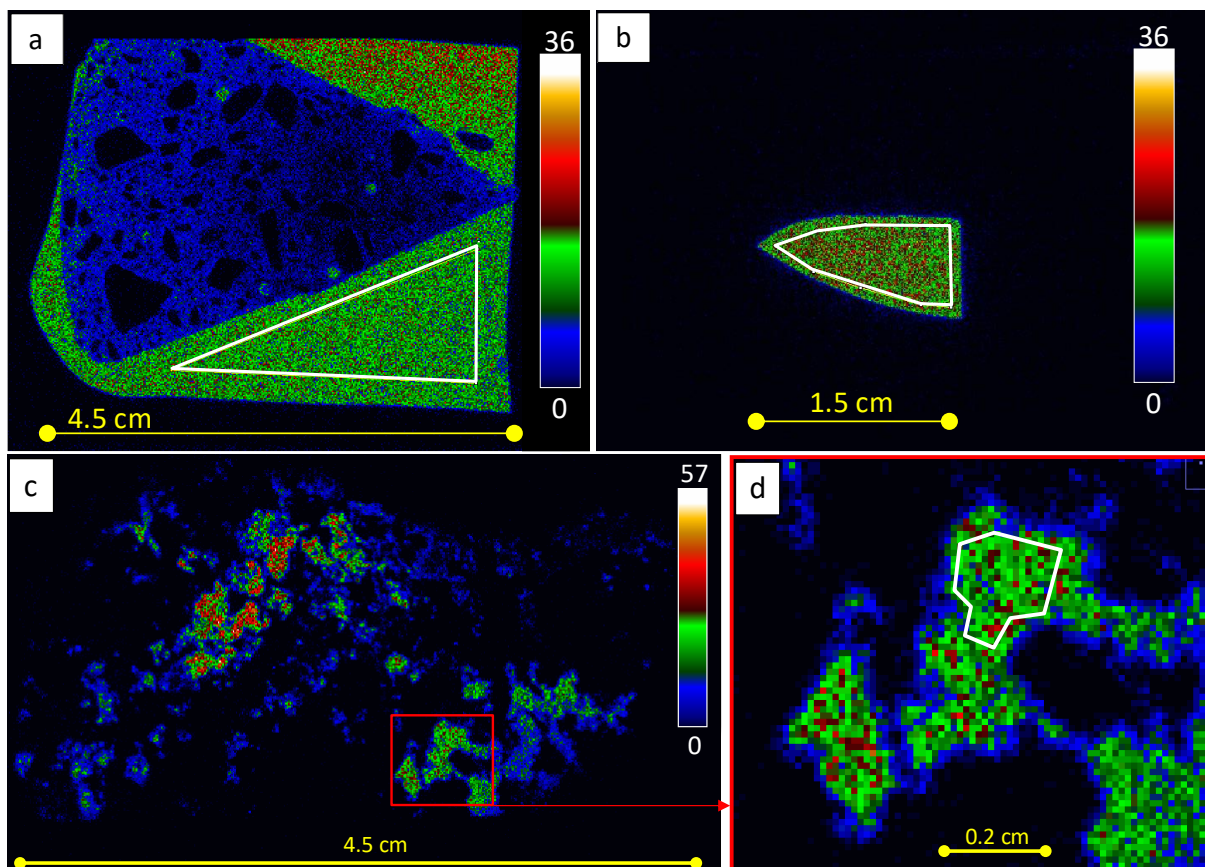
347 3. Results and discussion

348

3.1. Surface Count Rate measurements with BeaQuant™

349 Each of the three samples M6, M8 and M9 was acquired with the radioimager BeaQuant™
350 during one hour, in beta mode for M6 and M9, and in alpha mode for M8. On the
351 autoradiographs obtained, the Contaminated Surface areas S_C on which the count rate
352 measurements were performed are outlined with a white frame (Figure 5). S_C values (cm²) are
353 given in Table 2, together with the count rates measured. To calculate the Surface Count Rate
354 (SCR_C), note that S_C are always lower than S_D (Detector Surface area) in the present work.

355



356

357 **Fig. 5.** a- Autoradiography of the beta emissions from the M6 sample, a mortar (triangular
358 shape in blue and black) impregnated with ³H-PMMA resin. Resin surrounds also the mortar
359 (in green and red). b- Autoradiography of the beta emissions from a triangular piece of ¹⁴C-
360 PMMA (M9 sample). c- Autoradiography of the alpha emissions from uranium bearing
361 minerals (β -uranophane) included in the granite M8. D- Zoom on the red frame of image c.

362 Autoradiographs are acquired with BeaQuant™ and with the GS 12 × 8 cm² sample holder.
 363 Color scales give the particle counts in 1 hour, pixel size is fixed at 100 μm and surface count
 364 rate measurements have been performed on the surface areas delimited with a white frame

365

366 In M6 sample, the mortar corresponds to the central triangular area which is heterogeneous
 367 (Figure 5a). It is a mixture of non-radioactive grains (in black) and pores filled with the
 368 radioactive resin ³H-PMMA (in blue). The two homogenous areas surrounding the mortar are
 369 made of pure ³H-PMMA resin, on which the measurement was performed. The M9 sample
 370 shows a sub-triangular and homogeneous piece of ¹⁴C-PMMA resin (Figure 5b). Then, the
 371 autoradiography of M8 sample depicts the spatial distribution of the β-uranophane crystals into
 372 the rock (Figure 5c); a nearly homogeneous and small area corresponding to a cluster of β-
 373 uranophane crystals was chosen to perform the measurement (Figure 5d).

374

375 **Table 2** Measurements of the three samples with BeaQuant™ using the GS 12 × 8 cm² sample
 376 holder: Contamination Surface area S_C , Count Rate of this surface CR_C and Surface Count Rate
 377 SCR_C

Sample	Radionuclide	Particle	Used setting	S_C (cm ²)	CR_C (cps)	SCR_C
					for S_C	(cps cm ⁻²)
M6	³ H	beta	beta	$1.00 \times 10^{+00}$	38.21	38.10
M9	¹⁴ C	beta	beta	6.29×10^{-01}	30.09	47.85
M8	²³⁸ U decay chain	alpha	alpha	2.51×10^{-02}	1.60	63.90

378

379 Blank measurements were also performed in order to obtain the Background Surface Count
 380 Rate of the detector (SCR_B), using the sample holder GS (12 × 8 cm²) and the two different
 381 acquisition settings (Table 3). These blank values, needed to the further calculations of the

382 detection efficiency of BeaQuant™ (Eq. (2)), are typically 4 to 5 orders of magnitude lower
 383 than the three samples surface count rates.

384

385 **Table 3** Background measurements of BeaQuant™ (GS 12 × 8 cm² sample holder). Detector
 386 Surface area S_D , Background Count Rate CR_B and Background Surface Count Rate SCR_B

Setting	S_D (cm ²)	CR_B (cps)	SCR_B (cps cm ⁻²)
		for $S_D = 96$ cm ²	
beta	96	2.65	2.76×10^{-02}
alpha	96	0.13	1.35×10^{-03}

387

388 3.2. Surface Emission Rate estimations: from Bq cm⁻³ to Bq cm⁻²

389 The Volumetric Activities ($A_{V,K}$) in Bq cm⁻³ are known for the M6 and M9 samples, and
 390 calculated for each alpha emission of the M8 sample (due to the secular equilibrium of the ²³⁸U
 391 decay chain, all the daughters have the same activity) (Tables 1 and 4). The conversion method
 392 has been applied to estimate the Surface Activity ($A_{S,G}$) in Bq cm⁻² of the three samples, i.e.
 393 convert the Bq cm⁻³ (Table 1) into Bq cm⁻², using Eq. (6). Because P_E of ¹⁴C, ³H and each alpha
 394 of the ²³⁸U decay chain are equal to 1, these estimated Surface Activities $A_{S,G}$ are equal to the
 395 Surface Emission Rate SER_C (i.e. Bq = cps). The comparison between the Surface Count Rates
 396 (SCR_C) measured previously with BeaQuant™ and these estimated Surface Emission Rates
 397 (SER_C) would allow then to assess the detection efficiency of BeaQuant™ (next section), using
 398 Eq. (2). The three samples are indeed used as calibrated sources.

399 As previously mentioned, the first step of the conversion method is the calculation of the
 400 maximum ranges of particles emitting in the three samples. These R_{MAX} values are displayed in
 401 Table 4, with the related maximum energy used.

402 **Table 4** Estimation of the Surface Activity $A_{S,G}$ of the M6, M8 and M9 samples (since $P_E = 1$, $A_{S,G} = SER_C$ (Surface Emission Rate in cps cm^{-2})). The Known
403 Volumetric Activities $A_{V,K}$ and the Contamination thicknesses d_C have been reported from Table 1. Maximum Ranges values R_{MAX} have been calculated with
404 Geant4, using the maximum energy also reported. According Eq. (6), the Emission Fraction F_E is calculated for the smaller of the two, between d_C and R_{MAX} :
405 for M6 and M9 samples, $F_E(R_{MAX})$ is used to perform the conversion Bq cm^{-3} to Bq cm^{-2} , while for M8 sample $F_E(d_C)$ is used

Sample	Material	Radionuclide	Particle	Known Volumetric	Contamination	Maximum	Maximum	Emission	Emission	Estimated Surface
				Activity $A_{V,K}$ (Bq cm^{-3})	thickness d_C (cm)	Energy (keV)	Range R_{MAX} (cm)	Fraction $F_E(d_C)$	Fraction $F_E(R_{MAX})$	Activity $A_{S,G}$ (Bq cm^{-2}) (or SER_C (cps cm^{-2}))
M6	PMMA	^3H	beta	7 000 000	7×10^{-01}	18.6	8.40×10^{-04}	-	0.026	153
M9	PMMA	^{14}C	beta	330 000	5×10^{-02}	156.5	2.78×10^{-02}	-	0.034	312
M8 – ^{238}U Decay Chain	β -Uranophane mineral	^{238}U	alpha	26 409	3×10^{-03}	4198	1.73×10^{-03}	0.138	-	11
		^{234}U	alpha	26 409	3×10^{-03}	4775	2.09×10^{-03}	0.166	-	13
		^{230}Th	alpha	26 409	3×10^{-03}	4687	2.03×10^{-03}	0.162	-	13
		^{226}Ra	alpha	26 409	3×10^{-03}	4784	2.10×10^{-03}	0.167	-	13
		^{222}Rn	alpha	26 409	3×10^{-03}	5490	2.57×10^{-03}	0.204	-	16
		^{218}Po	alpha	26 409	3×10^{-03}	6002	2.92×10^{-03}	0.233	-	19
		^{214}Po	alpha	26 409	3×10^{-03}	7687	4.23×10^{-03}	0.316	-	25
		^{210}Po	alpha	26 409	3×10^{-03}	5304	2.44×10^{-03}	0.192	-	15
Average alpha							$2,51 \times 10^{-03}$	0.197		

Σ alpha	211 272	125
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409 Then the Emission Fraction F_E has been calculated with Geant4. As the Contamination
410 thicknesses d_C of the M6 and M9 samples (0.7 cm and 0.05 cm respectively) are higher than the
411 maximum range of the corresponding radiation in PMMA (8.4×10^{-4} cm for M6 and $2.78 \times$
412 10^{-2} cm for M9), F_E was estimated in a volume of thickness R_{MAX} (Table 4). Concerning the M8
413 sample, as the Maximum Ranges R_{MAX} of the eight alphas (Table 4) are of the same order of
414 magnitude than the sample Contamination thickness d_C (3×10^{-3} cm), the Emission Fraction F_E
415 of each alpha has been estimated for the Contamination thickness d_C (Table 4). Thus, the
416 Surface Activities $A_{S,G}$ ($= SER_C$ since $P_E = 1$) presented in Table 4 are calculated with Eq. (6a)
417 for M6 and M9 samples, and with Eq. (6b) for each alpha of M8 sample. The sum of the alphas
418 activity has been performed in order to be compared with the total alphas counting measured
419 with BeaQuant™.

420

421 To conclude, it is crucial to always consider that autoradiographic method remains a surface
422 measurement technique. For the analyzed samples of the present work, the Contamination
423 depths d_C were higher or equal to R_{MAX} . In the case of samples with $d_C < R_{MAX}$, $F_E(d)$ for ^3H and
424 ^{14}C radionuclides emitted in PMMA (plastic), concrete and steel are provided from $d = 0$ to d
425 $= R_{MAX}$ in the companion paper [25]. However, the analysis of a sample where d_C is unknown
426 can be problematic, especially if $d_C < R_{MAX}$. The unique way to solve properly that issue would
427 be to determine d_C by measuring the contamination profile as a function of depth.

428 3.3. Detection efficiency calculation of BeaQuant™

429 Comparing surface count rate measurements with surface emission rate estimations (Eq. (2)),
430 the detection efficiency of BeaQuant™ (sample holder reference GS), can be calculated for the
431 three samples which substitute the calibrated sources in the present work (Table 5). In the M6
432 sample emitting ^3H in PMMA (plastic), 25% of the particles emitted through the ASCV are
433 detected, whereas only 15% of betas emitted from ^{14}C -PMMA (M9 sample) are collected.

434 Concerning ^{238}U decay chain, alphas emitted in the β -uranophane crystals (M8 sample) are
 435 detected with an efficiency of 51%.

436

437 **Table 5** Instrument efficiencies ε of BeaQuantTM using the GS $12 \times 8 \text{ cm}^2$ sample holder. After
 438 correction from the Background Surface Count Rate SCR_B , the Surface Count Rate SCR_C is
 439 compared to the Surface Emission Rate SER_C expected for the samples (Eq. (2))

Sample	Radionuclide	Particle	Used setting	SCR_C (cps cm^{-2})	SCR_B (cps cm^{-2})	SER_C (cps cm^{-2})	ε
M6	^3H	beta	beta	38.10	2.76×10^{-02}	153	0.25
M9	^{14}C	beta	beta	47.85	2.76×10^{-02}	312	0.15
M8	^{238}U decay chain	alpha	alpha	63.90	1.35×10^{-03}	125	0.51

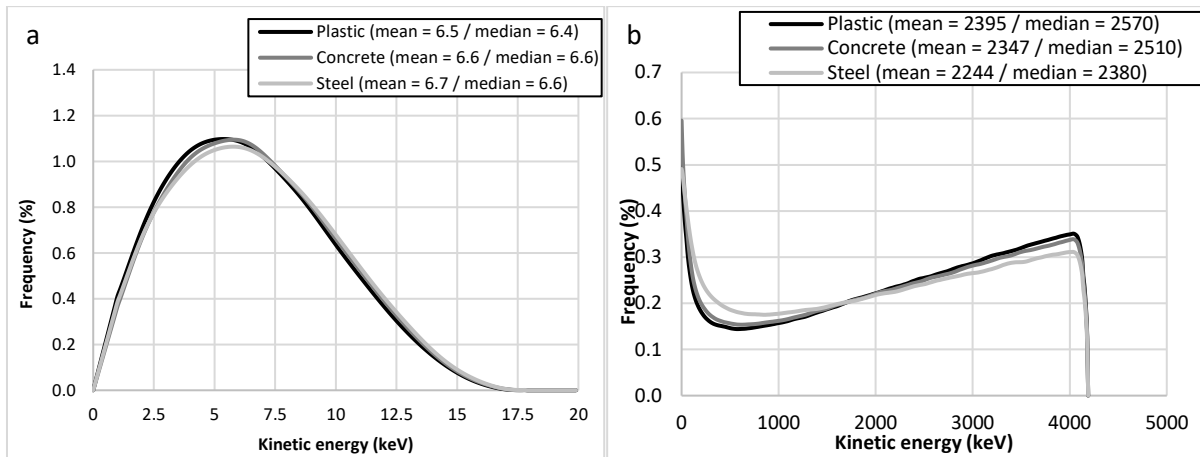
440

441 3.4. Matrix effect for the detection efficiency calculation

442 Figure 6 represents the frequency distributions (fd) of the kinetic energy for particles emitted
 443 from the upper face of a contaminated volume (i.e. the ASCV), in different matrix types. For
 444 alpha and beta, these fd are quite similar for a given radionuclide emission passing through the
 445 three different materials, regardless of the contamination thickness. Indeed, simulations were
 446 achieved considering two cases. In the first case, the thickness of the contaminated volume was
 447 set to R_{MAX} , which is variable from one material to another (Figure 6 and supplementary data
 448 Figure 1). In the second case, a constant thickness higher than R_{MAX} (30 μm , 100 μm and 1 mm
 449 for ^3H , ^{238}U and ^{14}C respectively) was used for each radionuclide, independently of the material
 450 (supplementary data Figure 2).

451 For more clarity, only one example of beta and alpha emitters is displayed in Figure 6.

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Fig. 6 Frequency distributions (fd) of the kinetic energy of particles when reaching the

Analyzed Surface of the Contaminated Volume (ASCV), supplied for three different

materials: plastic; concrete and steel. The mean and median values of the kinetic energy are

given for each fd. a- ^3H radionuclide (beta emission), contamination thickness = R_{MAX} . b- ^{238}U

radionuclide (alpha emissions only), contamination thickness = R_{MAX}

For beta emissions, a slight increase of mean and median energy values when material density increases (from plastic to steel) is observed, with a maximum relative standard deviation of 6 %. For alpha emissions, mean and median energy values decrease in denser materials, with a maximum relative deviation of 5 %. It can also be concluded that the detection efficiency of a given detector is the same for a given combination alpha/energy or beta/energy, regardless of the nature of the contaminated material. It means that the matrix effect on the detection efficiency of a given detector is very low or even negligible for the considered alpha and beta emissions.

4. Conclusion

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470

Autoradiography can be an efficient technique to measure traces of radioactivity on the surface of materials for biological and geological research fields but also more increasingly in the

471 framework of decommissioning. However, the calibration step is always an important issue, as
472 with all common Surface Contamination Meters (SCM).

473 The activity of standard specimens, needed to calibrate the autoradiography signal from films,
474 are most of the time provided in Bq g^{-1} , which is not consistent with the signal measured on
475 autoradiographs (DLU cm^{-2} for phosphor screens, grey level per pixel for silver halide
476 emulsions). The conversion tools proposed in the present work improve the calibration step for
477 these traditional methods of autoradiography.

478 Concerning radioimagers and SCMs, calibrated sources with certified surface emission rate are
479 often used to evaluate the detection efficiency needed to calibrate the instrument or to correct
480 the measurements. However, this kind of source are not easy to obtain (long national
481 procedures, usage regulations, transport...). Often it is necessary to use well characterized and
482 available laboratory samples, as is the case for the present study. Geant4 allowed to estimate
483 the Emission Fraction F_E , used to evaluate the Surface Activity A_S (or Surface Count Rate SCR ,
484 since $P_E = 1$) of the three laboratory samples chosen here, knowing their volumetric activities:
485 this illustrated the use of the conversion from Bq cm^{-3} to Bq cm^{-2} (Eq. (6)).

486 Results have demonstrated the importance of a good estimation of the emission fraction F_E to
487 apply the conversion method. Indeed, even if a contamination layer is extremely thin, F_E is
488 somewhat different from 0.5 (i.e. 50% of particles reaching the ASCV), as considered in the
489 ISO standard 7503-1 [24]. As an example, beta particles from ^3H have a short range in plastic:
490 for a contamination thickness equal to the relevant $R_{MAX} = 8.3 \mu\text{m}$, around 3 % of betas can
491 reach the ASCV (i.e. $F_E(R_{MAX} = 8.3 \mu\text{m}) = 0.03$). While the thickness of β -uranophane (the U-
492 bearing mineral including in the M8 sample) of $30 \mu\text{m}$ induces fraction emissions ranging from
493 0.14 to 0.32 according the alpha particle considered among the eight emitted in the ^{238}U decay
494 chain (i.e. $0.14 < F_E(30 \mu\text{m}) < 0.32$) (Table 5). The companion paper [25] supplies data of F_E
495 to deal with the cases where Contamination thickness $d_C < R_{MAX}$. $F_E(d)$ are also provided from

496 thickness $d = 0$ to $d = R_{MAX}$ of the considered particle type/energy and material. These $F_E(d)$ are
497 given for three materials (plastic, concrete and steel) and for ^3H , ^{14}C and ^{238}U , as well as other
498 radionuclides that can be encountered in nuclear dismantling facilities.

499 It was subsequently possible to calculate the detection efficiency of a radioimager, BeaQuant™
500 (GS $12 \times 8 \text{ cm}^2$ sample holder), for the three laboratory samples used as calibrated source in
501 this work. Since it has been shown that for a given instrument, the detection efficiency of both
502 beta/energy or alpha/energy combinations is independent of the composition of the material
503 contaminated (i.e. no matrix effect), the detection efficiency found for the three samples can be
504 generalized to the concerned radionuclides: $\varepsilon = 25\%$ for ^3H , $\varepsilon = 15\%$ for ^{14}C , $\varepsilon = 51\%$ for alpha
505 emitters in ^{238}U decay chain. These efficiency values ε can be used to calibrate or correct
506 BeaQuant™ measurements on materials contaminated with ^3H , ^{14}C or ^{238}U (Eq. (3)), performed
507 in the same configuration as used in the present study. Indeed, the detection efficiency of
508 BeaQuant™ is strongly dependent of the holder type and of the electric fields applied in the
509 detector (defined in the acquisition settings) (manufacturer's data). As an example, [9] found a
510 detection efficiency greater than 80% for alpha emitters in ^{238}U decay chain, using another
511 sample holder.

512 After detection efficiency correction (Eq. (3)), F_E parameter can be used to convert a measured
513 surface activity into volumetric activity (Eq. (6)). Such conversion remains nowadays a
514 challenge in decommissioning investigations where activity per gram is systematically required
515 to suitably manage the nuclear wastes. That is why we propose in the companion paper, a
516 second part "From Bq cm^{-3} to Bq cm^{-2} (and conversely) - Part 2: a useful conversion for
517 decommissioning operations", to provide a large set of data helping to deal with the conversion
518 problems possibly encountered in a dismantling context.

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526

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